

Toward Clean 2D Materials and Devices: Recent Progress in Transfer and Cleaning Methods

Wenlong Dong, Zhaohe Dai,* Luqi Liu,* and Zhong Zhang

Two-dimensional (2D) materials have tremendous potential to revolutionize the field of electronics and photonics. Unlocking such potential, however, is hampered by the presence of contaminants that usually impede the performance of 2D materials in devices. This perspective provides an overview of recent efforts to develop clean 2D materials and devices. It begins by discussing conventional and recently developed wet and dry transfer techniques and their effectiveness in maintaining material “cleanliness”. Multi-scale methodologies for assessing the cleanliness of 2D material surfaces and interfaces are then reviewed. Finally, recent advances in passive and active cleaning strategies are presented, including the unique self-cleaning mechanism, thermal annealing, and mechanical treatment that rely on self-cleaning in essence. The crucial role of interface wetting in these methods is emphasized, and it is hoped that this understanding can inspire further extension and innovation of efficient transfer and cleaning of 2D materials for practical applications.

properties make them highly attractive for a variety of electronics and optoelectronics applications, including semiconductor lasers, light-emitting diodes, and transistors.^[1,2,7,8] However, the practical use of 2D materials is often limited by the presence of contaminants that arise during the fabrication process or transfer process. For example, while the room-temperature mobility of monolayer graphene on a SiO₂ substrate is $\approx 40\,000\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ in theory,^[9] empirical measurements of transferred monolayers have yielded remarkably varied values ranging from ≈ 1140 to $\approx 48\,820\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$,^[10–17] often attributed to the influence of contaminants.^[10,13] While this is a serious problem even for a single layer, it is exacerbated when more layers are stacked into heterostructures, which have unlocked an array of properties and unprecedented synergistic effects. A concrete example can be

given by measured carrier mobilities from MoS₂/graphene/hBN devices ($\approx 60\,000\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$) and mica/graphene/hBN devices ($\approx 1000\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$), with the marked distinction stemming from the absence of self-cleaning mechanism at the graphene-mica interface.^[18] Therefore, an upsurge of research endeavors has been channeled into refining fabrication processes to preserve the cleanliness of 2D materials, scrutinizing their cleanliness, as well as exploring post-treatments capable of improving the cleanliness of 2D materials and their devices over the past decade.

Recent years have seen particular progress in the development of clean 2D materials and devices, and this perspective provides an overview of these advancements. Specifically, we discuss various transfer techniques that have been optimized or developed recently, multi-scale methodologies for assessing the cleanliness of 2D material surfaces and interfaces, and recent advances in passive and active cleaning strategies. It is important to note that this perspective is not meant to be an exhaustive overview of the broad field of 2D materials and devices. Rather, we emphasize the importance of understanding the interface phenomena of 2D materials, particularly the interfacial wetting behavior, in the design and selection of particular transfer and cleaning strategies. The goal is to inspire further innovation and extension of current methodologies towards clean 2D materials and devices and highlight the significance of continued efforts in this area to unlock the full potential of 2D materials in electronics and optoelectronics. Readers are encouraged to explore relevant reviews from different perspectives, which include bottleneck challenges

1. Introduction

Two-dimensional (2D) materials have gained tremendous attention in recent years due to a combination of exceptional electronic,^[1,2] optical,^[3,4] and mechanical^[5,6] properties. These

W. Dong, L. Liu
CAS Key Laboratory of Nanosystem and Hierarchical Fabrication and
CAS Center for Excellence in Nanoscience
National Center for Nanoscience and Technology
Beijing 100190, China
E-mail: liulq@nanoctr.cn

W. Dong
University of Chinese Academy of Sciences
Beijing 100049, China

Z. Dai
Department of Mechanics and Engineering Science
State Key Laboratory for Turbulence and Complex Systems
College of Engineering
Peking University
Beijing 100871, China
E-mail: daizh@pku.edu.cn

Z. Zhang
CAS Key Laboratory of Mechanical Behavior and Design of Materials
Department of Modern Mechanics
University of Science and Technology of China
Hefei 230027, China

 The ORCID identification number(s) for the author(s) of this article can be found under <https://doi.org/10.1002/adma.202303014>

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in 2D material-based electronics,^[7] large-scale synthesis of 2D materials,^[19] techniques for cleaning 2D material surfaces,^[20] deterministic transfer for 2D material assemblies,^[21] methods for producing van der Waals heterostructures,^[22–24] as well as some applications that utilize contaminated 2D materials.^[25–28]

2. Transfer Methods

Over the past decade, significant progress has been made in the development of various approaches for the fabrication of 2D materials, including mechanical exfoliation,^[29,30] liquid exfoliation,^[31,32] gas vapor growth,^[33,34] and chemical synthesis.^[35] The fabrication of 2D material-based devices, however, still requires transferring 2D materials from one substrate to another. The most common transfer methods can be divided into two main categories: wet and dry transfer, depending on whether liquids are used for layer-layer detachment during the process. In this section, we provide a brief overview of conventional transfer methods and some recently developed wet and dry transfer methods, with particular attention given to the cleanliness of these methods. It is important to note that each of the methods discussed below has its own set of advantages and disadvantages. Moreover, it is possible to use a combination of steps from different methods to achieve specific purposes.

2.1. Wet Transfer Methods

2.1.1. PMMA Layer-Assisted Transfer Method

The polymethyl methacrylate (PMMA) layer-assisted transfer has emerged as a prevalent method for transferring graphene and other 2D materials grown by chemical vapor deposition (CVD).^[36,37] Meanwhile, a slightly different version of this technique is also widely used for the transfer of exfoliated 2D materials.^[22,38] Here, we illustrate the main steps of the PMMA layer-assisted transfer method for the CVD case in **Figure 1a**. First, a thin PMMA layer is spin-coated onto the growth substrate. Next, the PMMA/graphene layer is detached from the pristine substrate by etching the surface of the substrate with etchants and washed with deionized water. The PMMA/graphene layer is then transferred onto the target substrate, and finally, the PMMA layer is removed using acetone treatment or high-temperature annealing, without little loss of the transferred 2D materials.

The PMMA layer has both benefits and drawbacks when it comes to protecting 2D materials: While it shields them from damage, it also leaves behind residues that can reduce the carrier mobility of graphene.^[10,13] Thus, several variants of this wet transfer method are developed to reduce contaminants. 1) One approach is to use alternative polymer layers that are soluble in organic solvents, such as paraffin,^[39] cellulose acetate (CA),^[40] polystyrene (PS),^[41] pentacene (C₂₂H₁₄),^[42] small molecule (naphthalene or camphor),^[43] rosin (C₁₉H₂₉COOH),^[44,45] or even Nail Polish.^[46] For example, the use of paraffin can lead to clean and wrinkle-reduced large-area graphene with highly uniform sheet resistance.^[39] However, these layers tend to have reduced mechanical strength, so a double-layer design

may be necessary to maintain the integrity of the transferred 2D materials.^[47] 2) Another approach is to use the electrochemically bubbling delamination method (Figure 1a).^[48] This makes the process less susceptible to etchants, but the substrate limitation and the tradeoff between the delamination rate and structural integrity of 2D materials still lock the coverage of the technique. New strategies, such as ultrasonication delamination^[49] and strain engineering,^[50] have been developed to address the challenge. 3) Similarly, water-soluble layers are used as sacrificed substrates to avoid the etching process.^[51,52] 4) Additionally, several post-treatments can help reduce PMMA residues,^[20] such as thermal annealing treatment (particularly the air or O₂ annealing),^[53] plasma, ultraviolet, or ozone treatment,^[54] electrochemical treatment,^[55] current-induced annealing treatment,^[56,57] modified Radio Corporation of America (RCA) clean method,^[58] and more.

