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TITLE:

Fabricating van der Waals Heterostructures with Precise Rotational Alignment

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SUMMARY:

In this work we describe a technique that is used to create new crystals (van der Waals heterostructures) by stacking ultrathin layered 2D materials with precise control over position and relative orientation.

ABSTRACT:

In this work we describe a technique for creating new crystals (van der Waals heterostructures) by stacking distinct ultrathin layered 2D materials. We demonstrate not only lateral control but, importantly, also control over the angular alignment of adjacent layers. The core of the technique is represented by a home-built transfer setup which allows the user to control the position of the individual crystals involved in the transfer. This is achieved with sub-micrometer (translational) and sub-degree (angular) precision. Prior to stacking them together, the isolated crystals are individually manipulated by custom-designed moving stages that are controlled by a programmed software interface. Moreover, since the entire transfer setup is computer controlled, the user can remotely create precise heterostructures without coming into direct contact with the transfer setup, labeling this technique as “hands-free”. In addition to presenting the transfer set-up, we also describe two techniques for preparing the crystals that are subsequently stacked.

INTRODUCTION:

Research in the burgeoning field of two-dimensional (2D) materials began after researchers developed a technique which enabled the isolation of graphene[1-3] (an atomically flat sheet of carbon atoms) from graphite. Graphene is a member of a larger class of layered 2D materials, also referred to as van der Waals materials or crystals. They have strong covalent intralayer bonding and weak van der Waals interlayer coupling. Therefore, the technique for isolating

graphene from graphite can also be applied to other 2D materials where one can break the weak interlayer bonds and isolate single layers. One key development in the field was the demonstration that just as the van der Waals bonds holding adjacent layers of two-dimensional materials together can be broken, they can also be put back together[2,4]. Therefore, crystals of 2D materials can be created by controllably stacking together layers of 2D materials with distinct properties. This spurred a great deal of interest, as materials previously inexistent in nature can be created with the goal of either uncovering formerly inaccessible physical phenomena[4-9] or developing superior devices for technology applications. Therefore, having precise control over stacking 2D materials has become one of the main goals in the research field[10-12].

In particular, the twist angle between adjacent layers in van der Waals heterostructures was shown to be an important parameter for controlling material properties[13]. For example, at some angles, the introduction of a relative twist between adjacent layers can effectively electronically decouple the two layers. This was studied both in graphene[14,15] as well as in transition metal dichalcogenides[16-19]. More recently, it was surprisingly found that it can also alter the state of matter of these materials. The discovery that bilayer graphene oriented at a "magic angle" behaves as a Mott insulator at low temperatures and even a superconductor when the electron density is properly tuned has sparked great interest and a realization of the importance of the angular control when fabricating layered van der Waals heterostructures[13,20,21].

Motivated by the scientific opportunities opened up by the idea of tuning the properties of novel van der Waals materials by adjusting the relative orientation between the layers, we present a home-built instrument along with the procedure to create such structures with angular control.

PROTOCOL:

1. Instrumentation for the transfer procedure

1.1. In order to visualize the transfer process, utilize an optical microscope that can operate under bright-field illumination. Since the typical sizes of the 2D crystals are 1–500 μm^2, equip the microscope with 5x, 50x, and 100x long working distance objectives. The microscope must also be equipped with a camera that connects to a computer (**Figure 1a**).

1.2. Use separate manipulators to individually control the position of the two crystals that are about to be stacked. Employ manipulators that are programmable and controllable by a computer to minimize vibrations during the transfer procedure.

NOTE: The manipulators responsible for the movement of the top substrate holder (**Figure 1b-c**) need only move in the X, Y, and Z direction. Importantly, the manipulators responsible for controlling the bottom substrate holder (**Figure 1e-f**) are also able to rotate by any angle Θ (translational and rotational motion).

1.3. In order to attach the samples onto the top stage manipulators, fabricate custom sample holders that can support a glass slide; the top crystal will be placed on the glass slide (**Figure 1d**).

1.4. For the bottom manipulators, place a flat heating element in a machined glass-ceramic holder (**Figure 1g**) and affix it to the rotating stage. Connect the heating element to a power supply and a temperature controller.

1.5. Program the controllers with an instrumentation software (e.g., LabVIEW) to control the relative position of the manipulators (**Figure 2**).

1.5.1. To perform the necessary motions, program the software with the following capabilities: individually or simultaneously move the manipulators; read, save and retrieve the position of each manipulator; easily adjust the speed of the manipulators, and autofocus the bottom stage. Build-in safety features to prevent any possible collisions between the sample and the lens.

2. Mechanical exfoliation of a 2D crystal.

2.1. Prepare a substrate for the mechanical exfoliation procedure.

2.1.1. Submerge 1 cm x 1 cm squares of silicon/silicon oxide (Si/SiO_2) wafers into a beaker filled with acetone and place the beaker into an ultrasonic cleaner for 10 min.

2.1.2. Individually remove the wafers from the beaker with tweezers and rinse them with isopropanol (IPA) then dry them with a nitrogen (N_2) gun.

NOTE: When working with acetone and IPA, it is suggested to do so under a fume hood while wearing the proper PPE.

2.2. Mechanically exfoliate the crystal onto the substrate.

2.2.1. Using tweezers, carefully remove a portion of the crystal and place it on a piece of semiconductor-grade adhesive tape.

2.2.2. Take a second piece of adhesive tape and firmly press it against the initial tape with crystal then peel away the two pieces of tape. After repeating several times, many thinner pieces of crystal will be found on the tape.

2.2.3. Press the adhesive tape with the thin 2D crystals onto a freshly cleaned substrate such that the crystal is in direct contact with the substrate and peel away the tape to leave exfoliated flakes on the substrate.

2.3. To remove any residual adhesive, place the resulting samples (substrates with 2D crystals exfoliated onto their surface) in a beaker filled with acetone for 10 min. Remove the samples using tweezers, rinse them with IPA, and dry with a N_2 gun.

2.4. Use an optical microscope to examine the exfoliated flakes. Estimate their thickness by

assessing the flake's optical contrast with the substrate[22]. Image the flakes using atomic force microscopy (AFM) in tapping mode (see the **Table of Materials**) to better quantify the surface morphology and to measure the flake thickness.

