

Cryogenic shock exfoliation for ultrahigh mobility rhombohedral graphite nanoelectronics

Corresponding Author: Professor Andrea Young

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Attachments originally included by the reviewers as part of their assessment can be found at the end of this file.

Version 0:

Reviewer comments:

Reviewer #1

(Comments for the Author)

1. Summary of the key results

In this manuscript, Holleis et al. present a new fabrication method to increase the yield and size of rhombohedral multilayer graphite (RMG), a metastable graphite phase with a specific stacking sequence. RMG has gained significant attention within the scientific community due to its unique band structure, which serves as a template for investigating various strongly correlated electronic effects. However, as the authors highlight, research progress has been hindered by the inherently low abundance of RMG in (natural) graphite. Here, the authors present a novel method for stabilizing RMG, fabricate various test devices from it, and apply a range of characterization methods.

2. Originality and significance: if not novel, please include reference

The present work demonstrates significant progress in the fabrication of RMG with respect to size and yield. Motivated by previous theoretical studies, the central approach involves inducing shear stresses to increase the intrinsic proportion of RMG relative to the more stable Bernal stacking of graphite. This is achieved by imposing thermal stresses onto a heterostructure consisting of graphite, a polymer tape, and an SiO₂/Si substrate. Following moderate heating, the heterostructure is then quickly immersed in liquid nitrogen (LN₂). The temperature change introduces thermal stresses, while the cryo-shock may contribute to stabilizing the fragile, metastable RMG phase. Consequently, the authors demonstrate that the percentage of RMG increases compared to the Bernal configuration, and the average area of the RMG regions expands significantly. While the approach of mechanically induced stress and rapid thermal quenching has a long history in materials science, this method is now being applied for the first time to stabilize metastable phases of 2D materials. The cryo-shock approach is a very simple technique, but much like the original "scotch tape method," it is readily foreseeable that the cryo-shock exfoliation will be rapidly adopted by the scientific community.

3. Data & methodology: validity of approach, quality of data, quality of presentation

To preserve the metastable material, a modified "force-free" stacking method is described, completing a comprehensive state-of-the-art process protocol. The authors employ a microscopic, freestanding polymer film for the (de)lamination of the flakes. One could mention that others have also used flexible membranes for 2D-transfer, [for example Wang, Wendong, et al. "Clean assembly of van der Waals heterostructures using silicon nitride membranes." *Nature Electronics* 6.12 (2023): 981-990] and even exploited soap bubbles as the ultimate soft film [Che, Lixuan, et al. "Soap film transfer printing for ultrathin electronics." *Small* 20.15 (2024): 2308312].

The authors present a wealth of results to demonstrate the exceptional quality of their RMG. These include far-field imaging (optical, IR), scanning microscopic techniques (AFM, scanning microwave impedance microscopy, scanning SQUID-on-tip microscopy), quantum capacitance, as well as electronic transport in the ballistic and hydrodynamic regimes. All these results are consistent with materials and devices of the highest quality; they achieve record-breaking mean free paths and offer a first glimpse into intriguing observations (see, for example, Ref. 25 from the same group, currently available only on arXiv) that we will most certainly see discussed in greater depth in the future. At the same time, this work will inspire others

to further develop this novel approach in new directions. The future will reveal its potential for scalability and application to other 2D material systems.

4. Appropriate use of statistics and treatment of uncertainties

The authors assessed a total of several hundred RMG flakes produced with "traditional" and "cryo-shock" methods to establish a clear advantage for the cryo-shock method. All characterization data is treated according to high standards.

5. Conclusions: robustness, validity, reliability

See also 3. & 4: The authors present a wealth of results to demonstrate the exceptional quality of their RMG. These include far-field imaging (optical, IR), scanning microscopic techniques (AFM, scanning microwave impedance microscopy, scanning SQUID-on-tip microscopy), quantum capacitance, as well as electronic transport in the ballistic and hydrodynamic regimes. All these results are consistent with materials and devices of the highest quality; they achieve record-breaking mean free paths and offer a first glimpse into intriguing observations (see, for example, Ref. 25 from the same group, currently available only on arXiv) that we will most certainly see discussed in greater depth in the future. At the same time, this work will inspire others to further develop this novel approach in new directions. The future will reveal its potential for scalability and application to other 2D material systems.

