Sensors and Materials, Vol. 35, No. 6 (2023) 1929–1939
MYU Tokyo

S & M 3299

Transfer of van der Waals Heterostructures of Two-dimensional Materials onto Microelectromechanical Systems

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(Received February 24, 2023; accepted April 18, 2023)

Keywords: two-dimensional materials, transfer technique, van der Waals junctions, MEMS

Two-dimensional (2D) materials are highly compatible with microelectromechanical systems (MEMSs), and the latter can be used to mechanically manipulate 2D materials with micrometer-level precision. To expand the application scope of 2D material/MEMS devices, the assembly of 2D materials into van der Waals heterostructures (vdWHs) is required. As well as enhancing the quality of the 2D materials, innovative physics emerges from the assembly of 2D materials into heterostructures owing to the interactions between different 2D materials. Therefore, the combination of MEMSs and vdWHs will open up new possibilities for nanodevices, enabling the *in situ* measurement of the intrinsic properties of vdWHs. However, the transfer of vdWHs onto patterned SiO_2/Si substrates is a nontrivial task, limiting the potential applications of vdWH/MEMS devices. Therefore, a stable method for transferring vdWHs onto MEMS surfaces must be established. In this article, we review techniques for transferring vdWHs onto SiO_2/Si substrates with a view to constructing vdWH/MEMS devices. Although wet methods are commonly used, dry methods are preferable for transferring vdWHs onto MEMS surfaces owing to the fragile nature of the suspended 2D membranes and MEMS substrates. In particular, we propose polyvinyl chloride as an ideal transfer medium for vdWHs on MEMSs.

1. Introduction

Two-dimensional (2D) materials research has rapidly progressed since the discovery of graphene in 2004.[1] Graphene is a nanosheet of carbon atoms arranged in a honeycomb-like structure. Although graphene has a thickness of one atomic layer, it has outstanding mechanical, electrical, and optical properties, and for this reason, it is referred to as a "supermaterial". In addition to graphene, various types of 2D materials exist—including semiconductors,[2–5] insulators,[6–9] superconductors,[10,11] ferromagnets,[12] and topological materials[13]—and the number of 2D materials continues to grow.

One of the most fascinating aspects of 2D materials is that they can be easily combined into vertical heterostructures known as van der Waals heterostructures (vdWHs).[14] Unlike conventional compound semiconductor heterostructures such as AlGaAs/GaAs, lattice matching at the 2D material interface should not be considered in constructing vdWHs. Therefore, any

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https://doi.org/10.18494/SAM4363

type of 2D material can be stacked in any order; in other words, there is complete freedom in designing vdWH materials. At the same time, the properties of vdWHs are, in some cases, dependent on the twist angle between adjacent layers. The most well-known example of twist-angle-dependent properties is the superconductivity in magic-angle-twisted bilayer graphene.[15] Graphene exhibits superconductivity when two graphene sheets are stacked with a small twist angle of approximately 1.1°, i.e., the magic angle. Recently, twist-angle-dependent phenomena have been intensively studied.[16]

The investigation of twist-angle-dependent phenomena in vdWHs requires the precise control of the twist angle. To observe superconductivity in a structure consisting of two graphene layers, the twist angle should be exactly 1.05°. A slight deviation from 1.1° can result in a marked change in graphene band structure, obscuring the flat band necessary for the emergence of superconductivity. However, fabricating vdWHs with the complete control of the twist angle is difficult. The interlayer twist angle often deviates from the target value by 0.1–0.3°, probably because of the pressure applied during the transfer process or because the angle tends to shift toward a point of stability.[17] Because the twist angle is fixed once the device is fabricated, multiple devices must be prepared to obtain a device with an optimized twist angle. As the twist angle is a continuous quantity, a full investigation of the angle-dependent properties is practically impossible because a large number of devices are required. To explore "twistronics" in vdWHs, the continuous control of the twist angle in a single device is highly desirable. Furthermore, strain is an important parameter that determines the properties of vdWHs. Strain modulates the interlayer friction,[18] bandgap,[19,20] and interlayer coupling[21] in vdWHs. Therefore, the development of *in situ* manipulation techniques for the twist angle and strain in vdWHs is crucial.