3. PMMA-PVA method for fabricating van der Waals heterostructures (top substrate preparation).

3.1. Prepare the top substrate for the transfer procedure by exfoliating the crystal on a poly(methyl methacrylate) (PMMA) film attached to a glass slide (**Figure 3a-d**).

3.1.1. Follow the procedure described in step 2.1. to obtain a clean substrate. Spin coat a layer of polyvinyl alcohol (PVA) on the substrate at 3000 rpm for 1 min by following the protocol described in the instrument's user manual.

NOTE: When using the spin coater, it is suggested to do so under a fume hood while wearing proper PPE.

3.1.2. Directly place the substrate on a hot plate and bake it uncovered in air at 75 °C for 5 min.

3.1.3. Spin coat a layer of PMMA on the substrate from step 3.1.2 by following a procedure similar to the one in step 3.1.1, but this time set the spinning parameters to an angular velocity of 1500 rpm for 1 min (**Figure 3a**).

3.1.4. Directly place the substrate on a hot plate and bake it uncovered in air at 75 °C for 5 min.

3.1.5. Remove the substrate from the hot plate and place pieces of adhesive tape along its edges to create a tape frame. Then, mechanically exfoliate a 2D crystal on the PMMA surface by following step 2.2 (**Figure 3b**).

3.1.6. Use sharp tweezers to separate the PMMA from the PVA by slowly peeling back the tape frame. The PMMA layer and exfoliated crystal along with the tape frame will detach from the PVA and Si/SiO_2 substrate (**Figure 3c**).

3.1.7. Invert the tape frame and place it on a machined support such that the crystal is facing downwards (**Figure 3d**).

NOTE: This support enables the user to place the tape frame under an optical microscope to inspect the exfoliation onto PMMA and identify a flake with the desired thickness and geometry.

3.1.8. Use sharp tweezers and the optical microscope to place a small washer (0.5 mm inner radius) precisely on the PMMA film such that it surrounds the desired flake (**Figure 3d**).

3.1.9. Lower a glass slide and adhere it to the polymer by pressing it against the exposed tape.

4. Polydimethylsiloxane (PDMS) stamping method for fabricating van der Waals heterostructures (top substrate preparation).

4.1. Prepare a polypropylene carbonate (PPC) solution for spin coating by mixing three parts PPC crystal with seventeen parts anisole. This is done by letting the solution mix in a stirrer for approximately 8 h or until the solution is homogeneous.

4.2. Prepare the top substrate for the transfer procedure by exfoliating crystal on a PPC film and by then placing it on a PDMS stamp attached to a glass slide (**Figure 4a-d**).

4.2.1. Follow the procedure in step 2.1. to obtain a clean substrate. Spin coat a layer of PPC on the substrate at 3000 rpm for 1 min (**Figure 4a**).

4.2.2. Directly place the substrate on a hot plate and bake it uncovered in air at 75 °C for 5 min.

4.2.3. Remove the substrate from the hot plate and place pieces of adhesive tape along its edges to create a tape frame.

4.2.4. Mechanically exfoliate the 2D layered crystal on the PPC coated substrate by following step 2.2 and use the optical microscope to identify a flake with the desired thickness and geometry. (**Figure 4b**).

4.2.5. Use scissors or a surgical blade to cut a piece of PDMS into a 2 mm x 2 mm square and place it in an oxygen plasma etcher for 2 min at 50 W and 53.3 Pa.

4.2.6. At the end of the cycle, press a glass slide on the PDMS stamp to bond the two together. Place the glass slide and PDMS stamp back into the oxygen plasma etcher to undergo the same cycle. Remove the glass slide when the cycle has ended.

4.2.7. Using tweezers, carefully peel back the tape frame and pick-up the PPC film with the exfoliated crystal (**Figure 4c)** and place it on the PDMS stamp such that the desired flake is located on the stamp.

5. Transferring flakes from the top substrate to the bottom substrate using the PMMA-PVA method (Figure 3e-h).

5.1. Place a substrate on the bottom stage of the transfer setup. On this substrate, identify the position of the desired flake. This flake will be the “bottom” crystal. Also, place the top substrate (the glass slide from step 3.1.9) into the top substrate holder of the transfer setup (**Figure 3e**).

5.2. Using a low-magnification objective (5x), bring the bottom substrate into focus and center the desired flake. Slowly lower the top substrate until it enters the depth of field of the objective. Adjust the lateral position and the rotational alignment of the two flakes.

5.3. Employ a higher-magnification objective (50x) and continue to lower the top substrate while adjusting the flake alignment. Lower the top substrate until the top flake entirely contacts the bottom flake. Contact is noticeable by a sudden change of color.

5.4. Heat the bottom substrate to 75 °C for better adhesion of PMMA to the bottom substrate. The PMMA will detach from the glass slide (**Figure 3f**).

5.5. Clean the bottom substrate following step 2.3 to remove the PMMA film (**Figure 3g-h**).

6. Transferring flakes from the top substrate to the bottom substrate using the stamping method (Figure 4d-f).

6.1. Place a substrate on the bottom stage of the transfer setup. On this substrate, identify the position of the desired flake. This flake will be the "bottom" crystal. Also, place the top substrate (the glass slide with PDMS from step 4.2.5–4.2.6) into the top substrate holder of the transfer setup (**Figure 4d**).

6.2. Heat the bottom substrate to 100 °C then follow steps 5.2–5.4 to align and bring into contact the top crystal with the bottom flake (**Figure 4e**).

6.3. Once complete contact is made between the two flakes (**Figure 4e**), slowly raise the top substrate. This results in the drop-off of the top flake from the stamp to the bottom substrate (**Figure 4f**).

REPRESENTATIVE RESULTS:

To illustrate the outcomes and effectiveness of our procedure we present a sequence of angle-controlled stacks of rhenium disulfide (ReS_2) thin crystals. To emphasize that the described method can also be applied to atomically thin layers, we also exemplify the construction of two relatively twisted monolayers of molybdenum disulfide (MoS_2).