2.1.2. Liquid Layer-Assisted Transfer Method

We refer to the wedging transfer method,^[59] surface energy-assisted transfer method,^[60] and capillary force-assisted transfer method^[14,61–63] available in the literature as liquid layer-assisted transfer method here. This method relies on the completely wetting property of the substrate-2D material interface, which is characterized by the spreading parameter $S = \gamma_{fs} - \gamma_{sl} - \gamma_{fl} > 0$,^[64] where γ_{ij} represents interface energy density of the *i*-*j* interface, *s*, *f* and *l* denote substrate, 2D material, and liquid, respectively. In this scenario, the liquid tends to enter the substrate-2D materials interface energetically, leading to spontaneous detachment. Therefore, liquid layer-assisted transfer is particularly useful for systems of high surface-energy liquid, hydrophilic substrate, and hydrophobic polymer film which are more likely to have a positive spreading parameter. One of the existing challenges in liquid layer-assisted transfer methods is effectively protecting the 2D materials from damage caused by the surface tension of the liquid used. There are different ways to perform such protection, and in Figure 1b, we schematically depict the method using a PDMS stamp. Initially, the surface of PDMS comes into contact with the 2D materials. Subsequently, a droplet is introduced to infiltrate the interface between the 2D materials and hydrophilic substrate, which leads to the delamination and pick-up of 2D materials onto PDMS. Finally, by leveraging the low surface energy of PDMS, 2D materials can be readily released onto the desired substrate.

Liquid layer-assisted transfer is a powerful etching-free method for transferring both CVD-grown and mechanically exfoliated 2D materials.^[60,62] However, it is subject to drawbacks such as creating extensive wrinkles and dramatic cracks in 2D materials, as well as bubbles at the interface between the 2D material and its target substrate.^[11] To address these issues, efforts have been made to find more reliable supporting layers such as PS,^[60] poly L-lactic acid (PLLA),^[65] polycarbonate (PC),^[66] and more. Modification of the wetting properties of the target substrate is another solution: Hydrophobic surfaces modified by self-assembled monolayers (SAMs) hinder the insertion of the water, resulting in wrinkle-less and crack-less transfer.^[67,68] In addition, the wettability of the transfer medium is a key factor: Kim et al.^[69] and Shen et al.^[70] demonstrated that the density of wrinkles is

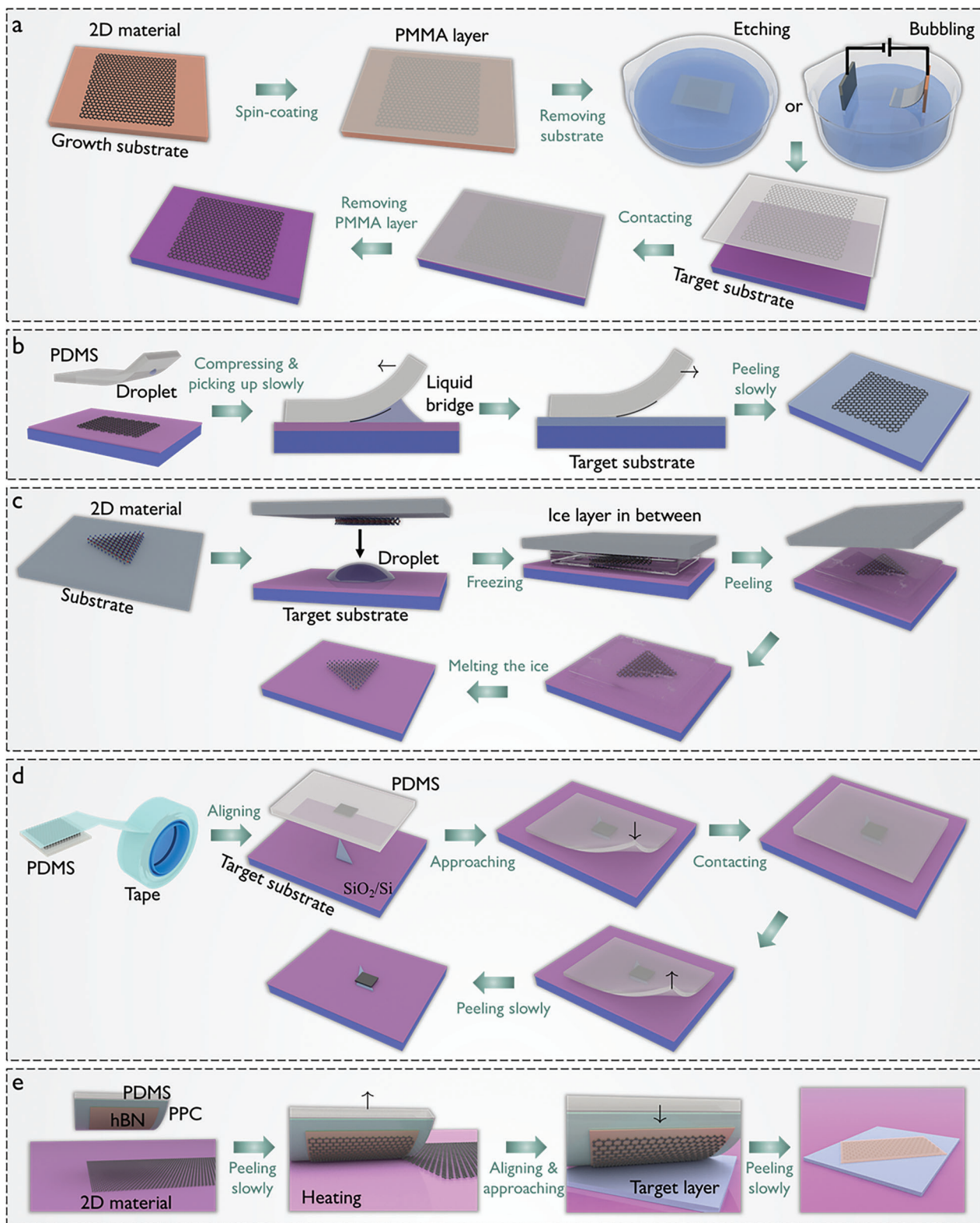


Figure 1. Schematic illustration for main steps in the wet (a-c) and dry (d,e) transfer techniques. a) PMMA layer-assisted transfer.^[36,37,48] b) Liquid layer-assisted transfer.^[14,61–63] c) Ice layer-assisted transfer.^[71] d) Viscoelastic dry transfer.^[72] e) Van der Waals pick-up.^[85]

mostly dependent on the transfer medium and a liquid droplet with low surface tension and ultra-strong wettability on the target substrate can minimize wrinkles and cracks.