One approach that could radically increase our chances of realizing *in situ* vdWH manipulation is to combine vdWHs and microelectromechanical systems (MEMSs). MEMS devices and 2D materials are highly compatible because MEMSs are capable of mechanically manipulating 2D materials with micrometer-scale precision.[22] In fact, many studies have been reported, in which 2D materials and MEMSs have been combined to explore the mechanical properties of the 2D materials.[18] Thus, vdWHs and MEMSs are also expected to be an excellent combination, enabling the direct measurement of the phenomena and behavior intrinsic to vdWHs that are governed by mechanical manipulation. However, there are almost no reports on combining vdWHs and MEMS in the literature. This is because transferring vdWHs onto MEMSs is very difficult owing to the delicate nature of the vdWHs and MEMS substrates.

In this article, we focus on transfer methods for vdWH/MEMS devices. In particular, we describe the advantages of a recently developed dry-transfer technique using polyvinyl chloride (PVC) to deposit vdWHs onto the fragile and rough surfaces of MEMSs.

2. From 2D Materials/MEMSs to vdWHs/MEMSs

MEMSs have been used for the mechanical manipulation of 2D materials, particularly for applying strain to 2D materials.[18] For example, strain can be manipulated by bending 2D materials on a flexible substrate,[20] by inflating 2D materials suspended over a hole,[23,24] or by increasing the width of a slit over which a 2D material is suspended (Fig. 1).[25] Strain modulation

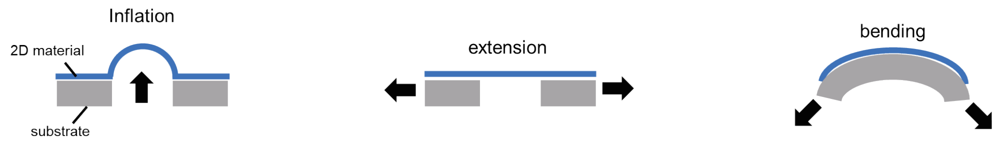

Fig. 1. (Color online) Schematics of three different strain modulation methods for 2D materials on MEMSs.

in 2D materials is important for various applications such as strain sensors and photodetectors.(18) In most previous studies of 2D materials/MEMSs, graphene has been used as the 2D material. Graphene has a high fracture toughness and excellent flexibility, and its electrical and optical properties are strongly dependent on mechanical strain. Therefore, graphene has great potential for use in resonators, strain sensors, and pressure sensors. Graphene prepared by chemical vapor deposition (CVD graphene) is often used for MEMS applications because of the large surface area of graphene sheets. To transfer CVD graphene, poly(methyl methacrylate) (PMMA) is commonly spin-coated onto it as a support for transfer (Fig. 2).(26) Then, the lower substrate (Cu, Ni) is etched, and the remaining PMMA/graphene is transferred onto an arbitrary substrate. There are two types of transfer methods for CVD graphene. The first is wet transfer, in which PMMA/graphene is floated on water and scooped up by the target substrate. The other method is dry transfer, in which PMMA/graphene is directly deposited onto the target substrate. In both cases, the PMMA layer is removed via thermal annealing. In general, dry transfer is preferable to wet transfer in terms of the quality of the transferred graphene.

Although the quality of the CVD graphene transferred by the above methods may be sufficient for simple sensor applications, it is far below the required level for state-of-the-art 2D material devices. As discussed in Introduction, 2D materials/MEMSs have great potential as a platform for studying the intrinsic properties of 2D materials. To expand the scope of application of 2D materials/MEMSs, the quality of the transferred 2D material should be improved. To achieve this, 2D materials/MEMSs should be assembled from higher-quality 2D materials. Although the growth of 2D materials by CVD results in large areas, the quality of these layers is lower than that of 2D layers exfoliated from bulk crystals. Most studies of the fundamental physics of 2D materials use 2D materials that have been mechanically exfoliated from bulk crystals using tape(6,10,15,27–30) as they are known to be of superior quality compared with materials prepared by all the other methods. Because the sizes of exfoliated 2D materials are smaller than those of CVD-grown ones (10–100 μm vs few inches), localized transfer methods must be used to manipulate them.