To demonstrate the angular alignment capabilities of the transfer setup we use rhenium disulfide (ReS_2). Because of its in-plane anisotropic lattice structure, this crystal mechanically exfoliates as elongated bars with well-defined edges[23,24]. This makes it a perfect candidate for the demonstration of angular alignment. Using the PDMS stamping method described in the protocol, we transferred a "top" ReS_2 crystal from the stamp onto a silicon substrate with previously exfoliated "bottom" ReS_2. Each time we aimed to align the edges at a specific angle. Employing the angular component of the transfer setup, the "top" ReS_2 flake was placed such that it would be twisted by the specific desired angle with respect to the already present "bottom" ReS_2.

Figure 5 shows an example of a transfer where a top flake of ReS_2 was placed on a bottom ReS_2 flake with an intended relative angle of 75°. The optical micrographs of the bottom and top crystals are shown in **Figure 5a** and **Figure 5b** respectively. Using the PDMS stamping method described in the protocol, a stack was fabricated and the resulting new crystal is shown in **Figure**

5c. Atomic force microscopy (AFM) was used to image the stack in **Figure 5d;** this demonstrates that the twist angle between the top and bottom flakes measured 74.6° ± 0.1° (mean ± SD). The error bar was calculated from the uncertainty in precisely defining the edges of the ReS_2 in the micrographs. To further display the angular precision of the transfer setup we repeated this process for several other samples with intended relative angles ranging from 15° to 90° in increments of 15° (**Figure 6**).

Using the PMMA-PVA procedure as described in the protocol, the transfer setup is successfully used to create a structure consisting of two monolayer flakes of molybdenum disulfide (MoS_2). The individual monolayers are exfoliated onto PMMA (**Figure 7a**) and Si/SiO_2 respectively (**Figure 7b**). Our transfer procedure results in the structure presented in the optical micrograph in **Figure 7c**. Its morphology is further characterized by atomic force microscopy, confirming the thickness and relative position of the stacked MoS_2 monolayer (**Figure 7d**).

FIGURE AND TABLE LEGENDS:

Figure 1: Components of the transfer setup. (**a**) The optical microscope equipped with the camera and three long working distance objectives. (**b**) The microscope with the top manipulators which are capable of moving in the XYZ (translational) directions. (**c**) The top manipulators**.** (**d**) A custom machined arm which fastens to the top manipulators and is used to hold a glass slide (top sample). (**e**) The microscope with the bottom manipulators which can move in XYZ (translational) and Θ (rotational). (**f**) The bottom manipulators. (**g**) A custom thermal insulating bottom stage holder (left) which is directly attached to the rotating stage. The temperature controlled flat heating element is shown on the right.

Figure 2: Instrumentation software front panel responsible for controlling the manipulators. The front panel is divided into two sections; the left section controls the bottom stage whereas the right section controls the top manipulators. The user can move the manipulators individually or simultaneously, read out the position of each manipulator, save positions, adjust the speed of the manipulators, and automatically focus.

Figure 3: PMMA-PVA substrate preparation and transfer procedure. (**a**) A bare Si/SiO_2 substrate is first spin coated with PVA followed by PMMA. (**b**) A tape frame is placed on the substrate to hold down the polymer during mechanical exfoliation. (**c**) The tape frame is peeled back to pick-up the PMMA layer with the exfoliated crystal. (**d**) The tape frame is inverted and placed on a machined support followed by an inspection under an optical microscope. A washer is placed on the polymer film such that it surrounds the desired flake. (**e**) A glass slide is adhered to the tape frame and is placed into the top manipulator of the transfer setup. A previously prepared substrate with exfoliated crystal is placed on the bottom stage. (**f**) The top and bottom flakes are aligned and brought into contact. By heating the bottom stage to 75 °C, the PMMA detaches from the glass slide leaving (**g**) the top flake, the PMMA film and the washer on the bottom substrate. (**h**) The washer is removed and the PMMA is washed away resulting in a heterostructure.

Figure 4: PDMS stamping preparation and transfer procedure. (**a**) A bare Si/SiO_2 substrate is spin

coated with PPC. (**b**) A tape frame is placed on the substrate to hold down the polymer during mechanical exfoliation. (**c**) The tape frame is peeled back to pick-up the PPC layer with the exfoliated crystal. (**d**) The glass slide is placed into the top manipulator of the transfer setup. A previously prepared substrate with exfoliated crystal is placed on the bottom stage and heated to 100 °C. (**e**) The top and bottom flakes are aligned and brought into contact. (**f**) At 100 °C, we can slowly raise the top stage which results in the drop-off of the flake from the stamp to the bottom substrate.

Figure 5: ReS_2 flakes transferred with a relative angle of 75°. Optical micrographs of (**a**) a ReS_2 crystal mechanically exfoliated on a Si/SiO_2 substrate, (**b**) a ReS_2 crystal mechanically exfoliated on a Si/SiO_2/PPC substrate, (**c**) the resulting structure after transfer. (**d**) An AFM topographic map of the final structure corresponding to the boxed area in (c). The measured angle between the ReS_2 flakes is 74.6° ± 0.1°.

Figure 6: ReS_2 flakes transferred at specific relative angles. (**a**–**f**) Optical micrographs of ReS_2 flakes transferred with the intention of forming angles ranging from 15° to 90° in increments of 15°.

Figure 7: Twisted MoS_2 bilayer. Optical micrographs of (**a**) the top MoS_2 flake on a PMMA film, (**b**) the bottom MoS_2 flake on a Si/SiO_2 substrate, (**c**) and the twisted MoS_2 structure. All scale bars in the optical images measure 5 μm. (**d**) AFM image of the twisted MoS_2 structure with a line profile (inset) along the direction indicated by the black dashed line.

DISCUSSION:

The home-built transfer setup presented here offers a method for building novel layered materials with both lateral and rotational control. Compared to other solutions described in the literature[10,25], our system does not require complex infrastructure, yet it achieves the goal of controlled alignment of 2D crystals.

The most critical step in the procedure is that of aligning and placing the top crystal in contact with the bottom one. Vibrations could be a cause for a failed alignment, therefore, one must minimize their effect. In this regard, the advantage of the “hands-free” set-up presented here is that the user does not risk introducing vibrations caused by the manual handling of the manipulators. Further improvements can be achieved by placing the set-up in an environment with lower vibrations or on a table equipped with vibration damping mechanisms.