6. Suggested improvements: experiments, data for possible revision

None

7. References: appropriate credit to previous work?

Yes

8. Clarity and context: lucidity of abstract/summary, appropriateness of abstract, introduction and conclusions

The manuscript is clearly written, the figures are of high quality, and the scientific results are excellent. The presented method holds great potential for broad application in the field of 2D vdW materials. I strongly recommend this manuscript for publication in nature nanotechnology.

(Remarks on code availability)

Reviewer #2

(Comments for the Author)

The manuscript by Holleis et al. reports on a novel exfoliation method that not only enhances the fraction of rhombohedral multilayer graphene (RMG) flakes but it also leads to a higher exfoliation yield overall. Both factors clearly lead to larger areas of RMG flakes. The method is based on shear stress generated by differential thermal contraction of polymers, tapes, graphite and silicon substrate upon rapid cooling in liquid nitrogen. After exfoliation, microscopy in the infrared regime as well as scanning microwave impedance microscopy are used to identify the stacking order and hence to evaluate the yield of the exfoliation procedure. The vdW assemblies based on these RMG flakes show high quality as evidenced by multiple characterization techniques (quantum transport, capacitance and magnetization measurements). The authors demonstrate ballistic transport limited by the device size (10 μm x 10 μm) and transport in the hydrodynamic regime, both underpinning the high electronic quality of the device.

The claim of the authors on the improved exfoliation is clearly supported by the data with a sufficiently large number of samples/devices. The quality of the data is high and its presentation is adequate. The fact that the results of the paper is built on the usage of this technique by multiple people shows its reproducibility and robustness – hence allowing other research groups to easily reproduce the technique.

The main finding of the manuscript is well captured in figure 1 g, where the higher yield of RMG flakes is clearly shown. The fraction of rhombohedrally stacked graphite flakes increases by roughly a factor 3 from 10-15% using conventional exfoliation techniques to ~32% using the novel shock exfoliation technique. A similar increase in flake size is observed at the same time. The authors use then a low-pressure transfer technique, which they claim is essential, to reach a high device yield in the end (e.g. to preserve the metastable rhombohedral stacking during the vdW assembly process).

While the increased yield, in area and flake size, of RMG is a clear improvement over conventional exfoliation and device fabrication, the authors do not detail the low-pressure transfer technique even though they claim it is essential to reach a final device yield of 90%, as shown in Fig. S4. The manuscript clearly lacks clarity and transparency regarding the overall vdW assembly process.

It is not clear what steps are used by the authors to achieve this large overall device yield (e.g. last paragraph of the cryogenic shock exfoliation section):

- Are microdomains of Bernal-stacking excluded
- Are cutting techniques such as AFM cutting or etching used to isolated pure rhombohedral stacking prior to vdW-

assembly?

The manuscript would clearly improve in devoting more space to the details of the vdW-assembly technique used, as it is essential for the main claim of the authors. Has the low-pressure assembly technique been developed by the authors or is this commonly known in the community? If so, are there references to cite?

In addition, the authors lack to compare to other exfoliation techniques that claim to increase the RMG yield, see for example Ref: <https://arxiv.org/pdf/2605.22282>. Could they comment on what makes their method more suitable?

The manuscript would substantially improve in quality and impact if the authors could motivate why the different transport regimes are so exciting in RMG devices. What is new compared to Bernal stacking for RMG devices in the hydrodynamic regime for example? Are there changes expected to the ballistic regime? How is it different from Bernal stacked graphite flakes?

As high-quality transport in graphene-based vdW devices is well-established (e.g. ballistic transport, hydrodynamic regime, ...), the main novelty of the manuscript lies in the novel exfoliation and vdW-assembly technique. This is surely worth publishing, however, there remains the question whether Nature Nanotechnology is the appropriate choice.

There are also the following questions / comments that should be addressed:

- Why is the rhombohedral stacking limited to ~30% of the surface? Do the authors envision protocols to further enhance that fraction? What is the limiting mechanism?
- Is it so surprising to find a ballistic transport regime in a 13-layer graphite flake in the high-density regime?
- How are the authors sure that the RMG flakes do not contain some polytypes that are not purely rhombohedral stacking (e.g. A-B-C-A-B-A-B-C compared to A-B-C-A-B-C-A-B)? How would it change the picture?
- Is there a reason why the graphite flake for the top gate is put down in the last step after vdW-assembly? Does the AFM cleaning, prior to the deposition of the top gate change the stacking order?