To further improve their quality, appropriate substrates should be selected for 2D materials. When 2D materials are directly deposited onto a silicone substrate, their quality is diminished by the surface roughness, surface phonons, and charged impurities on the SiO_2 surface.(31) To prevent this degradation, hexagonal boron nitride (h-BN) is used as an atomically flat insulating substrate.(32) The quality of 2D materials is markedly improved when capped with h-BN. To cap 2D materials with h-BN, the layers must be assembled into vdWHs by a transfer method. The advantages of vdWHs are not limited to quality improvement. There are many interesting

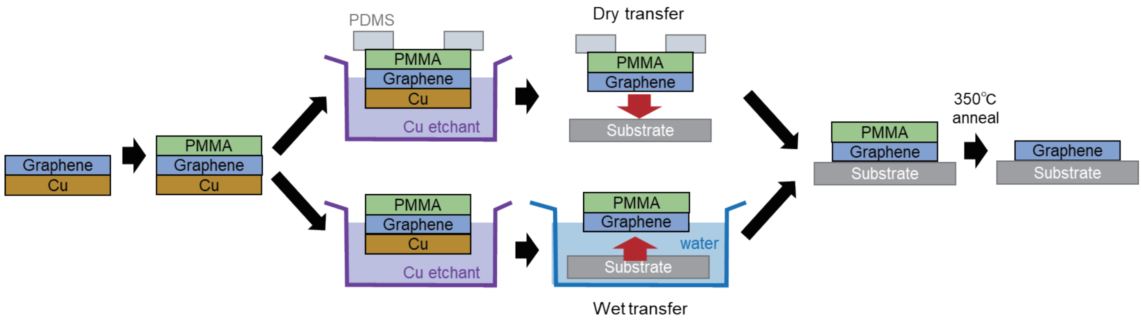

Fig. 2. (Color online) Methods for transferring CVD graphene onto arbitrary substrates.

physical phenomena that can be observed only when 2D materials are combined into vdWHs, and novel nanodevices can be realized by creating vdWHs, including vertical field-effect transistors (FETs),[33] magnetic tunnel junctions,[12] and Josephson junctions.[10] To summarize, improvements in the quality and observation of the fascinating physics of 2D materials can be realized only when these materials are in the form of vdWHs. Therefore, the use of vdWHs rather than isolated 2D materials is crucial for vdWH/MEMS applications.

By using high-quality vdWHs/MEMSs, the observation of the physical phenomena intrinsic to 2D materials, which is currently hindered by the poor quality of 2D materials/MEMSs, should become possible. Furthermore, combining vdWHs and MEMSs will realize an ideal platform for exploring the mechanics of vdWHs. As in the case of 2D materials/MEMSs, it should be possible to inflate, extend, and bend vdWHs using MEMS functionalities. In particular, it is expected that the *in situ* manipulation of the interlayer twist angle will be possible in vdWHs/MEMSs. The method currently used for investigating the twist-angle dependence of physical phenomena involves manipulation using an atomic force microscope (AFM) tip (Fig. 3).[34,35] In one study, an cross-shaped Au/graphene handle on a graphene/SiO_2 substrate was pushed and rotated using an AFM tip. The resistivity was recorded as a function of changing the twist angle and was observed to oscillate with a period of 60°. This oscillation indicated that the band structure of the two graphene sheets depended on the relative rotation angle of the sheets. Similar AFM techniques were used to study other vdWHs. This technique is innovative, but the degree of angle shift each time the AFM tip pushes the handle is not precisely controlled; it depends on the slippage between interfaces, which also depends on the twist angle. To realize the arbitrary and seamless control of the twist angle, it is preferable to fix the upper layers to a micron-sized rotation stage whose rotation angle can be controlled by applying a bias voltage. Although such a system has yet to be reported, vdWHs/MEMSs are promising candidates for their realization.

