

Editorial comments:

We thank the editors for carefully reading our manuscript and providing valuable feedback for improving the presentation of our work. Please find below a point-by-point response to the comments.

- 2. Please revise the Protocol text to avoid the use of any personal pronouns*
- 3. Please revise the Protocol to contain only action items that direct the reader to do something*
- 4. Lines 81-108: The Protocol should contain only action items that direct the reader to do something. Please either write the text in the imperative tense as if telling someone how to do the technique (e.g., “Do this,” “Ensure that,” etc.), or move the instrument information to the Table of Materials.*

We have revised each step of our protocol text to contain action items and to avoid personal pronouns.

- 5-13. Please add more details to your protocol steps.*

We have addressed all points 6-13 and added the necessary details.

- 14. Please combine some of the shorter Protocol steps so that individual steps contain 2-3 actions and maximum of 4 sentences per step.*

Where appropriate, we have combined our protocol steps, specifically in 3,4,5,6.

- 15-17 After you have made all the recommended changes to your protocol (listed above), please highlight 2.75 pages or less of the Protocol (including headings and spacing)*

We have highlighted the text necessary for the video.

- 18. Figure 1a: Please use lowercase x for magnification.*
- 19. Figure 3f and Figure 4e: Please include a space between the number and its temperature unit (75 °C, 100 °C).*

The figures have been updated to incorporate the comments.

- 20. Table of Materials: Please include a space between all numerical values and their corresponding units (100 µs, 11 nm, 15 mL, 37 °C, etc.). Please sort the items in alphabetical order according to the name of material/equipment.*

The Table of Materials has been updated to incorporate the comments.

- 21. Please include a Disclosures section, providing information regarding the authors' competing financial interests or other conflicts of interest. If authors have no competing financial interests, then a statement indicating no competing financial interests must be included.*

The Disclosures section and appropriate information was added.

Reviewers' comments:

Reviewer #1:

- *Minor Concerns:*

My only concern is that some of figures are confusing. For example, in figure 4d, the PDMS has some strange perspective (is it above or behind the glass slide?). In Figure 4 e,f,g, I presume the purple-colored block is the PDMS. But it is different from Figure 4d and it is not re-label after the color-change. etc. Figure 3 is also confusing. It helps to label the objects in the figures, so that they are self-explanatory.

We thank the reviewer for reading carefully and providing comments that helped us improved the presentation of our method. We have revised Figure 4 and Figure 3 by adding labels and removing some of the intermediary steps to avoid confusions.

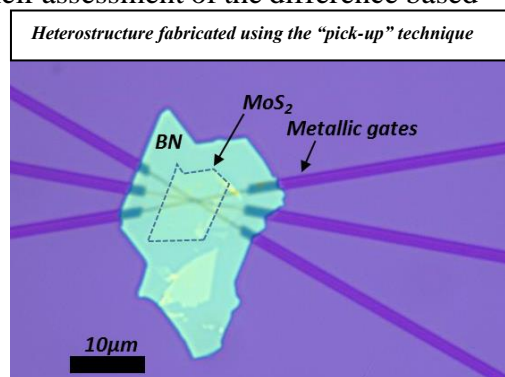
Reviewer #2:

- *In this manuscript, the authors presented their home-built computer-controlled transfer setup to stack 2D material flakes, as well as their complete dry transfer procedure. This setup can realize computer-controlled in-plane and angular alignment between flakes, which is good.*

We thank the reviewer for pointing out that the method and set-up presented in the manuscript are valuable.

- *The described technique itself is the last-generation dry transfer (layer-by-layer) technique, whereas the mainstream now is the pick-up technique which improves stack quality greatly. The major drawback of the layer-by-layer transfer technique is: by using this method, flakes can not avoid contacting polymers and acetone which greatly reduces device quality and can not be completely compensated by other cleaning efforts like annealing. (This is demonstrated by the huge bubbles shown in the AFM scans of Fig 7d. Note Fig 6d does not indicate any clean stack if the flakes are that thick.) As a result, no novelty exists in this manuscript, and the method is not good either.*

We thank the reviewer for carefully considering all the existing dry transfer methods and providing a comparison among them. We would like to clarify that in this manuscript we aim to emphasize the set-up for realizing heterostructures while also providing the reader with two distinct methods for preparing the top and bottom substrates. We agree with their assessment of the difference based on the “cleanliness” of the resulting heterostructure, however, optimizing the preparation methods for the goal of obtaining pristine surfaces is beyond the scope of our manuscript. To clarify that our method is fully compatible with the “pick-up technique”, we are presenting in the figure an optical micrograph of a heterostructure fabricated by means of the “pick-up technique” using the set-up described in this manuscript. We would also like to comment on the fact that for certain applications the “pick-up” technique might not be as practical as other two we are presenting, for example when a stack might not necessarily involve hBN.



- *The method no doubt works though, so there is no question about its validness, and this method still can be used in quality-insensitive studies. Given that the journal does not request novelty and what presented here can still be helpful to some researchers, I am not against publishing this manuscript...*

We thank the reviewer for recognizing the broader applicability of our presented method, it was indeed our goal to emphasise that point. Therefore, to clarify, we added a sentence in the discussion to stress that this method can be extended to other situations when two crystals or a crystal and another substrate need to be precisely aligned laterally and rotationally.

Reviewer #3:

- *Manuscript Summary:*
The manuscript JoVE59727 presents a thorough description of a motorized deterministic transfer setup used to fabricate van der Waals heterostructures with controlled lateral and rotational orientation. The topic is certainly interesting and it will be useful for the readership interested on 2D materials.

We thank the reviewer for carefully reading the manuscript and for their positive assessment of the utility of the method we present.

- *Major Concerns:*
The main novelty of the manuscript is the description of the setup. The authors claim that they present two transfer techniques but them have been thoroughly described in the literature already. The motorized transfer setup, although not described in full in the literature, has been already reported by other groups like Manchester and EPFL. Thus, as said the main novelty is the description of the setup. The Editorial team should judge if this is enough novelty to publish the manuscript. The authors claim that other transfer setups require of complex infrastructure but in reality this system is much more complex than those operated 'by hand' and the motorized ones are as complicated as this one.

We thank the reviewer for underscoring the goal of our manuscript, which is to present a method that is based on a set-up together with two examples of how to prepare the samples that will be stacked using the set-up. We agree with the reviewer that such a set-up has not been presented in full in literature, which prompted us to present it in this manuscript. Furthermore, the hope that the broader and interdisciplinary readership of JOVE might find this method - currently almost exclusively used for 2D materials - to have applicability in other areas.

- *Minor Concerns:*
In the first figure regarding the transfer of ReS₂. Why the authors choose 75°? There is no obvious reason. It seems as if they failed in getting 60° or 90° and afterwards they claim that they intended 75°.

We thank the reviewer for addressing this question. We would like to draw the reviewer's attention first to Figure 6 where we present a sequence of transfers from 15° to 90° in steps of 15°. Our goal was to present a general method, therefore we did not focus on the angles that have connection with the symmetries of the crystals (such as 30 ° or 60 °).

Figure 5 is, therefore, not a failed attempt, but simply one intermediate step in the sequence of Figure 6. We felt that it was important to choose one of the stacks in the sequence and thoroughly present the individual flakes before and after transfer as well as the characterization of the rotation angle using atomic force microscopy.