FINAL SCRIPT: APPROVED FOR FILMING

Submission ID #: 68540
Scriptwriter Name: Poornima G
Project Page Link: https://review.jove.com/account/file-uploader?src=20904748

Title: Residue-Free Fabrication of van der Waals Heterostructures of Two-Dimensional Materials

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Author Questionnaire

1. Microscopy: Does your protocol require the use of a dissecting or stereomicroscope for performing a complex dissection, microinjection technique, or something similar? **No**

2. Software: Does the part of your protocol being filmed include step-by-step descriptions of software usage? **No**

3. Filming location: Will the filming need to take place in multiple locations? **No**

Current Protocol Length

Number of Steps: 13

Number of Shots: 18

Introduction

Videographer: Obtain headshots for all authors available at the filming location.

1.1. **Changho Kim:** We aim to develop a residue-free fabrication method for 2D heterostructures that avoids exposure to any residues and temperature changes, while ensuring the devices remain clean and high quality.

1.1.1. INTERVIEW: Named talent says the statement above in an interview-style shot, looking slightly off-camera. *Suggested B-roll: 2.3.1*

What are the current experimental challenges?

1.2. **Minyoung Lee:** Transferring individual 2D flakes for multi-stacked heterostructures is technically challenging, and polymers or solvents often leave residues at the interface, disrupting clean stacking between layers.

1.2.1. INTERVIEW: Named talent says the statement above in an interview-style shot, looking slightly off-camera. *Suggested B-roll: 3.1.1*

What significant findings have you established in your field?

1.3. **Changho Kim:** We established a novel method that enables precise 2D heterostructure assembly using only van der Waals forces through bottom-up, top-down, and modular stacking techniques.

1.3.1. INTERVIEW: Named talent says the statement above in an interview-style shot, looking slightly off-camera. *Suggested B-roll: 4.2.1*

What research questions will your laboratory focus on in the future?

1.4. **Jae Hun Seol:** We plan to apply this method to various applications, such as optical or gas sensors, that require complex heterostructures with clean and high-quality interfaces.

1.4.1. INTERVIEW: Named talent says the statement above in an interview-style shot, looking slightly off-camera. *Suggested B-roll: 2.2.1*

Videographer: Obtain headshots for all authors available at the filming location.

Protocol

2. Acquiring the Residue-Free Region

Demonstrator: Minyoung Lee

Videographer: Please film the steps 2.1 to 2.5

2.1. To begin, prepare a piece of bulk crystal using a cotton swab and a razor blade **[1]**.

2.1.1. WIDE: Talent using a razor blade and cotton swab to extract a section of bulk crystal.

2.2. Attach the prepared crystal onto the adhesive surface of a pre-exfoliation stamp **[1]** and place another pre-exfoliation stamp on top of the crystal to form a sandwich configuration **[2]**.

2.2.1. Talent placing the crystal onto the sticky surface of the first stamp.

2.2.2. Talent aligning and pressing a second stamp over the crystal to make a sandwich.

2.3. Gently exfoliate the sandwiched crystal **[1]** and repeat this process 3 to 5 times to obtain a clean and uniform sub-micron-thick crystal **[2]**.

2.3.1. Talent lifting and pressing the stamps repeatedly to exfoliate the crystal.

2.3.2. Shot of a clean and uniform sub-micron-thick crystal.

2.4. Now, place the pre-exfoliated crystal onto the sample stage **[1]** and secure the tape stamp with a tilt angle of at least 5 degrees onto the magnetic plate of the stamp manipulator using a magnet **[2]**.

2.4.1. Talent positioning the exfoliated crystal on the flat sample stage.

2.4.2. Talent attaching the tilted tape stamp to the magnetic holder using a small magnet.

2.5. Align the tape stamp above the pre-exfoliated crystal by adjusting the sample stage **[1]**.

2.5.1. Talent slowly turning knobs or sliding controls to center the tape stamp over the crystal.
AUTHOR'S NOTE: Please use the shot showing adjustment of the sample stage's x-axis. Although we also recorded a version adjusting the stamp manipulator's x-axis, this should not be used

2.6. Attach the tape stamp to the top surface of the crystal by adjusting the stamp manipulator in the negative Z direction and gently exfoliate the thin residue-free region by adjusting the stamp manipulator in the positive Z direction **[1]**.