To realize vdWH/MEMS devices, the development of appropriate transfer techniques for vdWHs is a prerequisite. The transfer of vdWHs onto MEMSs is much more difficult than the conventional transfer onto SiO_2/Si because of the delicate structure of the MEMSs. The vdWHs should be transferred onto dumps, holes, and trenches of the MEMSs without breaking the vdWHs and MEMS substrates. In the following sections, we discuss transfer methods and their applicability to vdWH/MEMS devices.

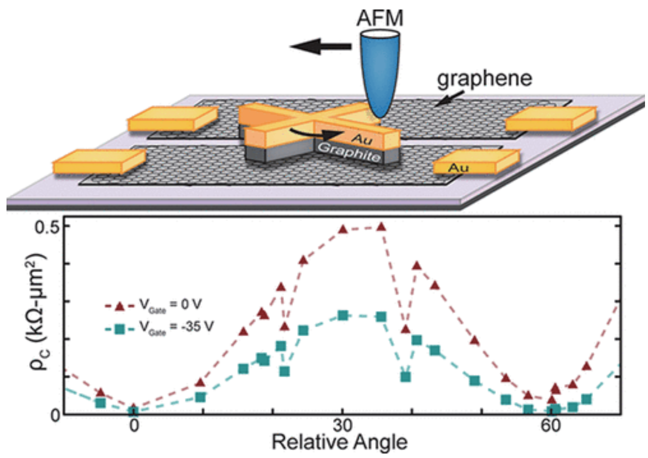

Fig. 3. (Color online) Control of interlayer twist angle between two graphene sheets using an AFM tip. Reprinted from Ref. 35 with permission.

3. Transferring vdWHs

In this section, we describe representative methods for the transfer of 2D materials for the assembly of high-quality vdWHs. First, 2D flakes must be prepared on a SiO_2/Si substrate by mechanical exfoliation using tape.(14) Mechanical exfoliation is the most basic but widely used method for preparing high-quality 2D atomic layers. 2D crystals are placed on a piece of tape, and the tape is folded and unfolded several times to cleave the crystal. The freshly cleaved surface of the crystal is attached to a SiO_2/Si substrate, and the tape is slowly removed. 2D flakes can be identified using a conventional optical microscope utilizing the optical interference at 2D material/SiO_2/Si interfaces. The SiO_2 thickness is selected to maximize the optical contrast of the 2D material. Typical thicknesses are ~300 nm for SiO_2 and ~100 nm for graphene.(36) To increase the visibility of the 2D materials on the MEMS surface, the thickness of the oxidized layer of the MEMS substrate should be considered.

Flakes of 2D materials on a SiO_2/Si substrate are picked up by sticky polymers to be assembled into vdWHs. We now describe a representative transfer method using polycarbonate (PC),(37) which is widely used to fabricate high-quality vdWHs such as superconducting twisted bilayer graphene. Figure 4 shows a schematic diagram of the transfer process using PC. (i) A thin PC layer is laid on a piece of dome-shaped poly-dimethylpolysiloxane (PDMS) on a glass slide and attached to it using tape. (ii) The PC/PDMS stamp is placed in contact with a 2D flake on a SiO_2/Si substrate at temperature T = 130 °C, which is close to the glass transition temperature of PC. (iii) The PC/PDMS stamp is detached from the substrate, and the 2D flake is picked up. (iv) By repeating this process, a vdW stack is assembled on the PC layer. (v) Next, the PC/PDMS stamp is placed in contact with another SiO_2/Si substrate at T = 180 °C, and (vi) the stack is dropped onto the substrate along with the melted PC. (vii) The PC residue is removed by immersion in acetone, and (viii) the transfer is complete. Because the stack is assembled from top to bottom, this type of transfer is called the top-down or pickup method. This top-down