As rotational alignment is becoming a more and more important parameter to consider when creating van der Waals heterostructures, the rotational capability of this transfer setup is one of its strengths. The fact that the optical microscope is limited in resolving the edges of both crystals represents main the limitation in the alignment precision.

Not all 2D materials are inert in air. Crystals such as black phosphorus (BP)[26] or chromium triiodide (CrI_3)[27,28] are known to degrade upon exposure to air. Therefore, to create heterostructures using these materials, and preserve the pristine interface, the transfer

procedure would have to take place in an inert environment such as inside a glovebox. Because the transfer instrumentation presented here is “hands-free”, it can be operated in a glovebox where these inert materials could be used.

Lastly, the stacking method presented here can have broader applicability and can be extended to situations when two crystals or a crystal and a substrate need to be precisely aligned laterally and rotationally.

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DISCLOSURES:

The authors have nothing to disclose. The authors have no competing financial interests.

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Figure 1

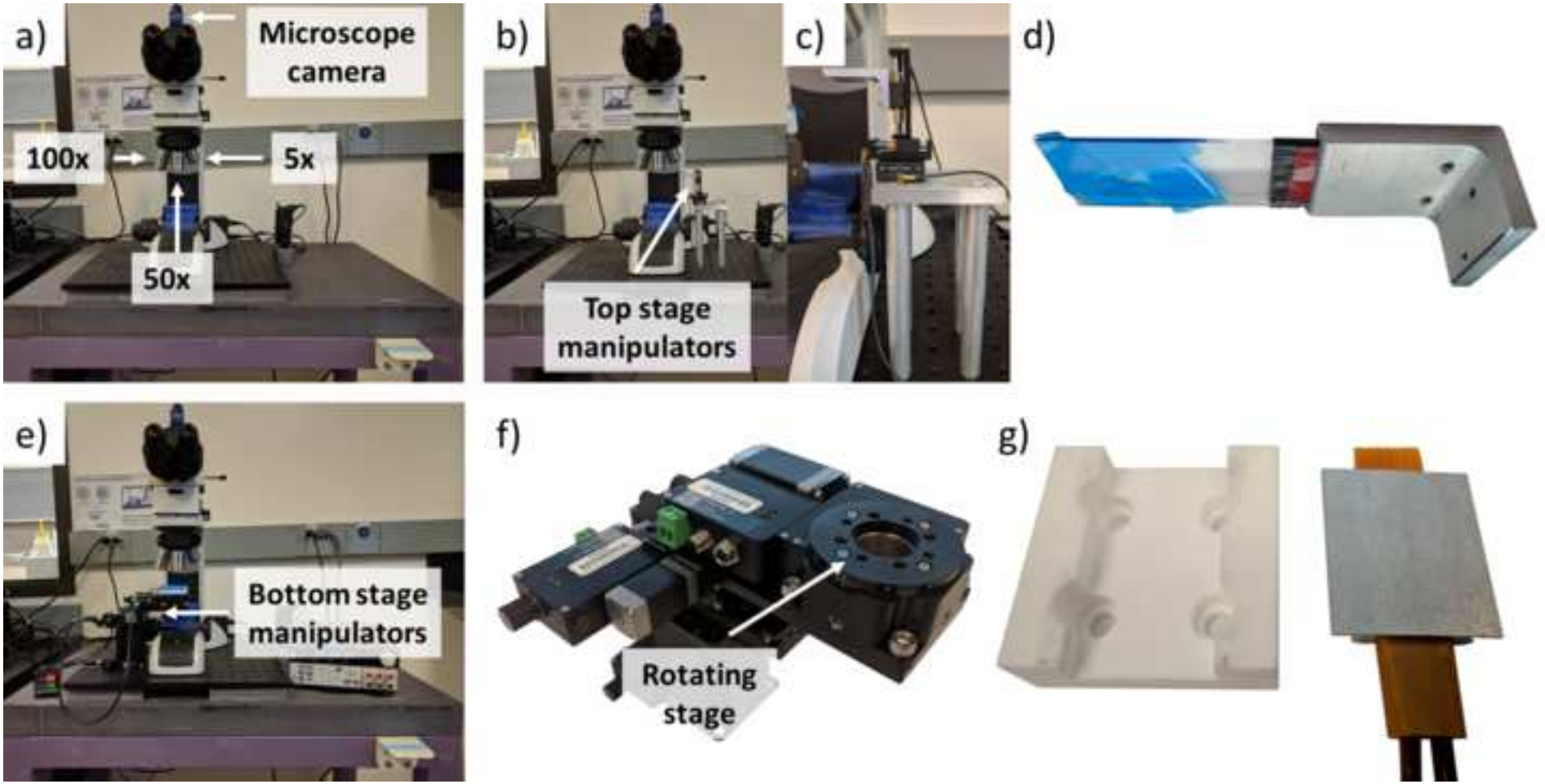

Figure 2

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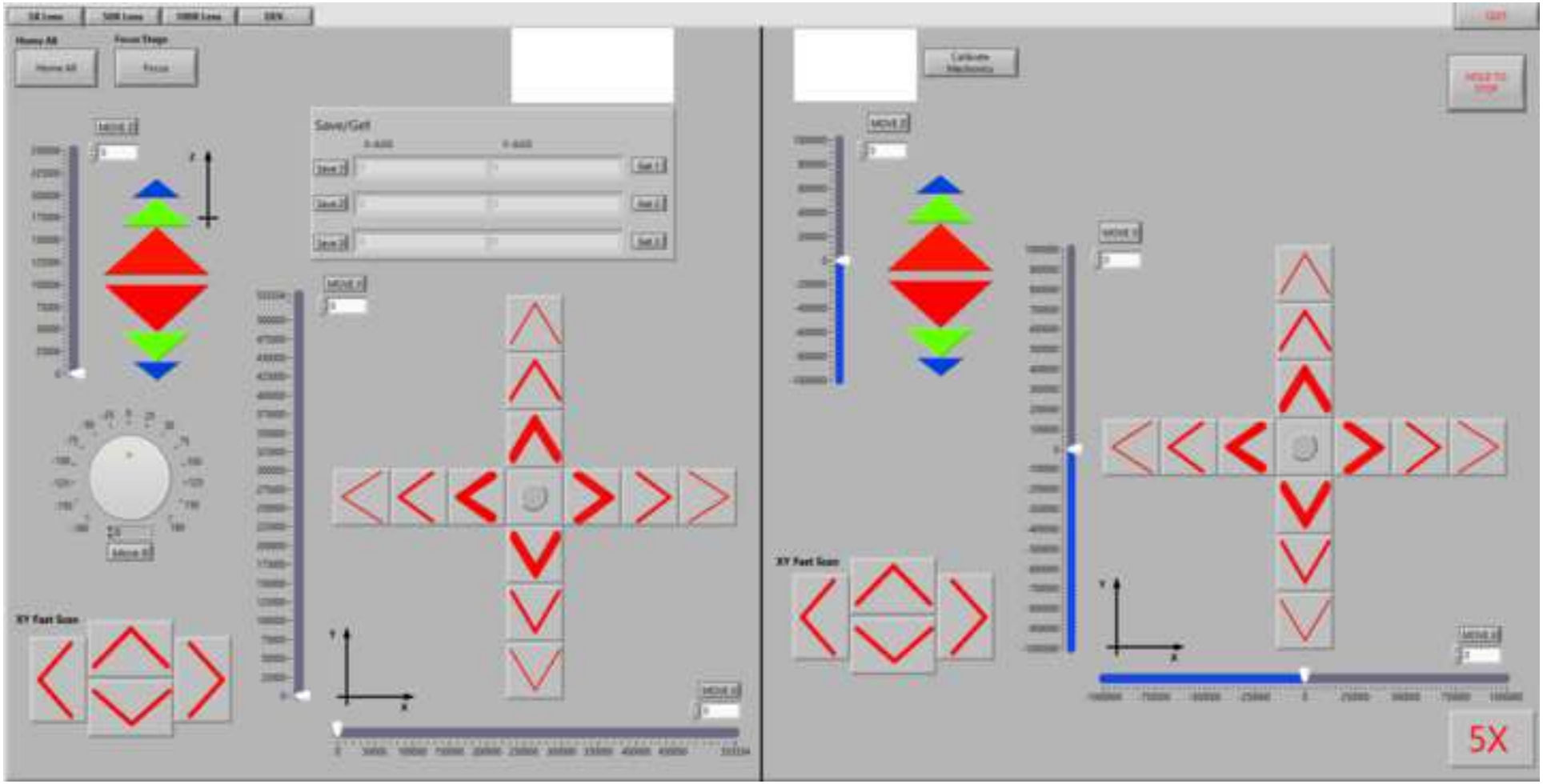

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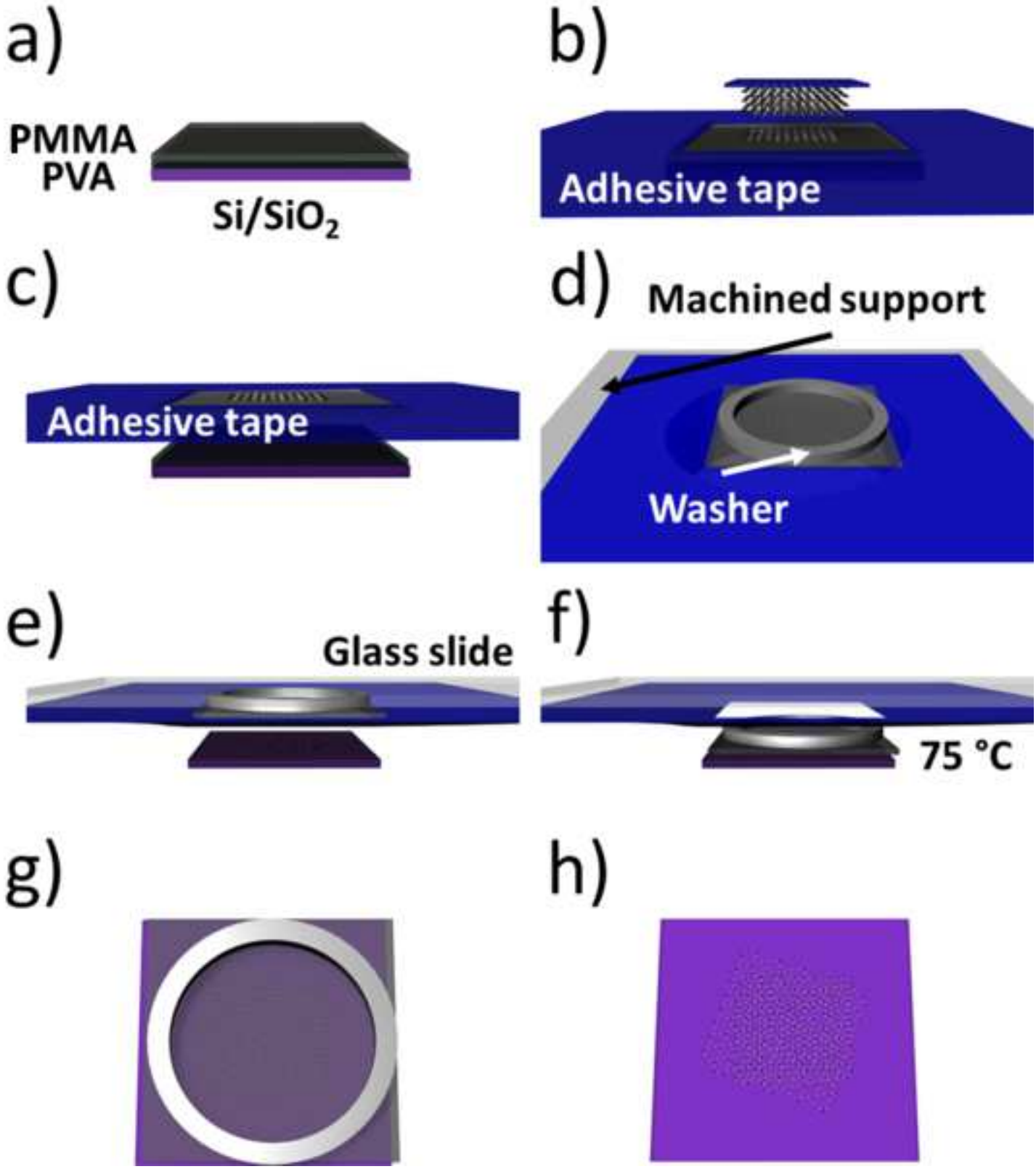