2.1.3. Ice Layer-Assisted Transfer Method

Though liquid layer-assisted transfer is often considered a relatively “clean” method for transferring 2D materials, it still introduces trace contaminants to the surface due to the required contact between 2D materials and their protecting polymer layer. Furthermore, this method is limited to systems with positive spreading parameters. Very recently, an ice layer-assisted transfer method has been developed^[71] that not only eliminates the need for polymer layers and organic solvents in the process but also works for a broader range of systems, including those with negative spreading parameters. Figure 1c schematically illustrates the main steps of the ice layer-assisted transfer process. To begin, a water droplet is placed on the target substrate. Approaching the initial substrate to the target substrate can squeeze the droplet into a ≈ 100 -micrometer thick liquid layer, which can then be frozen to create an ice layer between the two substrates. After the initial substrate is peeled off, the ice layer remains on the target substrate, and any interfacial strain caused by the inhomogeneous interfaces between the substrate, 2D material, and ice is relaxed. Finally, the ice layer is melted to complete the transfer process.

There are several interesting facts about the reported ice layer-assisted transfer process. First, the ice layer can be used as a stamp for transferring 2D materials to the next substrate. Second, it has been found that a high initial water temperature (60 °C) and a low freezing temperature (−20 °C) are critical factors for successful transfer. Third, at a relatively higher temperature (the exact temperature depending on the specific sample and substrate), an active surface cleaning mechanism occurs during the peeling of the ice stamp, which can remove some of the contaminants initially absorbed on the 2D material surface and leave the 2D material on its substrate intact. While the usefulness of this relatively new method needs to be examined over time, there are concerns regarding interfacial bubbles since it involves the process of ice melting and thin film dewetting, which brings the 2D material into contact with the target substrate.

2.2. Dry Transfer Methods

2.2.1. Viscoelastic Dry Transfer Method

In contrast to wet transfer methods discussed in the previous subsection, Castellanos-Gomez et al. have developed a dry transfer method that eliminates the use of liquids and instead employs viscoelastic stamps, such as polydimethylsiloxane (PDMS).^[72] This technique provides inherent advantages in transfer speed and cleanliness. Figure 1d illustrates the key steps involved in this dry transfer method. Similar to the “Scotch tape” method, 2D materials can be directly exfoliated onto the surface of the PDMS stamp. Then the stamp is pressed onto the desired location on the target substrate, and the PDMS is slowly peeled off, leaving the 2D flake on the top of other 2D materials on the target sub-

strate. The use of PDMS or other viscoelastic polymers for transfer methods can facilitate the rapid preparation of van der Waals heterostructures, but it also presents several challenges. First, direct contact between 2D materials and the polymer can lead to trace contaminants. When these contaminants are trapped at the 2D material interfaces and form bubbles, significant efforts are required to clean them up (this will be discussed in more detail later on). To mitigate this issue, pretreating the PDMS with UV-ozone cleaning has been reported to effectively reduce organic residues.^[73] Second, PDMS fails to pick up 2D materials on growth substrates due to low adhesion energy to 2D materials, which limits further applications. Thus, polymers with strong adhesion such as epoxy^[74] and photocurable resins^[75] are used to ensure the successful pick-up of 2D materials, but this may result in cracks or wrinkles on the flake. Possible solutions to this issue include using soft gels with good stress relaxation characteristics as a lamination layer^[16] and using a chemically modified polymer with tunable adhesion.^[12,76] In addition, different adhesion requirements of polymers during the pick-up and release process can pose a dilemma. To address this issue, the adhesion between 2D materials and polymers, such as PMMA,^[77] polyvinyl alcohol (PVA),^[78,79] polypropylene carbonate (PPC),^[80,81] and more, has been found tunable via a heating/cooling system. Based on the thermal adhesion strategy, it is even possible to transfer and stack 2D materials in a vacuum to avoid airborne contaminants.^[82] Furthermore, polymer handles with microdome-shaped structures can be used to manipulate 2D flakes such as sliding, rotating, folding, flipping, and exfoliating owing to their controlled contact area.^[83,84]

2.2.2. Van der Waals Pick-up Method

To achieve even higher sample cleanliness, Wang et al. developed an alternative assembly technique for van der Waals heterostructures using hexagonal boron nitride (hBN) as the pick-up stamp.^[85] This technique involves a glass/PDMS/PPC structure adhering to the hBN stamp (Figure 1e). First, a flat PPC layer is spin-coated on a SiO₂/Si substrate. Then, two methods can be used to obtain suitable hBN flakes on the PPC layer: 1) exfoliating hBN flakes onto the surface of the PPC, which is then transferred onto the PDMS stamp, or 2) transferring the PPC layer onto the PDMS surface first and then picking up a suitable hBN flake at 40 °C. Due to the good transparency of PDMS and PPC, the target 2D material can be aligned using an optical microscope and picked up from its pristine substrate by the hBN stamp. It is worth noting that a high temperature is usually required to enhance the van der Waals force between hBN and the target 2D material if the substrate has been treated with plasma.^[86] The atomically smooth surface of the hBN layer enables 2D materials to be transferred without the use of polymers, thereby avoiding polymer residues that are inevitable in other methods.

The hBN pick-up method has a primary advantage in ensuring that the inner layers of the heterostructure remain free from contact with polymers or solvents, resulting in highly clean and homogeneous interfaces. Furthermore, despite the challenge of removing the hBN layer after the process, it can be utilized as a substrate or encapsulation layer owing to its chemical inertness and ability to reduce the scattering effect on carriers compared to

SiO₂/Si.^[85] Nonetheless, this advantage must be weighed against the increased complexity and reduced fabrication yield of the hBN method, relative to the other simpler methods such as the viscoelastic dry transfer method. On the other hand, recent studies have demonstrated that certain metals exhibit highly robust interactions with 2D crystals, enabling them to serve as an adhesion layer while also facilitating the production of high-quality and scalable 2D flakes.^[87,88] For instance, Moon et al. demonstrated that graphene with varying layers can be exfoliated in a scalable manner by depositing corresponding metals onto the graphite.^[89] Huang et al. developed a gold-assisted mechanical exfoliation technique that effectively yielded 40 distinct single-crystalline monolayers.^[90] By depositing metals on the thermal release tape, large monolayer 2D materials can be exfoliated onto the tape or peeled from the substrate by the tape and then transferred to target substrates.^[91–94] Similarly, Li et al. designed an Au-mesh film as a peeling tape to restrict the contact between MoS₂ crystal and Au, where the noncontact region guarantees that the monolayers can be mechanically released without additional treatments.^[95] Unfortunately, due to the excessively strong adhesion between the 2D material and metal, the successful transfer of the 2D material onto a desired substrate may necessitate the functionalization of the substrate to enhance its adhesion to the 2D material.

2.3. Controlling the Interlayer Twist

We complete this section by briefly discussing how to implement the method described above to regulate the twist angle between individual layers of 2D materials in a van der Waals structure (Figure 2). The moiré patterns between these layers can manifest with characteristic sizes varying from a few nanometers to several microns, depending on the twist angle.^[96] The properties of van der Waals structures are tightly linked to the moiré patterns they exhibit, thereby prompting significant efforts to manipulate the twist angle.^[97]

In this regard, the “tear-and-stack” method is now widely used in the preparation of twisted 2D material layers, as illustrated in Figure 2a. Similar to the van der Waals pick-up transfer method, Kim et al. used a hemispherical PMDS or epoxy handle with a spin-coated polymer to lift the hBN layer on the SiO₂/Si substrate (Figure 2a, middle panel).^[98] This hBN layer can then tear graphene into various pieces with the same crystal direction and is subsequently employed to vertically stack graphene pieces with a controlled twist angle. Note that the graphene layer can be sectioned before the transfer process by electron beam lithography or AFM tip to improve the controllability of the twist angles. Alternatively, the bottom panel in Figure 2a depicts the wet PMMA layer-assisted transfer method used to fabricate twist-controlled bilayer and trilayer graphene.^[99] Aside from the transfer steps, a distinct feature of this method is the use of femtosecond laser micromachining to cut graphene/PMMA layer into two or three pieces with straight and parallel edges, thus enabling precise control over the cutting lines.