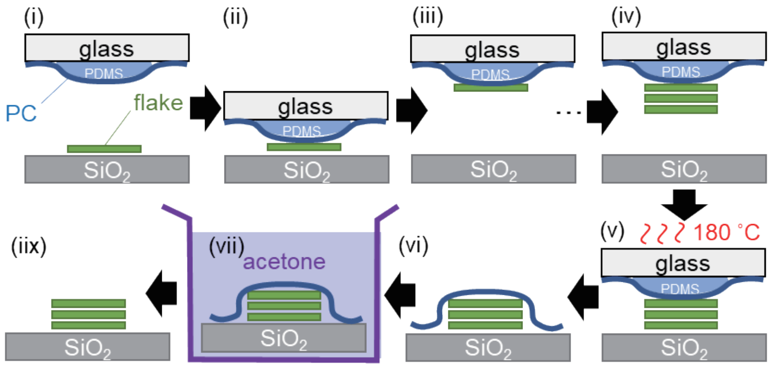

Fig. 4. (Color online) Transfer of 2D materials using PC/PDMS stamp.

transfer procedure can be conducted using different polymers such as polypropylene carbonate (PPC)[38] and PMMA.[39]

In top-down transfer, 2D flakes, except for the topmost layer, are not in direct contact with the polymer stamp. Therefore, the polymer contamination of the 2D flakes can be minimized. However, thin flakes should not be picked up directly by the polymer stamp because they are easily broken or wrinkled by the pressure applied during transfer. To transfer layers with a thickness of a few atoms, 2D materials are exfoliated directly onto a polymer sheet and then transferred to a target substrate. Figure 5 shows an example of this transfer process using PPC.[36] (i) A PPC layer is spin-coated onto a SiO_2/Si substrate. 2D flakes are exfoliated onto the PPC layer using tape. (ii) After a flake with the desired properties has been located, the PPC layer is transferred to a piece of PDMS on a glass slide and (iii) attached to the slide using tape, with the target flake positioned at the center of the PDMS. (iv, v) With 2D flake/PPC/PDMS in contact with the substrate, at the desired location, at $T = 70$ °C, the PC layer is slowly detached from the substrate, (vi) leaving the 2D flake on the substrate. If the flake is not deposited on the substrate, the PPC layer can, if desired, be dropped onto the substrate by raising the temperature to $T = 130$ °C. Because the 2D layers are stacked one layer at a time from bottom to top, this type of transfer is called the bottom-up method. The bottom-up method can also be performed by exfoliating 2D flakes onto a PDMS sheet,[40] which is then inverted over a SiO_2/Si substrate.

For both top-down and bottom-up transfers, various polymers have been used. The choice is dependent on the pick-up/release temperatures, adhesivity, and applicability to different 2D materials. Recently, a novel transfer technique using PVC has been demonstrated (Fig. 6).[41] PVC is a versatile transfer medium for 2D materials. A fascinating aspect of PVC is the fact that its adhesiveness varies enormously with temperature.[42] 2D flakes strongly adhere to PVC at $T \sim 90$ °C, whereas the adhesion is very weak at $T > 130$ °C. This marked adhesion change enables both the pick-up and release of 2D flakes using a PVC/PDMS stamp. Therefore, PVC is one of the few polymers that can be used to both pick up and release 2D flakes simply by controlling the temperature. Crucially, PVC can release 2D flakes onto a SiO_2/Si substrate at temperatures far below its melting point [Fig. 7(a)],[43] and hence it can release vdWHs without

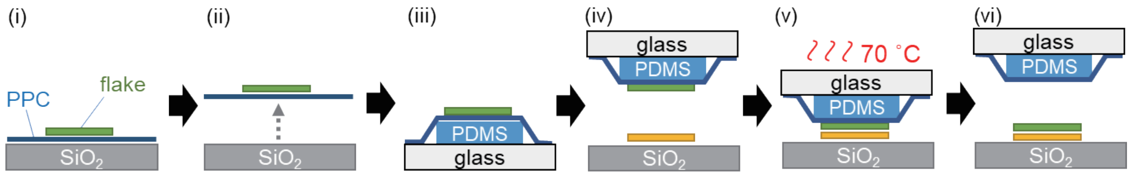

Fig. 5. (Color online) Transfer of 2D materials using PPC/PDMS stamp.