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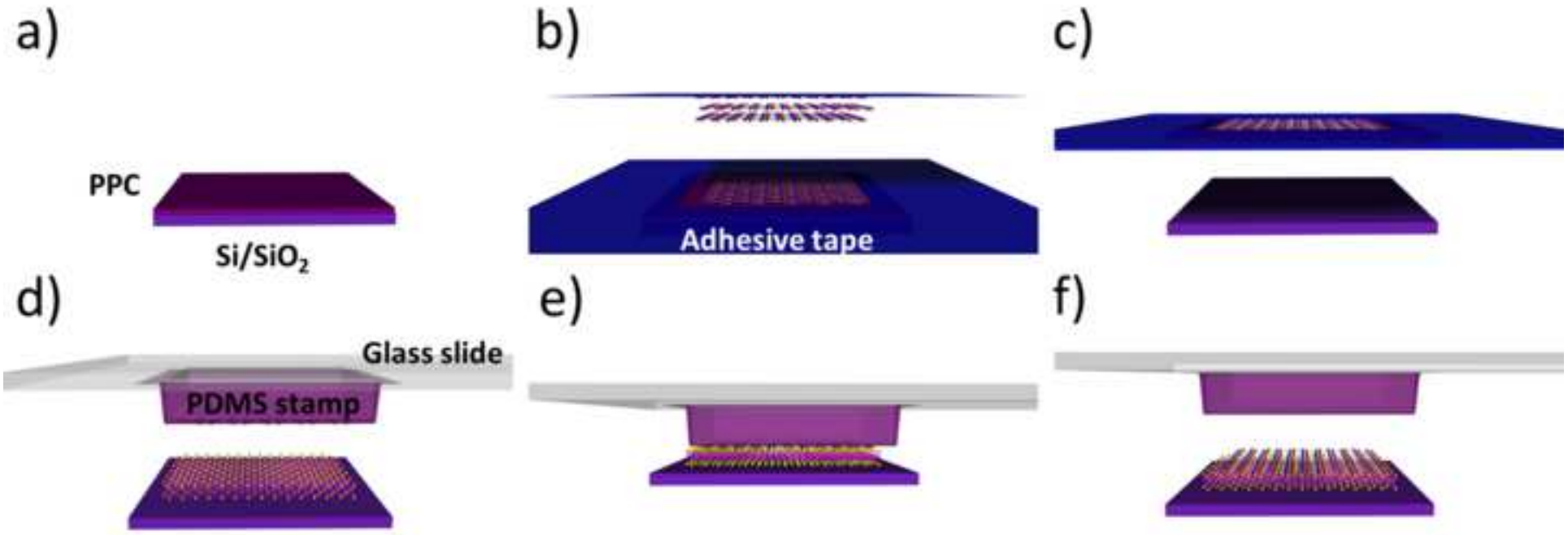

Figure 5

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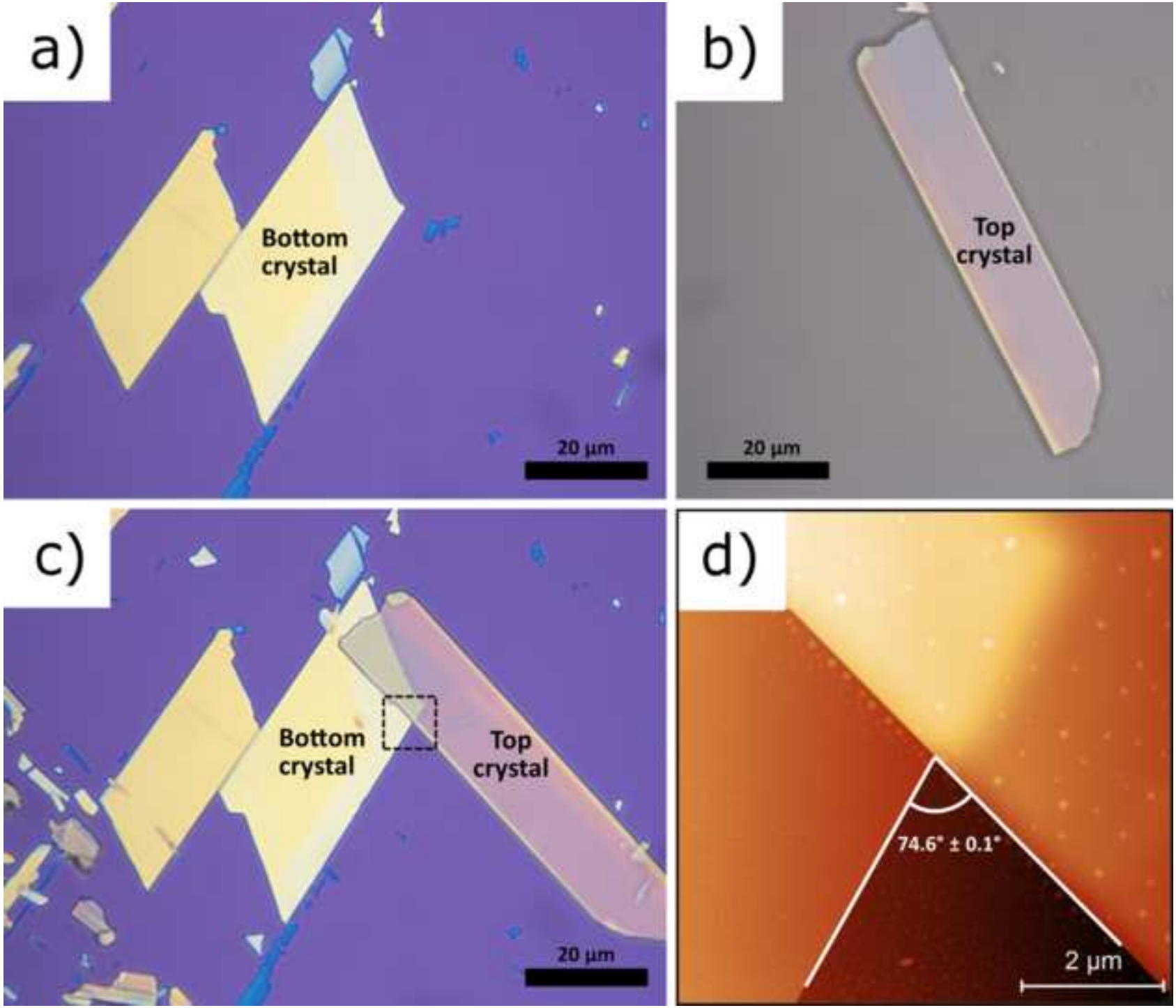