Several methods for folding or unfolding 2D materials might be employed to produce stacked layers with controlled twist angles.^[100–102] For example, as illustrated in Figure 2b, Wang et al. used the liquid layer-assisted method and modified sub-

strate with a semi-hydrophilic and semi-hydrophobic surface to fabricate bilayer graphene with different twist angles.^[100] Similarly, Zhao et al. showed defined folding regions by modifying the PVA substrate with the electron beam, resulting in arrays of folded patterns.^[101] Additionally, a tiny folded area can be created by precisely manipulating a graphene sheet on a graphite surface at low temperatures using a scanning tunneling microscope (STM) tip^[103] or by using nanoimprint-style contact techniques to nucleate and direct the self-folding of graphene ribbons.^[104]

3. How Clean is Clean?

In the previous section, we discussed various transfer methods and advanced strategies for minimizing contaminants. However, even van der Waals heterostructure devices constructed by the cleanest van der Waals pick-up method perform below theoretical expectations, suggesting that there are possible contaminants present at the surfaces and interfaces of 2D materials. In this section, we will briefly explore the possible sources of contaminants and methods for assessing cleanliness (Figure 3). Note that different perceptions of cleanliness can vary depending on the scale of observation, and thus interface cleanliness is a relative concept that depends on the size of samples and scale of observation.

3.1. Source of the Contaminants

Contaminants are inevitably introduced into material surfaces due to exposure to environments.^[18,105] In addition, trace contaminants can be trapped at the interfaces by attractive van der Waals forces between the deposited flake and the target substrate, resulting in either uniform layers of contaminants^[18,106] or topographically detectable bubbles of contaminants (which will be further discussed in the next section).^[18,64,107,108]

On the one hand, water and hydrocarbons in ambient and sample storage environments can readily adsorb onto the surface of 2D materials.^[25] 1) Airborne hydrocarbons of an organic nature are commonly found on surfaces of both exfoliated and CVD-grown 2D materials.^[109–111] More recently, Tilmann et al. have highlighted the formation of thin polymeric adlayers, likely composed of hydrocarbons, on freshly exfoliated MoS₂ samples, even though they are not in direct contact with the adhesive tape used for the mechanical exfoliation.^[109] Layers of hydrocarbon contaminants in the order of a few Ångström rapidly form on the surfaces of 2D materials after tens of minutes of exposure to air.^[112–115] The total surface energy of 2D materials decreases due to the contaminant by hydrocarbons upon exposure to air, causing hydrophilic 2D material surfaces to become hydrophobic.^[112–115] 2) Surfaces possessing hydrophilic characteristics, exhibit an inherent affinity for forming spontaneous water adlayers.^[116] For example, nanodroplets of water, ranging from 10 to 100 nm in radius, can be adsorbed onto graphene surfaces while both the density and radii of these droplets can be reduced in conditions of low relative humidity (RH).^[117] Indeed, the effective mechanical exfoliation of monolayer graphene needs to avoid too much humidity within the laboratory environment, which could potentially cause condensation.^[118] Notably, the surface adsorption between hydrocarbons and water is a competition. It was found that the presence of water can slow the

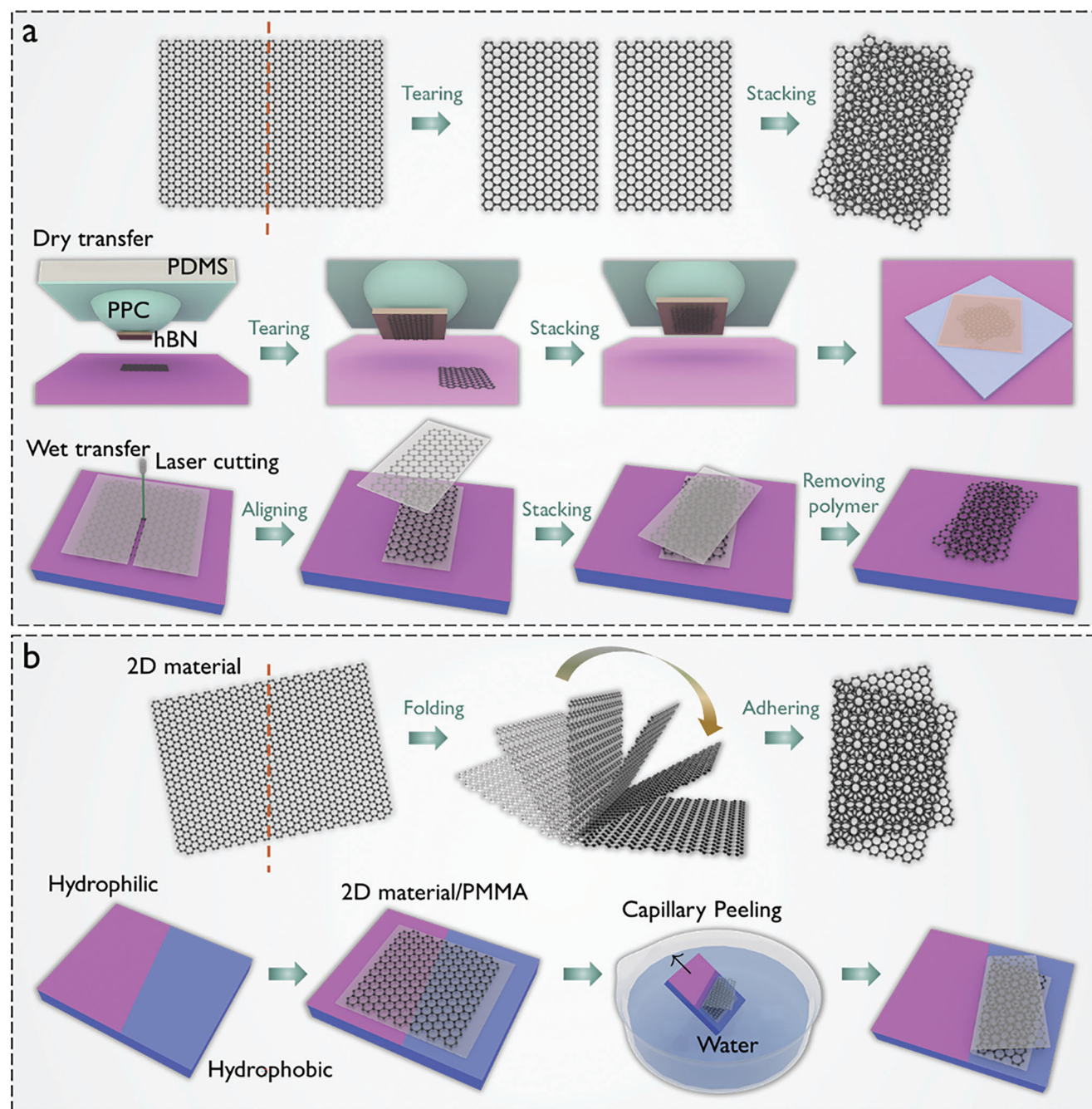


Figure 2. Schematic illustration for the mechanisms and main steps in preparing twisted 2D materials. a) Tear/cut and stack method.^[98,99] b) Controlled folding method.^[100]