(Remarks on code availability)

Reviewer #3

(Comments for the Author)

In this manuscript, the authors introduce a novel exfoliation technique that demonstrates higher yield of large-area rhombohedral multilayer graphene (RMG) with respect to conventional approaches. The technique relies on rapid cooling in liquid nitrogen, resulting in shear stress on graphite flakes, which drives a conversion from Bernal to rhombohedral stacking. The authors further refine the van der Waals assembly sequence as to preserve the rhombohedral regions and succeed in fabricating high-mobility field effect devices. The electronic quality of the resulting samples is assessed via transport and scanning probe experiments, complemented with numerical simulations, which indicate large electronic mean free paths (> 200 μm) and gate-tunable transport regimes.

The cryogenic shock technique is surely relevant for a broad community and its introduction timely. The data presented are of excellent quality and convincingly support the main conclusions of the paper. However, several aspects of the manuscript need to be improved before considering it for publication in Nature Nanotechnology.

Major comments:

- The properties of RMG strongly depend on the layer number. However, this is not adequately considered in the paper. The authors should extend the statistics of Fig.1g,h also to the thickness of the obtained flakes and RMG domains therein. Additionally, the RMG thickness of flakes and devices shown throughout the paper should be indicated.
- The community is reporting exciting new physics in the thin limit of RMG (e.g. 4-layers show chiral superconductivity [15] and coexisting superconductivity-QAH [14]). While the realization of thick (13-layer) RMG devices is convincingly discussed, there is no evidence that the thin limit can be explored with this new technique, highlighting a possible key drawback. The authors should report on whether thin RMG devices can be obtained via cryogenic shock exfoliation and, if possible, discuss their capacity to support correlated electronic phases.
- The authors claim to combine "all known techniques to reduce relaxation". However, they do not consider/mention cutting the crystals to isolate the RMG domains (which was done in previous works from the same group, e.g. Ref.[2]). Are the crystals cut before stacking? Or is it no longer required with the new technique?
- Regarding the narrow channel device, the paper reports a simulation of the spatial current flow (Fig.4h), but no experimental Biot-Savart imaging. The authors should consider reporting these data, or explain why they are not present.

Minor comments:

- Promotion of rhombohedral stacking from mechanical stress was already shown in CVD-grown graphene [10.1038/s41467-019-14022-3, 10.1016/j.carbon.2021.02.082]. While the domains obtained by CVD are rather small in lateral dimension, the fraction of RMG over Bernal and the amount of material from a single growth cycle can be considerable. This should be mentioned when commenting on the relative scarcity of RMG from previous techniques.
- A device with area >1300 μm^2 is claimed: is an optical image of this specific device shown in the paper?
- Which graphite crystals are used as starting material? Which tape is employed? These details could be useful for other

groups to adopt this new technique, increasing the potential impact of the paper.

- The “single sequence transfer” mentioned in Fig. S4b (x axis) is not properly defined in the paper or methods.

(Remarks on code availability)

Version 1:

Reviewer comments:

Reviewer #2

(Comments for the Author)

The authors have clarified all points raised by us and by the other two referees adequately. The updated manuscript clearly improved in quality and clarity and allows now for easier reproduction by other research groups. We can recommend publication in Nature Nanotechnology after addressing a minor point.

Minor comment:

- The newly added Ref. 47, concerning membrane transfer techniques, seems to be unrelated.

(Remarks on code availability)

Reviewer #3

(Comments for the Author)

The authors satisfactorily answered all of the remarks on the original manuscript, appreciably providing new data. Thus, I support the publication of the paper in its current form in nature nanotechnology.

(Remarks on code availability)

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Response to Reviewers

“Cryogenic shock exfoliation for ultrahigh mobility
rhombohedral graphite nanoelectronics”

Ludwig Holleis et al.

Submitted to Nature Nanotechnology

June 19, 2026

We thank all three reviewers for their careful reading of our manuscript and their constructive feedback. All three reviewers are positive about the work; Reviewers 1 and 3 recommend publication, while Reviewer 2 raises several points which we address below.

Reviewer 1

Summary: Reviewer 1 strongly recommends publication, describing the cryo-shock method as simple yet transformative. We thank the reviewer for their positive evaluation of our work and recommendation for publication.