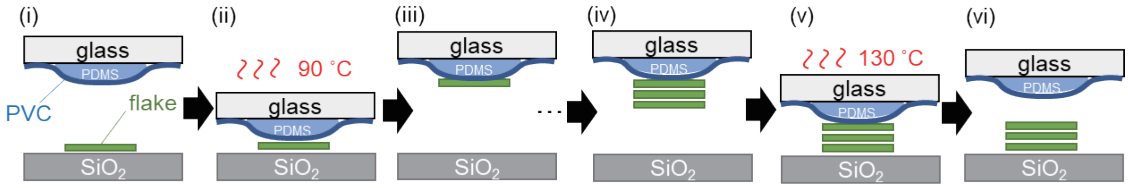

Fig. 6. (Color online) Transfer of 2D materials using PVC/PDMS stamp.

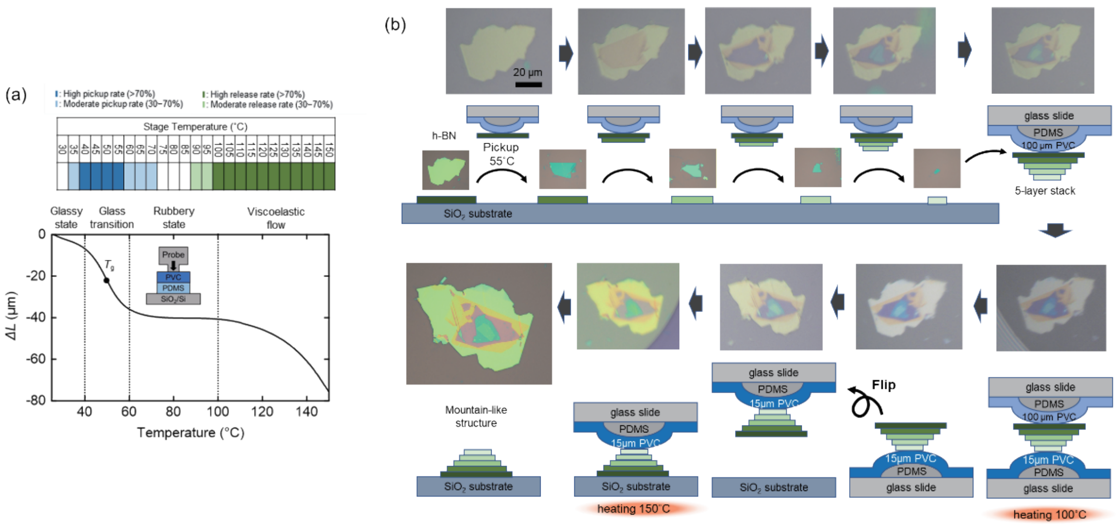

Fig. 7. (Color online) (a) Thermomechanical analysis of PVC/PDMS stamp. (b) Stacking and flipping h-BN layers using PVC/PDMS stamp. Reprinted from Ref. 43 with permission.

melting. This release of vdWHs without polymer melting is termed "dry release" because it does not require any organic solvent immersion to remove polymer residues.