adsorption of the hydrocarbons.^[119] 3) Adsorption of airborne gas molecules is another tricky issue, especially for oxygen. Oxygen is a typical p-type doping agent for 2D materials, leading to Fermi-level shifts of 2D materials.^[120–122] Besides, oxygen usually acts synergistically with water to cause hysteresis in field effect transistors.^[123–126] Prolonged contact with oxygen and water molecules, 2D materials would be subject to chemical reactions which can ultimately lead to degradation. For highly sensitive materials that are even isolated from the air environment by hBN en-

capsulation, the degradation cannot be prevented, demonstrating the presence of oxygen at the interface.^[127]

On the other hand, while conducting the exfoliation and transfer under vacuum can diminish the origin of ambient contaminants, the introduction of residues during the transfer process remains a potential source of contaminants. 1) In procedures employing wet transfer methods discussed above, there exists the possibility of liquid entrapment at the interface between the 2D material and the substrate.^[64,108] Interestingly,

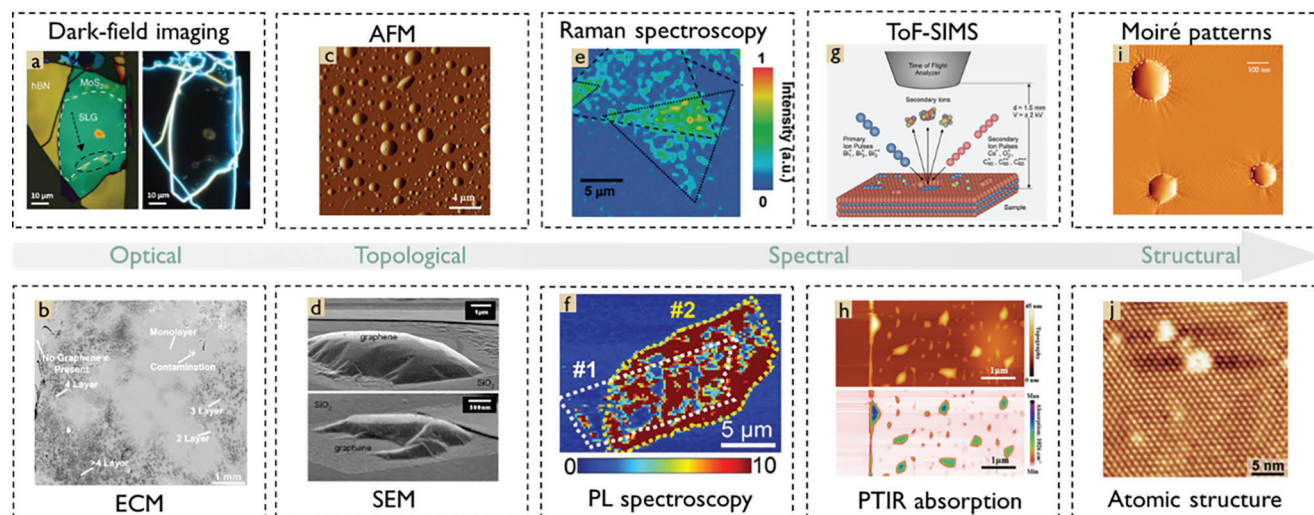


Figure 3. Summary of 2D material cleanliness assessment techniques. The optical imaging methods include a) optical dark-field imaging and b) ECM. a) Reproduced under terms of the CC-BY license.^[132] Copyright 2018, published by Springer Nature. b) Reproduced with permission.^[133] Copyright 2018, American Chemical Society. The topological characterizations include c) AFM and d) SEM. c) Reproduced with permission.^[108] Copyright 2022, Wiley-VCH. d) Reproduced with permission.^[105] Copyright 2009, American Chemical Society. Spectral characterizations include e) Raman spectroscopy, f) photoluminescence spectroscopy, g) ToF-SIMS, and h) PTIR. e) Reproduced with permission.^[139] Copyright 2014, American Chemical Society. f) Reproduced with permission.^[106] Copyright 2020, American Chemical Society. g) Reproduced under terms of the CC-BY license.^[131] Copyright 2015, published by Springer Nature. h) Reproduced with permission.^[129] Copyright 2019, American Chemical Society. Structural imaging methods include i) moiré patterns of twisted bilayer 2D materials and j) atom-level structural imaging. i) Reproduced under terms of the CC-BY license.^[147] Copyright 2021, published by Springer Nature. j) Reproduced with permission.^[149] Copyright 2018, Wiley-VCH.

certain applications, such as liquid cell microscopy, have deliberately harnessed the presence of this trapped liquid introduced through such means.^[128] 2) A significant proportion of the contaminants stem from remnants of supporting polymer layers, particularly noticeable when employing techniques like the PMMA layer-assisted transfer method and viscoelastic dry transfer method—methods that allow direct contact between the 2D materials and polymers.^[66,109] Even for the van der Waals pick-up method that can ensure noncontact between 2D materials and polymers, trace of PC contaminants was still detected at the interfaces through techniques such as photothermal induced resonance using the photothermal induced resonance (PTIR) technique^[129] and cathodoluminescence (CL) spectroscopy.^[130] 3) The etchant used in the substrate etching and polymer removal procedures often persists on the surface of the materials. For example, Zhou et al. analyzed the planar chemical composition of 2D heterostructures and identified residual $(\text{NH}_4)_2\text{S}_2\text{O}_8$ used for copper foil etching and $\text{C}_2\text{H}_6\text{O}$ used for the PMMA removal.^[131] 4) When using the PMMA layer-assisted transfer method, the presence of substrate residues (such as copper foil) as potential contaminants cannot be disregarded.^[131]

3.2. Assessment of Cleanliness

It is often necessary to use the optical microscope and related techniques, such as optical dark-field imaging^[132] and ellipsometric contrast micrography (ECM),^[133] to quickly screen for possible surface and interfacial residues (Figure 3a, b). Additionally, assessing the cleanliness of 2D materials often involves checking for the vertical undulation of 2D materials using atomic force mi-

croscopy (AFM)^[108] or scanning electron microscope (SEM)^[105] (Figure 3c,d). However, it is important to note that a flat surface does not necessarily correspond to a clean interface. For instance, in systems with a positive spreading number, contaminants may appear as a thin, uniform layer instead of bubbles at the interface.^[106] Spectroscopy is a well-established method for characterizing materials, providing valuable insights into the cleanliness of 2D materials (Figure 3e-h). For example, Raman and photoluminescence (PL) spectroscopy can detect changes in the signal that reveal information on the charge doping,^[134,135] strain,^[134–136] defects,^[137,138] and interlayer coupling,^[139,140] allowing for indirect evaluation of material quality and cleanliness. Besides, contaminants themselves can induce strong signals in Raman spectroscopy,^[66] PL spectroscopy,^[140] CL spectroscopy,^[130] energy dispersive X-ray spectroscopy (EDX) spectroscopy,^[141] X-ray photoelectron spectroscopy (XPS),^[10,142] Auger electron spectroscopy (AES),^[143,144] ultraviolet photoelectron spectroscopy (UPS),^[145] time-of-flight secondary ion mass spectrometry (ToF-SIMS),^[76,109,131] and infrared (IR).^[114,129] Note that ToF-SIMS has been proven exceptionally effective in the detection of atomically thin layers of contaminants, which have been elusive when using common methods, such as Raman spectroscopy, XPS, or SEM.^[109]