1. Summary of the key results

In this manuscript, Holleis et al. present a new fabrication method to increase the yield and size of rhombohedral multilayer graphite (RMG), a metastable graphite phase with a specific stacking sequence. RMG has gained significant attention within the scientific community due to its unique band structure, which serves as a template for investigating various strongly correlated electronic effects. However, as the authors highlight, research progress has been hindered by the inherently low abundance of RMG in (natural) graphite. Here, the authors present a novel method for stabilizing RMG, fabricate various test devices from it, and apply a range of characterization methods.

2. Originality and significance: if not novel, please include reference

The present work demonstrates significant progress in the fabrication of RMG with respect to size and yield. Motivated by previous theoretical studies, the central approach involves inducing shear stresses to increase the intrinsic proportion of RMG relative to the more stable Bernal stacking of graphite. This is achieved by imposing thermal stresses onto a heterostructure consisting of graphite, a polymer tape, and an SiO/Si substrate. Following moderate heating, the heterostructure is then quickly immersed in liquid nitrogen (LN₂). The temperature change introduces thermal stresses, while the cryo-shock may contribute to stabilizing the fragile, metastable RMG phase. Consequently, the authors demonstrate that the percentage of RMG increases compared to the Bernal configuration, and the average area of the RMG regions expands significantly. While the approach of mechanically induced stress and rapid thermal quenching has a long history in materials science, this method is now being applied for the first time to stabilize metastable phases of 2D materials. The cryo-shock approach is a very simple technique, but much like the original “scotch tape method”, it is readily foreseeable that the cryo-shock exfoliation will be rapidly adopted by

the scientific community.

3. Data and methodology: validity of approach, quality of data, quality of presentation

To preserve the metastable material, a modified force-free stacking method is described, completing a comprehensive state-of-the-art process protocol. The authors employ a microscopic, freestanding polymer film for the (de)lamination of the flakes. One could mention that others have also used flexible membranes for 2D-transfer, (for example Wang, Wendong, et al., Clean assembly of van der Waals heterostructures using silicon nitride membranes. *Nature Electronics* 6.12 (2023): 981-990) and even exploited soap bubbles as the ultimate soft film (Che, Lixuan, et al., Soap film transfer printing for ultrathin electronics. *Small* 20.15 (2024): 2308312).

We thank the reviewer for bringing these references to our attention and have added both in the appropriate places.

The authors present a wealth of results to demonstrate the exceptional quality of their RMG. These include far-field imaging (optical, IR), scanning microscopic techniques (AFM, scanning microwave impedance microscopy, scanning SQUID-on-tip microscopy), quantum capacitance, as well as electronic transport in the ballistic and hydrodynamic regimes. All these results are consistent with materials and devices of the highest quality; they achieve record-breaking mean free paths and offer a first glimpse into intriguing observations (see, for example, Ref. 25 from the same group, currently available only on arXiv) that we will most certainly see discussed in greater depth in the future. At the same time, this work will inspire others to further develop this novel approach in new directions. The future will reveal its potential for scalability and application to other 2D material systems.

4. Appropriate use of statistics and treatment of uncertainties

The authors assessed a total of several hundred RMG flakes produced with "traditional" and "cryo-shock" methods to establish a clear advantage for the cryo-shock method. All characterization data is treated according to high standards.

5. Conclusions: robustness, validity, reliability

See also 3. 4: The authors present a wealth of results to demonstrate the exceptional quality of their RMG. These include far-field imaging (optical, IR), scanning microscopic techniques (AFM, scanning microwave impedance microscopy, scanning SQUID-on-tip microscopy), quantum capacitance, as well as electronic transport in the ballistic and hydrodynamic regimes. All these results are consistent with materials and devices of the highest quality; they achieve record-breaking mean free paths and offer a first glimpse into intriguing observations (see, for example, Ref. 25 from the same group, currently available only on arXiv) that we will most certainly see discussed in greater depth in the future. At the same time, this work will inspire others to further develop this novel approach in new directions. The future will reveal its potential for scalability and application to other 2D material systems.

6. Suggested improvements: experiments, data for possible revision

None

7. References: appropriate credit to previous work?

Yes

8. Clarity and context: lucidity of abstract/summary, appropriateness of abstract, introduction and conclusions

The manuscript is clearly written, the figures are of high quality, and the scientific results are excellent. The presented method holds great potential for broad application in the field of 2D vdW materials. I strongly recommend this manuscript for publication in nature nanotechnology.