2.6.1. SCOPE: Video 1.avi 00:00 – 00:11

2.7. Adhere the residue-free region firmly to the substrate by adjusting the stamp manipulator in the negative Z direction **[1]**.

2.7.1. SCOPE: Video 2.avi 00:00 – 00:02 and FILE: Video 3.avi 00:04 – 00:04

2.8. Then, exfoliate the single flake by adjusting the stamp manipulator in the positive Z direction **[1]**.

2.8.1. SCOPE: Video 2.avi 00:04 – 00:08 and FILE: Video 3.avi 00:06 – 00:12

3. Fabrication of Residue-Free van der Waals Heterostructure Assemblies Through Bottom-Up Stacking Process

3.1. Pick up the molybdenum disulfide flake with the MoS_2 *(M-O-S-2)* residue-free stamp **[1-TXT]** and release it onto the h-BN *(H-B-N)* flake, to assemble the structure **[2-TXT]**.

3.1.1. SCOPE: Video 4.avi 00:00 – 00:05. **TXT: MoS_2: Molybdenum Disulfide**

3.1.2. SCOPE: Video 4.avi 00:08 – 00:14 **TXT: h-BN: Hexagonal Boron Nitride**

3.2. Pick up another h-BN flake and release it on the MoS_2 and h-BN structure **[1]**.

3.2.1. SCOPE: Video 4.avi 00:16 – 00:31

3.3. Next, pick up the h-BN flake using the MoS_2 residue-free stamp **[1]**.

3.3.1. SCOPE: Video 5.avi 00:04 – 00:07

3.4. Use the picked-up HBN flake to cover the MoS_2 flake, and then pick up the MoS_2 flake with the already picked-up h-BN flake, forming a h-BN and MoS_2 structure **[1]**.

3.4.1. SCOPE: Video 5.avi 00:08 – 00:12

3.5. Finally, release the combined structure onto another h-BN flake **[1]** and then release the Hetero B onto the Hetero A **[2]**.

3.5.1. SCOPE: Video 5.avi 00:13 – 00:20

3.5.2. SCOPE: Video 6.avi 00:04 – 00:11

Results

4. Results

4.1. The symmetrical hexagonal atomic structure of residue-free MoS_2 was confirmed using high-resolution transmission electron microscopy and selected area electron diffraction pattern analysis **[1]**.

4.1.1. LAB MEDIA: Figure 7.

4.2. Energy-dispersive X-ray spectroscopy mapping revealed strong molybdenum and sulfur signals **[1]** and minimal carbon and oxygen, confirming the absence of polymer residue and oxidation on the molybdenum disulfide flakes **[2]**.

4.2.1. LAB MEDIA: Figure 7A. *Video editor: Highlight the EDS maps showing MO and S images*.

4.2.2. LAB MEDIA: Figure 7A. *Video editor: Highlight the EDS maps showing C and O images.*

4.3. Atomic force microscopy analysis showed that the thicknesses of transferred h-BN and MoS_2 flakes were 18.34 nanometers and 5.27 nanometers, respectively **[1]**.

4.3.1. LAB MEDIA: Figure 8. *Video editor: Highlight the values 18.34 nm and 5.27 nm in B and E*.

4.4. The root-mean-square roughness values of 0.143 nanometers for h-BN and 0.153 nanometers for MoS_2 confirmed excellent flatness with minimal surface irregularities **[1]**.

4.4.1. LAB MEDIA: Figure 8. *Video editor: Highlight the blue boxes displaying the Rq values 0.143 nm and 0.153 nm in B and E.*

4.5. Raman spectroscopy analysis revealed peak positions at approximately 1365 and 383 inverse centimeters for h-BN and MoS_2, respectively, indicating strain-free transfer **[1]**.

4.5.1. LAB MEDIA: Figure 8. *Video editor: Highlight the peaks E2g in C and E12g in F*.

4.6. Distinct manipulation methods allowed successful assembly of the heterostructures via both bottom-up and top-down stacking **[1]**.