After these conventional characterizations, the overall cleanliness of 2D materials can then be inferred from the performance of 2D material-based devices,^[18] since contaminants can have a detrimental impact on the functional performance of 2D materials as they can introduce doping, strain, and charge scattering effects.^[10,13,18,146] In case transferred samples that are considered clean may not exhibit electrical properties comparable to high-quality exfoliated 2D material, the cleanliness should be assessed

at a smaller scale. In the case of bilayer samples, moiré patterns can emerge when a slight twist angle is introduced between two layers due to their different periodicity, which can be used to indicate the atomic cleanliness of the interface.^[147,148] For example, distorted moiré patterns near interfacial bubbles and their disappearance inside provide a visual tool for observing the quality of the interface (see Figure 3i).^[147] Moreover, atomic resolution images can be obtained through STM and transmission electron microscopy (TEM), allowing for the assessment of sample quality at the sub-nano scale (Figure 3j).^[96,149,150]

4. Cleaning Methods

After discussing various transfer methods, the source of contaminants, and the characterization method for the cleanliness of 2D materials, we are finally in a position to examine the existing methods for passively and actively cleaning contaminated 2D materials. In fact, in the section on transfer methods, we have discussed a number of details regarding how to gently or aggressively remove polymer residues from 2D material surfaces. A comprehensive discussion on this topic can be found in a previous review,^[20] where six major treatments for cleaning 2D material surfaces are summarized. These treatments include thermal annealing treatment, plasma and ion beam treatment, electron beam treatment, light treatment, mechanical treatments, and chemical methods. However, when the cleaning target is the 2D material interfaces or devices, the contaminants are often covered by either protective layers or the 2D material layers themselves, making surface cleaning techniques ineffective. Therefore, we will focus on cleaning techniques for 2D material interfaces in the remainder of this section.

4.1. Interfacial Bubbles and Self-Cleaning

4.1.1. The Formation Mechanism

The integration of 2D materials into practical devices involves creating vast interfaces between different materials, which are highly susceptible to contaminants. Earlier studies have revealed that interfacial contaminants can be pushed and clumped together into bubbles or pockets (Figure 4a).^[141,151] The occurrence of bubbles has been attributed to that the affinity between the two materials is stronger than the affinity between the material and the trapped contaminants.^[152,153] More specifically, this interfacial process takes place at interfaces characterized by a negative interfacial spreading parameter ($S < 0$), bearing resemblance to the dewetting of liquid films on a surface.^[64,154] In other words, when $S < 0$, it is energetically favorable for the two materials to create a common interface as large as possible while compensating for some elastic strain energy to form bubbles that are used to contain the squeezed contaminants.^[64,105,107,108]

While bubbles of contaminants may initially appear as a nuisance, they have garnered significant attention due to their intriguing ability to reveal the adhesion of 2D materials^[155] as well as to cause large mechanical strain that can modulate a wide range of the material properties (a phenomenon known as strain engineering).^[25–28,156] Such ability has spurred research to understand the mechanical behavior of 2D material bubbles. Khestanova et al.^[107] first reported on the self-similar shape of

2D material bubbles, and subsequent studies have observed an almost constant aspect ratio in bubbles formed by tens of 2D material pairs.^[107,108,157–162] More recently, Ma et al. conducted experiments to measure the lateral retention force required to move the bubble with an AFM tip in all directions (Figure 4a).^[163] They found that the motion of the bubble in the graphite-graphite interface is isotropic, and the lateral retention forces increase almost linearly with the bubble's contact line, as well as with increasing resting times at the interface. There are still ongoing efforts to elucidate the multiscale nature of the mechanical behavior of 2D material bubbles.^[160]

4.1.2. The Self-Cleaning Mechanism

The formation of bubbles has also been advantageous for high-performance 2D material devices. This is due to a self-cleaning mechanism revealed by TEM experiments involving these bubbles.^[141,151] Interfaces between 2D materials and their substrates outside of the bubble regions exhibit atomic-level smoothness and are free of any contaminants.^[141] In addition, Hou et al. also observed moiré patterns of twisted bilayer graphene outside the nanobubble regions, further indicating the atomic cleanliness at the graphene-graphene interface.^[147] Moreover, these nanobubbles display remarkable mobility. After the manipulation of nanobubbles via tip scanning, the initially contaminated region can fully recover to a contaminant-free state, marked by the emergence of perfect moiré patterns.^[147] This self-cleaning mechanism has demonstrated its use at the device level, as discussed in the introduction. Specifically, carrier mobilities of MoS₂/graphene/hBN devices (where $S < 0$, leading to interfacial bubbles of contaminants) and mica/graphene/hBN devices (where $S > 0$, leading to interfacial adlayers of contaminants) are $\approx 60\,000$ and $1000\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$, respectively.^[18] Nonetheless, despite the presumption of atomic-level cleanliness in bubble-free areas, the existence of random contaminant bubbles limits the size of the clean area to a few microns. Consequently, the design of gates within devices must adopt a serpentine configuration to circumvent direct interaction with these bubbles, potentially hindering the practical application of 2D materials.^[18] Recognizing these limitations, there has been a proactive pursuit of methodologies aimed at expanding the bubble-free area, a topic briefly touched upon in the subsequent discussion.

4.2. Exploiting the Self-Cleaning

4.2.1. Thermal Annealing

Thermal annealing is commonly used to activate self-cleaning and increase the clean area of heterostructures (see Figure 4c). At high temperatures, the diffusion of pollutants is promoted, allowing small bubbles to aggregate into larger ones.^[18,149] Similarly, introducing high temperatures during the pick-up or drop-off process in the transfer of 2D materials is beneficial for achieving a bubble-free interface.^[86] More recently, it has been found that laminating the heterostructure at $\approx 180\text{ }^\circ\text{C}$ maximizes the clean area of the interface.^[132] However, for systems with a positive spreading number where the self-cleaning mechanism is absent, annealing can make things “worse”: the contaminants can