Reviewer 2

Summary: Reviewer 2 is broadly positive about our work and we would like to thank them for their assessment. They raise several important points, all of which we address below. We hope that, with these improvements, the reviewer will now support publication of our work in Nature Nanotechnology.

Reviewer 2 (Comments for the Author):

The manuscript by Holleis et al. reports on a novel exfoliation method that not only enhances the fraction of rhombohedral multilayer graphene (RMG) flakes but it also leads to a higher exfoliation yield overall. Both factors clearly lead to larger areas of RMG flakes. The method is based on shear stress generated by differential thermal contraction of polymers, tapes, graphite and silicon substrate upon rapid cooling in liquid nitrogen. After exfoliation, microscopy in the infrared regime as well as scanning microwave impedance microscopy are used to identify the stacking order and hence to evaluate the yield of the exfoliation procedure. The vdW assemblies based on these RMG flakes show high quality as evidences by multiple characterization techniques (quantum transport, capacitance and magnetization measurements). The authors demonstrate ballistic transport limited by the device size (10 μm x 10 μm) and transport in the hydrodynamic regime, both underpinning the high electronic quality of the device. The claim of the authors on the improved exfoliation is clearly supported by the data with a sufficiently large number of samples/devices. The quality of the data is high and its presentation is adequate. The fact that the results of the paper is built on the usage of this technique by multiple people shows its reproducibility and robustness – hence allowing other research groups to easily reproduce the technique. The main finding of the manuscript is well captured in figure 1 g, where the higher yield of RMG flakes is clearly shown. The fraction of rhombohedrally stacked graphite flakes increases by roughly a factor 3 from 10-15% using conventional exfoliation techniques to 32% using the novel shock exfoliation technique. A similar increase in flake size is observed at the same time. The authors use then a low-pressure

transfer technique, which they claim is essential, to reach a high device yield in the end (e.g. to preserve the metastable rhombohedral stacking during the vdW assembly process).

We thank the reviewer for their positive assessment of our work.

While the increased yield, in area and flake size, of RMG is a clear improvement over conventional exfoliation and device fabrication, the authors do not detail the low-pressure transfer technique even though they claim it is essential to reach a final device yield of 90%, as shown in Fig. S4. The manuscript clearly lacks clarity and transparency regarding the overall vdW assembly process. It is not clear what steps are used by the authors to achieve this large overall device yield (e.g. last paragraph of the cryogenic shock exfoliation section):

- Are microdomains of Bernal-stacking excluded
- Are cutting techniques such as AFM cutting or etching used to isolated pure rhombohedral stacking prior to vdW-assembly?

The manuscript would clearly improve in devoting more space to the details of the vdW-assembly technique used, as it is essential for the main claim of the authors.

We thank the reviewer for requesting clarification on these details as we want to be as specific as possible to allow for easy adaptation of our techniques by the community. We have added the key steps that we used to achieve 90% yield like isolating rhombohedral domains via AFM cutting in the main text and significantly expanded the method section on the vdW-assembly process.

Has the low-pressure assembly technique been developed by the authors or is this commonly known in the community? If so, are there references to cite?

To the best of our knowledge, we are unaware of any literature that employs this version of the membrane transfer. As reviewer 1 pointed out, there are other membrane transfer techniques which we now cite. We want to note, however, that none of these have been successfully used to transfer rhombohedral multilayer graphene, based on what is available in the literature.

In addition, the authors lack to compare to other exfoliation techniques that claim to increase the RMG yield, see for example Ref: <https://arXiv.org/pdf/2605.22282>. Could they comment on what makes their method more suitable?

We indeed have noticed this publication; however, want to note that it was submitted to arXiv about a month after our initial submission and hence have not discussed it. We have tested this different bending method described in the reference, but were not able to reproduce the result or notice any difference in rhombohedral yield. In general, it is not our task to review this paper, and since it has been uploaded to a preprint server significantly after our work, we will leave it to the peer review process and the community to evaluate this arXiv article. We want to stress that we do contextualize our work within the peer-reviewed literature of manipulating multilayer graphene stacking orders in favor of rhombohedral structure (citations 33-39 in the revised manuscript).