4.6.1. LAB MEDIA: Figure 9. *Video editor: Highlight the labeled regions "Hetero A" and "Hetero B" in panels A and B.*

4.7. The assembled heterostructures exhibited randomly distributed gas-filled blisters at the heterointerface without any applied force **[1]**.

4.7.1. LAB MEDIA: Figure 10B. *Video editor: Highlight the optical image of "as assembled" showing "gas blisters".*

4.8. Application of increasing forces from 30 to 1000 nano-newtons progressively reduced blister size and count **[1]**.

4.8.1. LAB MEDIA: Figure 10B. *Video editor: Sequentially highlight the images and the corresponding graphs from "30" to "1000 nN".*

Pronunciation Guide:

1. van der Waals

Pronunciation link (Merriam-Webster):
https://www.merriam-webster.com/dictionary/van%20der%20Waals
IPA: /væn dər ˈvɑlz/
Phonetic Spelling: van-der-VAHLZ

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2. heterostructure

Pronunciation link (Merriam-Webster — hetero-):
https://www.merriam-webster.com/dictionary/hetero
(and structure)
IPA: /ˌhɛtərəʊˈstrʌktʃər/ → American /ˌhɛtərəˈstrʌktʃɚ/
Phonetic Spelling: HET-uh-roh-STRUK-cher

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3. molybdenum disulfide (MoS_2)

Pronunciation link (Merriam-Webster — molybdenum):
https://www.merriam-webster.com/dictionary/molybdenum
IPA: /məˈlɪbdəˌnəm daɪˈsʌlfaɪd/
Phonetic Spelling: muh-LIB-duh-num die-SUL-fide

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4. hexagonal boron nitride (h-BN)

Pronunciation link (Merriam-Webster — hexagonal):
https://www.merriam-webster.com/dictionary/hexagonal
IPA: /hɛkˈsægənəl ˈbɔrɒn naɪˌtraɪd/
Phonetic Spelling: hek-SAG-uh-nul BOR-on NY-tride

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5. exfoliate

Pronunciation link (Merriam-Webster):
https://www.merriam-webster.com/dictionary/exfoliate
IPA: /ɪkˈsfoʊliˌeɪt/
Phonetic Spelling: ik-SFOH-lee-ayt

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6. nanometer

Pronunciation link (Merriam-Webster):
https://www.merriam-webster.com/dictionary/nanometer
IPA: /ˈnænəˌmiːtər/

Phonetic Spelling: NAN-uh-mee-ter

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7. atomic force microscopy

Pronunciation link (Merriam-Webster — atomic):
https://www.merriam-webster.com/dictionary/atomic
Pronunciation link (Merriam-Webster — microscopy):
https://www.merriam-webster.com/dictionary/microscopy
IPA: /əˈtɑmɪk fɔrs maɪˈkrɑskəpi/
Phonetic Spelling: uh-TOM-ik force my-KROS-kuh-pee

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8. Raman spectroscopy

Pronunciation link (Merriam-Webster — Raman):
https://www.merriam-webster.com/dictionary/raman
Pronunciation link (Merriam-Webster — spectroscopy):
https://www.merriam-webster.com/dictionary/spectroscopy
IPA: /ˈrɑːmɑn spɛkˈtrɑskəpi/
Phonetic Spelling: RAH-man spek-TRAW-skuh-pee

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9. energy-dispersive X-ray spectroscopy (EDS/XPS)

Pronunciation link (Merriam-Webster — dispersive):
https://www.merriam-webster.com/dictionary/dispersive
IPA: /ˈɛnərdʒi dɪˈspɜrsɪv ɛks reɪ spɛkˈtrɑskəpi/
Phonetic Spelling: EN-er-jee dis-PUR-siv ex RAY spek-TRAW-skuh-pee

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10. nanonewton

Pronunciation link (Merriam-Webster — newton):
https://www.merriam-webster.com/dictionary/newton
IPA: /ˈnænəˈnuˌtɑn/
Phonetic Spelling: NAN-uh-NEW-ton