As well as avoiding organic solvent immersion, dry release expands the scope of vdWH fabrication. Recently, a flip-over technique for 2D flakes has been demonstrated using PVC. In this study, the pickup and release temperatures of PVC, T_{pickup} and T_{release}, respectively, were tuned by carefully selecting the PVC film thickness; the thicker the film, the lower the T_{pickup} and T_{release} values. The difference between T_{pickup} and T_{release} was utilized to transfer 2D flakes from a thick PVC film to a thin PVC film, hence, making it possible to "flip-over" (invert) a vdWH [Fig. 7(b)].[43] This technique allows greater flexibility in the design of device structures and is especially useful for exposing a thin, fragile layer at the top of a vdWH stack.

Moreover, owing to the exceptionally strong adhesion between 2D flakes and PVC, this polymer can be used for 2D layer manipulation. For example, 2D flakes can be pulled, folded, and torn by carefully selecting and/or moving the localized contact area between PVC and 2D flakes (Fig. 8).[41] The strong adhesive forces between 2D flakes and PVC have been systematically investigated,[42] and it was revealed that PVC strongly adheres to both the surface and edges of 2D flakes. In general, the cleaved edges of 2D flakes have a higher roughness and adhere more strongly to a polymer than the flake surface. However, the strong adhesion to the flake surface is the reason for the high pick-up yield when PVC is used as the transfer material.

4. Appropriate Transfer Method for vdWHs/MEMSs

Thus far, we have discussed representative transfer methods for vdWHs. As mentioned earlier, stacking processes of vdWHs can be classified into two categories: top-down stacking and bottom-up stacking. Release methods for vdWHs can also be classified into two categories: wet release and dry release. Then, transfer methods for vdWHs can be categorized into four types, as summarized in Fig. 9: (i) top down/wet release, (ii) top down/dry release, (iii) bottom up/wet release, and (iv) bottom up/dry release.

To improve the vdWH quality, top-down stacking is preferable to bottom-up stacking because direct contact between the vdW interface and the polymer is avoided. However, in terms of vdWH transfer on MEMSs, the release process is more important than the stacking process. The difference between the wet release and dry release processes lies in whether immersion in organic solvents is required to remove the residual polymer. In wet release, vdWHs are dropped onto a substrate by melting the polymer, and the polymer residue must be removed from the 2D material on the substrate by immersion in an organic solvent such as acetone or chloroform. Wet

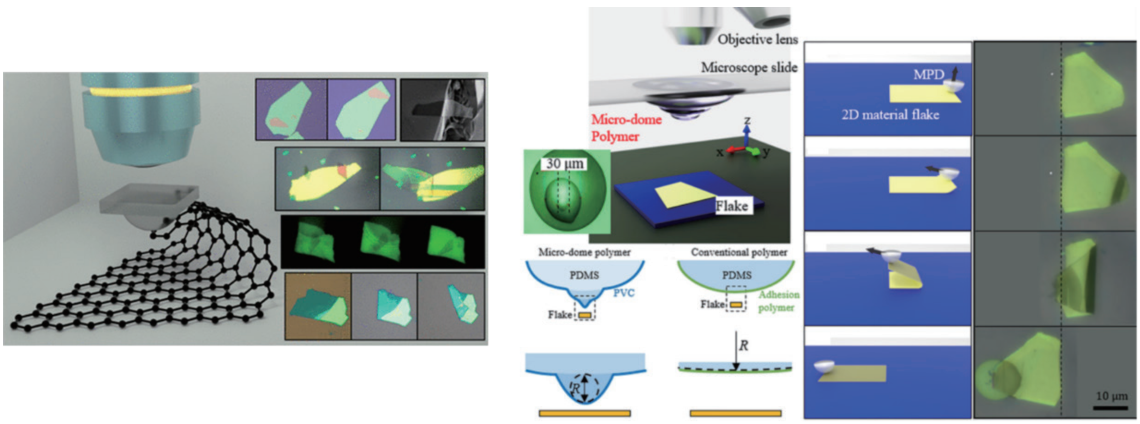

Fig. 8. (Color online) Versatile manipulation of 2D materials using PVC/PDMS stamp. To minimize the PVC contact area, a high-curvature PDMS dome is used. Reprinted from Ref. 41 with permission.