The manuscript would substantially improve in quality and impact if the authors could motivate why the different transport regimes are so exciting in RMG devices. What is new compared to

Bernal stacking for RMG devices in the hydrodynamic regime for example? Are there changes expected to the ballistic regime? How is it different from Bernal stacked graphite flakes?

We thank the reviewer for pointing out the lack of motivation of the current flow section and have added clarifying text in the appropriate positions for improved flow. We have included this section for two reasons: First, ballistic electron flow is just a further illustration of the high sample quality, and tuning to the hydrodynamic regime is also only possible when the momentum relaxation length (or mean-free-path) is large. Second, we were able to observe the first device size driven crossover from Poiseuille to porous flow. Porous flow has been previously only reported in a Corbino geometry due to the absence of boundaries by default. Hence, we report a new physical regime that is only achievable in samples of the size that are produced by cryogenic shock exfoliation.

Ballistic transport is also present in Bernal stacked samples, and indeed we do not expect rhombohedral and Bernal samples to behave differently in this regime. However, no electron hydrodynamics has been observed in Bernal stacked samples as electron-electron interactions are significantly weaker, and the flat bands of rhombohedral graphene are essential to access this regime. While it may be in principle possible to observe similar regime in Bernal *bilayer* graphene—equivalent to rhombohedral bilayer graphene!—nobody seems to have explored this possibility to date. We do expect that future work may indeed reveal the universality of this physics following our work here.

As high-quality transport in graphene-based vdW devices is well-established (e.g. ballistic transport, hydrodynamic regime, ...), the main novelty of the manuscript lies in the novel exfoliation and vdW-assembly technique. This is surely worth publishing, however, there remains the question whether Nature Nanotechnology is the appropriate choice.

We respectfully disagree with the reviewer’s assessment of the impact of our novel the exfoliation and assembly technique. Our work simultaneously addresses multiple long-standing challenges of device size and yield. Taken together, and viewed in the broader context of the 2D materials community, this is — to our knowledge — the first time a **strongly correlated, metastable** material, whether rhombohedral or moiré, has been reliably fabricated into large devices suitable for genuine mesoscopic physics experiments. We deliberately reserved this point for the conclusion, and we hope its significance becomes apparent to any 2D experimentalist from the broad body of results preceding it. The reviewer points to high quality signatures of effectively uncorrelated electrons; our manuscript differs in that the *the samples* also show, e.g., isospin ferromagnetism, and superconductivity as can be seen in the data presented in the main text and on a rhombohedral hexalayer device added in the Extended Data (though we have not explored these phases in detail in the current manuscript, which is focused on fabrication).

There are also the following questions / comments that should be addressed:

- Why is the rhombohedral stacking limited to 30% of the surface? Do the authors envision protocols to further enhance that fraction? What is the limiting mechanism?

That is an excellent question, and we do hope to improve on this number further, testing modifications to the cryogenic shock exfoliation technique. To date, the method described in the article is our most successful approach.

- Is it so surprising to find a ballistic transport regime in a 13-layer graphite flake in the high-density regime?

It is indeed not surprising to find a ballistic transport regime at high densities. The large mean-free-path exceeding $200\mu\text{m}$ is quantitatively surprising as this has not been measured at low temperatures, especially in rhombohedral devices; it likely indicates the absence of, e.g., interlayer stacking faults.

- How are the authors sure that the RMG flakes do not contain some polytypes that are not purely rhombohedral stacking (e.g. A-B-C-A-B-A-B-C compared to A-B-C-A-B-C-A-B)? How would it change the picture?

That is an excellent question as the IR and sMIM images only give relative and not absolute contrast. Initially, we confirmed rhombohedral stacking via Raman maps. More recently, we have skipped this step as all devices fabricated from uniform (seemingly) RMG domains produced by cryogenic shock exfoliation flakes end up in the rhombohedral configuration. This might have two reasons: first, it is energetically unlikely to get such large domains of the intermediate polytypes. Second, the intermediate domains might relax to the rhombohedral configuration during the transfer process. We are certain, however, that the final devices only contain rhombohedral stacking from both the local scanning nSQUID-on-tip images and the global capacitance measurements that present an area average, and only show rhombohedral features.

- Is there a reason why the graphite flake for the top gate is put down in the last step after vdW-assembly? Does the AFM cleaning, prior to the deposition of the top gate change the stacking order?