Figure 6

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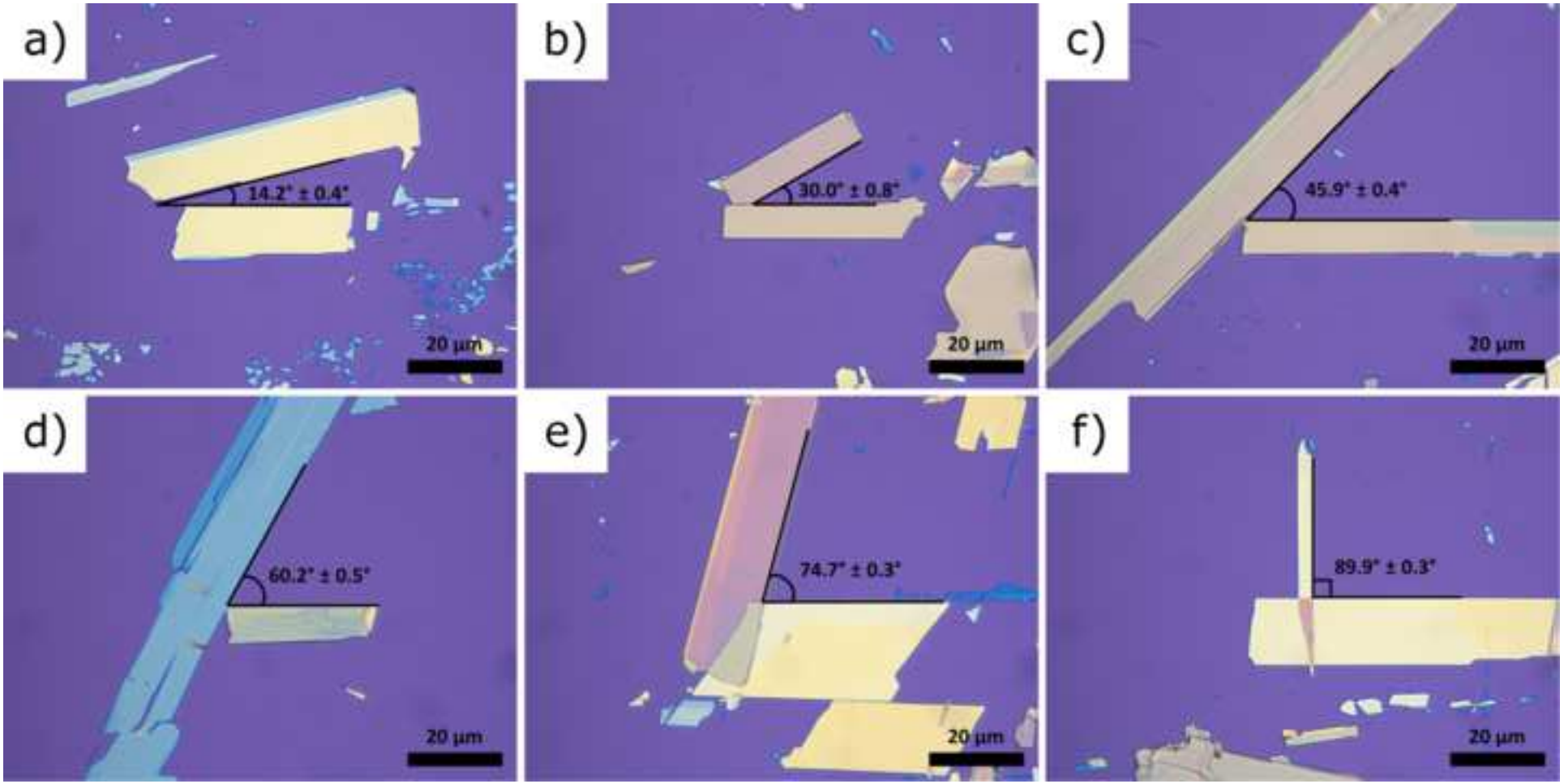