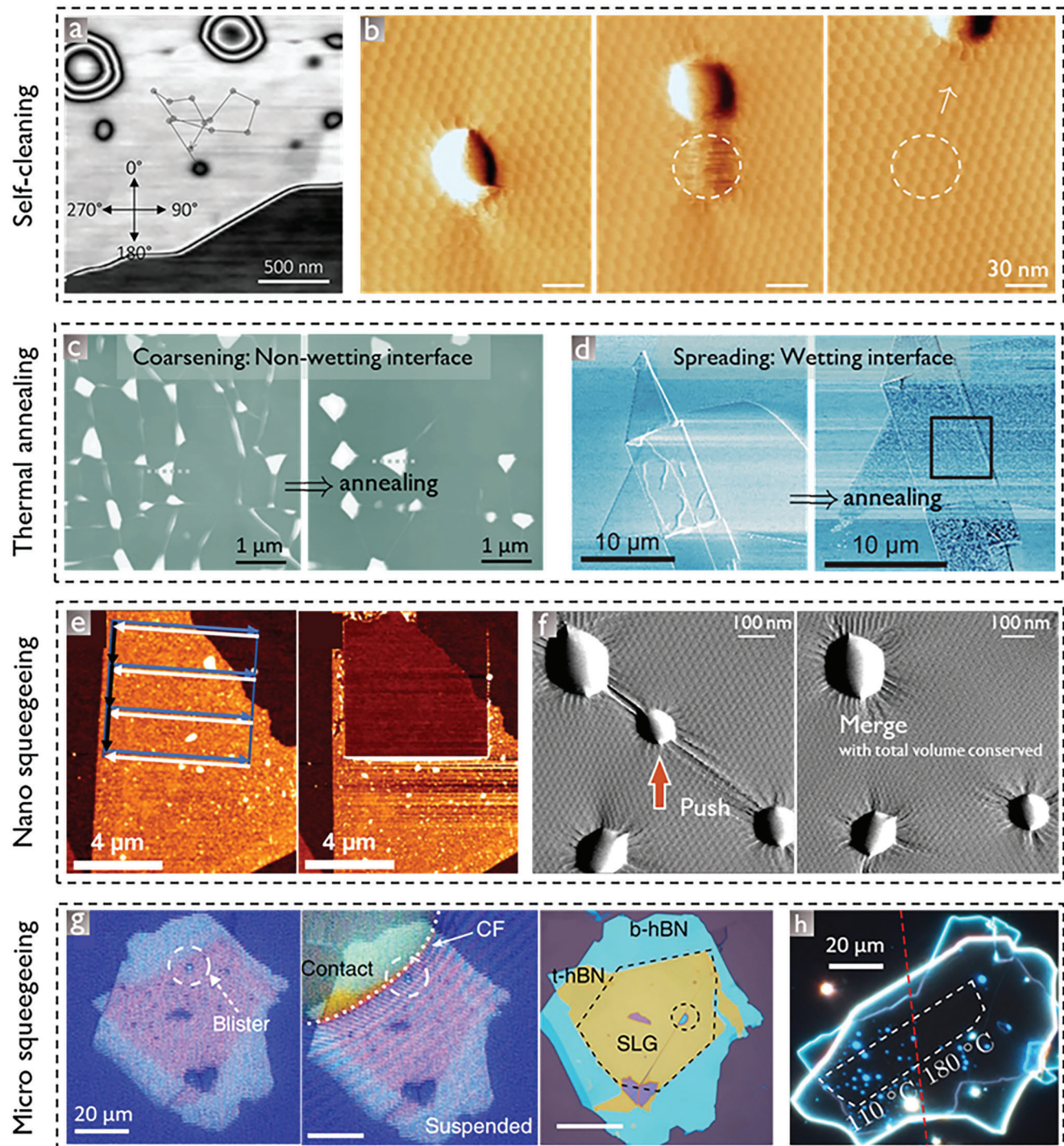


Figure 4. Summary of cleaning method for 2D material interfaces. a,b) AFM topography of an area with bubbles and the trajectory of a moving bubble in subsequent scans. a) Reproduced with permission.^[163] Copyright 2023, American Chemical Society. b) Reproduced under terms of the CC-BY license.^[147] Copyright 2021, published by Springer Nature. c,d) AFM topography images of graphene on hBN c) and hBN on hBN d) before and after annealing. c) Reproduced with permission.^[77] Copyright 2015, IOP Publishing. d) Reproduced with permission.^[106] Copyright 2020, American Chemical Society. e) AFM topography images of CVD WSe₂ on hBN before and after the squeegeeing treatment. The arrows show the general raster pattern used to flatten an area. Reproduced with permission.^[167] Copyright 2018, American Chemical Society. f) AFM deflection images in lateral force mode of suspended twisted bilayer graphene before and after mechanical treatment. The arrow shows the direction of the mechanical treatment. Reproduced under terms of the CC-BY license.^[147] Copyright 2021, published by Springer Nature. g) Optical images of the hBN-graphene-hBN structure during the micro squeegeeing process at a temperature of 180 °C. Reproduced under terms of the CC-BY license.^[132] Copyright 2018, published by Springer Nature. h) Optical dark field image of the hBN-graphene-hBN structure after cleaning where the part to the left of the red dashed line is mechanically treated at 110 °C, while the part to the right is treated at 180 °C. Reproduced under terms of the CC-BY license.^[132] Copyright 2018, published by Springer Nature.

spread to the entire 2D material interface after thermal annealing treatment (see Figure 4d).^[106]

4.2.2. Mechanical Treatment

Direct mechanical treatment using tools like AFM tips provides an additional method for utilizing self-cleaning. Previous reports have utilized contact mode AFM to scan the surface of graphene in a broom-like manner, and it was discovered that the processed area appeared much cleaner than untreated areas.^[164,165] Likewise, a “nano-squeegee” technique has been found useful in creating micron-sized clean interfaces by using an AFM tip to carefully squeeze contaminants out from between 2D layers and their substrates (Figure 4e).^[146,164,166–168] Hou et al. reported that the coalescence of nanopockets induced by mechanical treatment can result in larger atomically clean graphene-graphene interfaces (as evidenced by moiré patterns, Figure 4f).^[147] Additionally, a micrometer-scale “squeegee” can be achieved by controlling the contact angle of the stamp to the substrate during the van der Waals pick-up transfer process (Figure 4g).^[169,170] It was found that contaminant bubbles can be efficiently swept away from the 2D material interface by utilizing an inclined substrate and a lens-like stamp. Notably, at a temperature of 180 °C (Figure 4h), the bubbles could become physically mobile, allowing for a sweeping speed of $>10\ \mu\text{m s}^{-1}$ and resulting in bubble-free areas that are limited only to the size of the exfoliated 2D materials.^[86,132]

5. Discussion

In this section, we will explore the challenges and opportunities for further research and innovation in this field by examining the significance of cleanliness, the current limitations of cleaning and transfer techniques, as well as fundamental questions that arise from interfacial wetting.

5.1. Significance of Bubble-Free Interfaces

Randomly distributed bubbles can create sources of unwanted disorder. Fortunately, by utilizing interfacial cleaning methods, it is possible to generate large areas free of bubbles. These clean interfaces facilitate the investigation of properties in heterogeneous systems: 1) Clean interfaces provide a platform for studying the intrinsic nature of 2D materials. Treated samples have shown improved electrical properties comparable to those of high-quality exfoliated samples.^[166] Furthermore, due to a reduction in charge and strain inhomogeneities, graphene can exhibit ballistic transport properties in bubble-free channels of hBN/graphene/hBN heterostructures, and the quantum Hall effect can be observed.^[169] 2) The removal of interfacial contaminants can reduce interlayer spacing, thereby increasing the interlayer coupling effect. This can significantly reduce the PL line width, indicating an improvement in the spatial homogeneity of the PL spectrum.^[167] In addition, interlayer excitons and a bilayer-like Raman signature can be observed in MoSe₂/WSe₂ heterostructures, providing evidence of strong interlayer coupling. 3) Bubble-free interfaces are crucial for twistronics due to

the strain effect. Specifically, reduction in strain inhomogeneity homogenizes the twist angle between layers, enabling the study of novel properties of moiré superlattices, such as room-temperature superconductivity^[171–173] and abnormal quantum Hall effect,^[174] among others.