The top graphite gate prevents us from confirming the stacking order with the IR camera during and after transfer which is why we drop it after finishing the stack. AFM cleaning does not change the stacking order. In fact, we almost never see the RMG relax to Bernal after its initial pickup from the SiO_2 substrate. We have added our logic for AFM cleaning and transferring the top gate as a final step in the methods section.

Reviewer 3

Summary: Reviewer 3 finds the data excellent and the technique timely and relevant, and recommends publication after addressing a few major and minor points concerning layer-number statistics, evidence for thin-limit RMG, crystal cutting and Biot-Savart imaging. We thank the reviewer for their positive assessment and support for publication, and address their concerns below.

In this manuscript, the authors introduce a novel exfoliation technique that demonstrates higher yield of large-area rhombohedral multilayer graphene (RMG) with respect to conventional approaches. The technique relies on rapid cooling in liquid nitrogen, resulting in shear stress on graphite flakes, which drives a conversion from Bernal to rhombohedral stacking. The authors further refine the van der Waals assembly sequence as to preserve the rhombohedral regions and succeed in fabricating high-mobility field effect devices. The electronic quality of the resulting samples is assessed via transport and scanning probe experiments, complemented with numerical

simulations, which indicate large electronic mean free paths (> 200 nm) and gate-tunable transport regimes.

The cryogenic shock technique is surely relevant for a broad community and its introduction timely. The data presented are of excellent quality and convincingly support the main conclusions of the paper. However, several aspects of the manuscript need to be improved before considering it for publication in Nature Nanotechnology.

Major comments:

- The properties of RMG strongly depend on the layer number. However, this is not adequately considered in the paper. The authors should extend the statistics of Fig.1g,h also to the thickness of the obtained flakes and RMG domains therein. Additionally, the RMG thickness of flakes and devices shown throughout the paper should be indicated.

We thank the reviewer for raising this point. We have included a breakdown of the exfoliation statistics by layer number in extended data figure S2 which shows a relatively even distribution of uniform RMG areas across layer number. The thicker multilayer graphene flakes tend to be a bit larger and have fewer cracks or wrinkles, and warming the flakes slowly in an inert atmosphere helps achieve a bit more uniform thinner multilayer graphene flakes, although not strictly necessary. We also now consistently label the layer number throughout the manuscript.

- The community is reporting exciting new physics in the thin limit of RMG (e.g. 4-layers show chiral superconductivity (15) and coexisting superconductivity-QAH (14)). While the realization of thick (13-layer) RMG devices is convincingly discussed, there is no evidence that the thin limit can be explored with this new technique, highlighting a possible key drawback. The authors should report on whether thin RMG devices can be obtained via cryogenic shock exfoliation and, if possible, discuss their capacity to support correlated electronic phases.

Indeed, this is an important point. Cryogenic shock exfoliation produces high quality and uniform rhombohedral domains independently of layer number (as shown in the extended data figure S1). These show no difference from flakes produced by the standard method, and devices are generally of the same quality as reported in the literature. To this end, transport and capacitance data from a device fabricated with cryogenic shock exfoliation is shown in the recent arXiv preprint from our group arXiv:2605.30316. We also included another supplementary figure of a six layer sample aligned to hBN that shows both (in-plane field induced) superconductivity as well as topological phases at the same quality as reported by other and our group previously.

- The authors claim to combine “all known techniques to reduce relaxation”. However, they do not consider/mention cutting the crystals to isolate the RMG domains (which was done in previous works from the same group, e.g. Ref.(2)). Are the crystals cut before stacking? Or is it no longer required with the new technique?

We apologize for the confusion. We image the flakes with sMIM to confirm the absence of microdomains and domain walls and then isolate the uniform domains as the reviewer described. We have clarified this point in the methods and extended data figure.

- Regarding the narrow channel device, the paper reports a simulation of the spatial current flow

(Fig.4h), but no experimental Biot-Savart imaging. The authors should consider reporting these data, or explain why they are not present.

The reviewer makes a valuable suggestion, and we have included measurements we have taken for the narrow channel. The data is analogous to the imaging across a narrow channel reported by our group previously in this preprint arXiv:2603.11175. We find that the current concentration agrees with our assignment of hydrodynamic, diffusive, and ballistic current flow following the analysis of the above article.