Figure 7

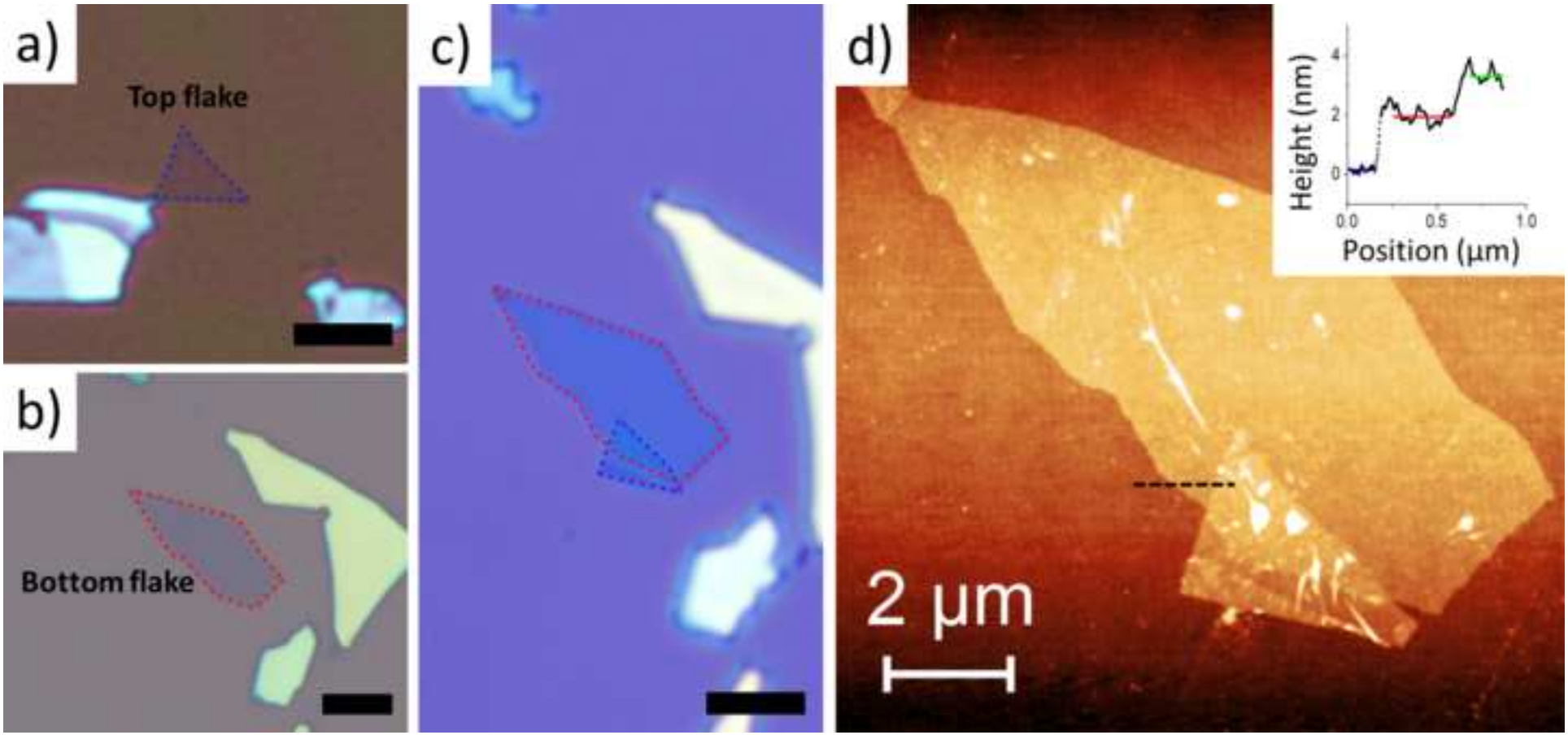

Table of Materials

Click here to access/download;Table of Materials;JOVE_Materials_Final2.0.xlsx

Name of Material/ Equipment	Company	Catalog Number
5X objective lens	Nikon Metrology	MUE12050
50X objective lens	Nikon Metrology	MUE21500
100X objective lens	Nikon Metrology	MUE21900
Acetone	Sigma-Aldrich	270725
Adhesive tape	Ultron Systems, Inc.	
Anisole	MicroChem	
Atomic force microscope	Bruker	Dimension Icon
Bottom stage rotation manipulator	Zaber Technologies	X-RSW60A-PTB2
Bottom stage X manipulator	Zaber Technologies	X-LSM025A-PTB2
Bottom stage Y manipulator	Zaber Technologies	X-LSM025A-PTB2
Bottom stage Z manipulator	Zaber Technologies	X-VSR40A-KX14A
Isopropanol	Sigma-Aldrich	563935
LabVIEW software	National Instruments	
Macor	McMaster-Carr	8489K238
Microscope camera	Zeiss	426555-0000-000
Molybdenum disulfide (MoS_2)	HQ Graphene	
Optical breadboard	Thorlabs, Inc.	MB4545/M
Optical microscope	Nikon Metrology	LV150N
Oxygen plasma etcher	Plasma Etch, Inc.	PE-50
PDMS stamp	Gel-Pak	PF-20-X4
PMMA 950 A6	MichroChem Corp.	M230006 0500L1GL
Polypropylene carbonate	Sigma-Aldrich	389021-100g
PVA Partall #10	Composites Canada	
Rhenium disulfide (ReS_2)	HQ Graphene	
Si/SiO_2 substrate	Nova Electronics Materials	HS39626-OX
Spin coater	Laurell Technologies	WS-650-23
Temperature controller	Auber Instruments	SYL-23X2-24
Top stage controller unit	Mechonics	CF.030.0003
Top stage X manipulator	Mechonics	MS.030.1800
Top stage Y manipulator	Mechonics	MS.030.1800
Top stage Z manipulator	Mechonics	MS.030.3000
Ultrasonic bath	Elma Schmidbauer GmbH	Elmasonic P 30 H

Comments/Description

23.5 mm working distance and 0.15 numerical aperture
19 mm working distance and 0.4 numerical aperture
4.5 mm working distance and 0.8 numerical aperture
Purity ≥99.90%

We typicall use the ScanAsyst mode
360° travel with step size of 4.091 µrad
25 mm travel with step size of 47.625 nm
25 mm travel with step size of 47.625 nm
40 mm travel with step size of 95.25 nm
Purity 99.999%

5 megapixel, 47 fps live frame rate, exposure time of 100 µs - 2 s, color camera

Controls the temperature of the bottom stage via a J type thermocouple

18 mm travel with step size of 11 nm
18 mm travel with step size of 11 nm
30 mm travel with step size of 11 nm

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Author(s): Adina Luican Mayer, Justin Bohdanowicz, Ryan Plumadore

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CORRESPONDING AUTHOR

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Title:	Assistant Professor
Signature:	
Date:	23 Jan 2019

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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:

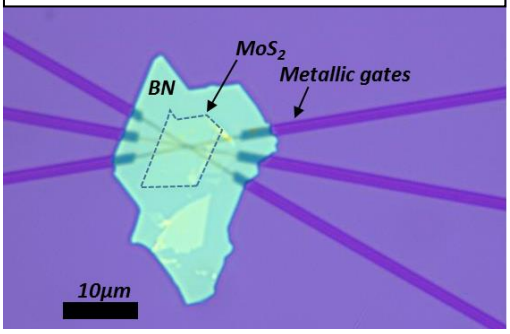
- *In this manuscript, the authors presented their home-built computer-controlled transfer setup to stack 2D material flakes, as well as their complete dry transfer procedure. This setup can realize computer-controlled in-plane and angular alignment between flakes, which is good.*

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.

Heterostructure fabricated using the "pick-up" technique



- *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 motonized 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 75º? There is no obvious reason. It seems as if they failed in getting 60º or 90º and afterwards they claim that they intended 75º.

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 15º to 90º in steps of 15º. 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 º or 60 º).

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.