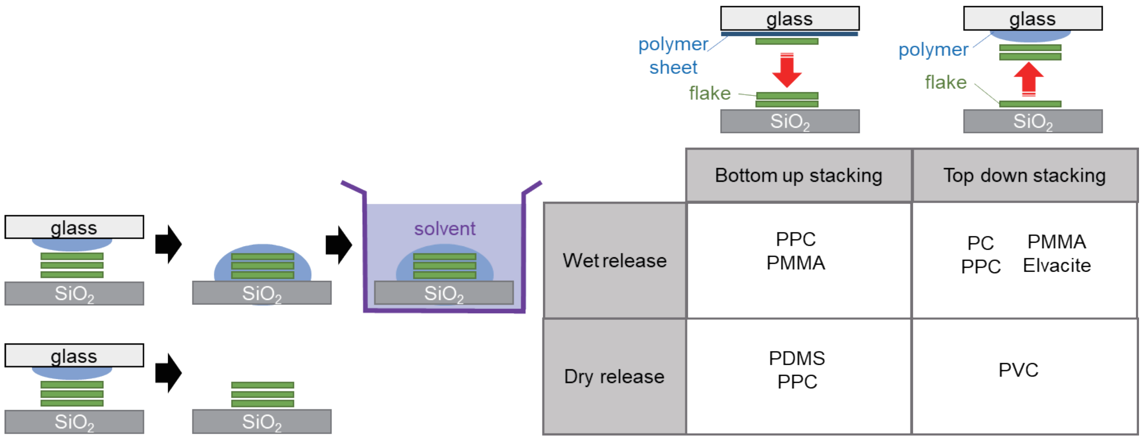

Fig. 9. (Color online) Categorization of representative polymers used for the transfer of 2D materials.

release is unsuitable for MEMS applications because suspended vdWHs are often broken or stripped away by immersion in organic solvents. In contrast, only the vdWH is released onto the substrate during dry release, bypassing the need for immersion in any liquid.

Considering the above points, the optimal transfer method for MEMSs should be top down/dry release transfer. However, top down/wet release transfer is more commonly used than top down/wet release transfer because an appropriate polymer that satisfies the requirements of both the pickup and dry release of 2D flakes has only recently been identified. For a successful dry transfer, a rise in temperature should cause the sufficient weakening of the adhesion between the polymer and 2D flakes without polymer melting. The most commonly used polymers, such as PC, PPC, and PMMA, start to melt before the adhesion to the flakes becomes sufficiently weak to release it. In contrast, the adhesiveness of PVC is significantly reduced at temperatures far below its melting temperature. Indeed, there is a difference of approximately 50 ℃ between the dry release and melting temperatures of PVC. Therefore, dry release using PVC is robust. Because the adhesiveness of PVC is highly tunable, we recommend this polymer for the transfer of vdWHs to MEMSs. Note also that PVC can be used for a wide range of vdWH manipulations, including sliding, folding, tearing, and flipping, which further expands the application scope of vdWH/MEMS.

5. Conclusion

We reviewed the current situation in the field of 2D materials/MEMS devices and fabrication techniques of vdWHs. We also discussed future possibilities of vdWH/MEMS devices and techniques for transferring vdWHs to MEMSs. The top down/dry release transfer method is suitable for vdWH/MEMS devices as it prevents polymer contamination and mechanical damage to fragile vdWHs and MEMS substrates during transfer. Considering its excellent adhesion tunability and high dry release rate, we recommend PVC as an ideal transfer medium for constructing vdWH/MEMS devices.

Acknowledgments

This work was supported by JST-CREST and JST-Mirai (Grant Numbers JPMJCR15F3, JPMJCR20B4, and JPMJMI21G9); JSPS KAKENHI (Grant Numbers JP20H00127, JP21H05232, JP21H05234, JP21K20345, and JP22K14559); Tokuyama Science Foundation and Support Center for Advanced Telecommunications Technology Research Foundation.

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