Minor comments:

- Promotion of rhombohedral stacking from mechanical stress was already shown in CVD-grown graphene [10.1038/s41467-019-14022-3, 10.1016/j.carbon.2021.02.082]. While the domains obtained by CVD are rather small in lateral dimension, the fraction of RMG over Bernal and the amount of material from a single growth cycle can be considerable. This should be mentioned when commenting on the relative scarcity of RMG from previous techniques.

We thank the reviewer for bringing these references to our attention and we have included them in the list of previous techniques.

- A device with area $>1300 \text{ um}^2$ is claimed: is an optical image of this specific device shown in the paper?

This device is shown in extended data figure 5 as referenced in the main text.

- Which graphite crystals are used as starting material? Which tape is employed? These details could be useful for other groups to adopt this new technique, increasing the potential impact of the paper.

We thank the reviewer for encouraging us to add these details. Since submission, we have found that the source of graphite can influence the exfoliation result. We have added these details to the methods section and provided a supplementary video illustrating the cryogenic shock exfoliation process.

- The “single sequence transfer” mentioned in Fig. S4b (x axis) is not properly defined in the paper or methods.

We have added more explanation clarifying this detail.

Review "Cryogenic shock exfoliation for ultrahigh mobility rhombohedral graphite nanoelectronics"
by Ludwig Holleis et. al.

In this manuscript, Holleis et al. present a new fabrication method to increase the yield and size of rhombohedral multilayer graphite (RMG), a metastable graphite phase with a specific stacking sequence. RMG has gained significant attention within the scientific community due to its unique band structure, which serves as a template for investigating various strongly correlated electronic effects. However, as the authors highlight, research progress has been hindered by the inherently low abundance of RMG in (natural) graphite. Here, the authors present a novel method for stabilizing RMG, fabricate various test devices from it, and apply a range of characterization methods.

The present work demonstrates significant progress in the fabrication of RMG with respect to size and yield. Motivated by previous theoretical studies, the central approach involves inducing shear stresses to increase the intrinsic proportion of RMG relative to the more stable Bernal stacking of graphite. This is achieved by imposing thermal stresses onto a heterostructure consisting of graphite, a polymer tape, and an SiO₂/Si substrate. Following moderate heating, the heterostructure is then quickly immersed in liquid nitrogen (LN₂). The temperature change introduces thermal stresses, while the cryo-shock may contribute to stabilizing the fragile, metastable RMG phase. Consequently, the authors demonstrate that the percentage of RMG increases compared to the Bernal configuration, and the average area of the RMG regions expands significantly. While the approach of mechanically induced stress and rapid thermal quenching has a long history in materials science, this method is now being applied for the first time to stabilize metastable phases of 2D materials. The cryo-shock approach is a very simple technique, but much like the original "scotch tape method," it is readily foreseeable that the cryo-shock exfoliation will be rapidly adopted by the scientific community.

To preserve the metastable material, a modified "force-free" stacking method is described, completing a comprehensive state-of-the-art process protocol. The authors employ a microscopic, freestanding polymer film for the (de)lamination of the flakes. One could mention that others have also used flexible membranes for 2D-transfer, [for example Wang, Wendong, et al. "Clean assembly of van der Waals heterostructures using silicon nitride membranes." *Nature Electronics* 6.12 (2023): 981-990] and even exploited soap bubbles as the ultimate soft film [Che, Lixuan, et al. "Soap film transfer printing for ultrathin electronics." *Small* 20.15 (2024): 2308312].

The authors present a wealth of results to demonstrate the exceptional quality of their RMG. These include far-field imaging (optical, IR), scanning microscopic techniques (AFM, scanning microwave impedance microscopy, scanning SQUID-on-tip microscopy), quantum capacitance, as well as electronic transport in the ballistic and hydrodynamic regimes. All these results are consistent with materials and devices of the highest quality; they achieve record-breaking mean free paths and offer a first glimpse into intriguing observations (see, for example, Ref. 25 from the same group, currently available only on arXiv) that we will most certainly see discussed in greater depth in the future. At the same time, this work will inspire others to further develop this novel approach in new directions. The future will reveal its potential for scalability and application to other 2D material systems.

The manuscript is clearly written, the figures are of high quality, and the scientific results are excellent. The presented method holds great potential for broad application in the field of 2D vdW materials.

I strongly recommend this manuscript for publication in nature nanotechnology.