5.2. Limitations of Current Methodologies

We have discussed various transfer techniques falling under the categories of wet and dry transfer. It is crucial to acknowledge that each of the aforementioned methods comprises multiple steps and possesses distinct merits and demerits. Additionally, it is feasible to merge steps from diverse approaches to achieve specific goals. For instance, we can anticipate that transferring 2D materials and devices across a large area, without incurring damage or contaminant, would be facilitated by techniques that integrate strategies for detaching PMMA layers from 2D materials in wet transfer with the approaches employed in viscoelastic dry transfer. However, compared to transfer methods, which have provided various options and demonstrated ongoing innovation, there are relatively few options available for clean methods, especially when dealing with the extensive interfaces present in 2D materials and devices. Current strategies, including both thermal and mechanical treatment, rely heavily on the self-cleaning mechanism, which only works for systems with negative spreading parameters. Even in systems where contaminants de-wet into bubbles, this mechanism has limitations, such as breakdown for nanoscale interfacial bubbles in tiny-angle twisted bilayer graphene, as demonstrated in a recent study.^[175] While mechanical treatment at elevated temperatures shows promise as a solution,^[86,132] further research is required to determine the optimal loading dynamics.

5.3. Large-Scale Production of 2D Materials

While mechanically exfoliated 2D materials currently offer the highest-quality samples for showcasing their properties, the realization of their ultimate practical applications demands the availability of large-area 2D materials. Achieving this goal necessitates scalable approaches such as CVD synthesis.^[19] However, the dry and wet transfer methods, as discussed earlier, possess limitations as they are mostly effective for materials ranging from a few microns to centimeters in size. In contrast, the roll-to-roll process has emerged as a promising technique for achieving scalable transfers.^[176] This is underscored by successful demonstrations such as the fabrication of a 100-meter-long graphene sheet.^[177] Ongoing endeavors are focused on refining the roll-to-roll method to enable mature commercial production of large-scale 2D materials, such as reducing costs, minimizing mechanical damage, and eliminating residues during the transfer.^[178–185] Nevertheless, the most critical challenges in this area may revolve around the growth of high-quality 2D materials since they are significantly plagued by intrinsic defects, grain boundaries, wrinkles, and other imperfections.^[19,186]

5.4. Questions from the Perspective of Interfacial Phenomena

Achieving clean 2D materials and devices presents not only an engineering problem but also a series of outstanding fundamental questions to be answered, particularly regarding the mechanics of 2D material interfaces.^[155] We list a few examples below and believe that properly studying these questions can provide an in-depth understanding of the interaction between contaminants and 2D materials and facilitate the development of novel transfer and cleaning methods.

How to characterize and predict the spreading parameter of 2D material systems – an important factor to consider when selecting specific transfer and cleaning methods? It seems sensible to determine the spreading parameter via the measurement of the film-substrate adhesion Γ , film contact angle θ_f , substrate contact angle θ_s , and surface tension γ_l of the involved liquid since the spreading parameter can also be expressed as $S = \Gamma - \gamma_l (\cos \theta_s + \cos \theta_f)$.^[64] However, the intrinsic contact angle of 2D materials remains a topic of debate as evidenced by the large variation in the previously measured values (again often attributed to the contaminant effect).^[187] Fortunately, discerning whether S is positive or negative is comparatively simpler as it is just a matter of whether interfacial bubbles can form. So far there have been tens of pairs of 2D material interfaces that have exhibited $S < 0$ when exposed to typical contaminants such as water and hydrocarbons, whereas only a limited number of pairs have shown $S > 0$.^[18,64,106,153] Nevertheless, accurately establishing S for various material pairs in the presence of diverse contaminants remains demanding, due to the complexity of the chemistry involved in the contaminant and the vast array of possible combinations.^[131]

What are the ideal kinetics for the wet transfer method, which involves a liquid layer at the interface between 2D materials and substrate? In the case of dry transfer methods, such as employing a viscoelastic stamp, pioneering research has established that removing the stamp away from the substrate with sufficiently high peel velocity can lead to adhesion that is strong enough to pick up the target material away from the substrate. Conversely, a sufficiently low peel velocity can be used to separate the target material from the stamp and drop it off onto the substrate.^[188] In contrast, the dynamics governing wet transfer methods exhibit a higher level of complexity due to the interplay between the viscous liquid layer and the elastic 2D materials.^[189–191] Currently, a comprehensive understanding of the kinetic strategies required for the deterministic pick-up and drop-off of 2D materials using wet transfer techniques is still lacking.^[192]

What are the structures and phase behaviors of different substances when they are confined to a nanoscale interface? Recent experiments and molecular simulations have unveiled the fascinating size-dependent shape characteristics of contaminant bubbles.^[160] Notably, the minimum height of these bubbles measures a few Ångström, equivalent to a phase of a single molecular layer of interfacial contaminants.^[157] Meanwhile, the confining pressure shows an inverse correlation with the bubble size with potential intensities up to tens of GPa.^[64,107,193] The confined contaminants are likely a mix of water, hydrocarbons, and other trace elements^[109,110,131] or can be partially controlled.^[128,147,194–196] The combination of these understandings may offer a unique avenue for investigating the phase separation phenomena within multicomponent liquid mixtures under the extreme of nanoscale

confinement.^[197] The problem is more complicated when considering the dynamic evolution of an array of contaminant bubbles over time or under thermal and mechanical stimuli. Although the recent process has revealed the ripening and merging mechanism underlying the coarsening behavior of bubbles,^[147,195,196] there are still questions of both fundamental and practical importance to be addressed, such as how and how fast the coarsening takes place toward the final patterns of bubbles that have been observed.

6. Conclusion

In conclusion, the pursuit of clean and pristine 2D materials has presented a considerable challenge to the field, but progress has been made in recent years through the development of simpler and faster transfer techniques, as well as multi-scale methodologies for evaluating material cleanliness. The emergence of passive and active cleaning strategies has opened up new possibilities for achieving and maintaining the desired level of cleanliness in 2D materials and devices, enabling fundamental studies of their electronic, optical, and mechanical properties, and paving the way for their integration into a wide range of applications. However, despite significant progress, there are still limitations and outstanding fundamental questions that must be addressed. It is imperative to work further in developing novel transfer and cleaning methods that can facilitate the attainment of pristine 2D materials and devices while gaining an in-depth understanding of the interaction between contaminants and 2D materials. Properly studying these questions is expected to lead to breakthroughs in the field and further advancements in the development of 2D materials for various applications.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

2D materials, clean, contaminants, interface, transfer methods

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Wenlong Dong is currently a Ph.D. candidate under the supervision of Prof. Luqi Liu at the National Center of Nanoscience and Technology, China. He received his B.S. in Materials Science and Engineering from the University of Chinese Academy of Sciences in 2020. His research interests are strain engineering of 2D materials.



Zhaohe Dai is currently an Assistant Professor at the College of Engineering, at Peking University. He obtained his B.Sc. degree in Theoretical and Applied Mechanics from the University of Science and Technology of China in 2013, and his M.S. degree in Solid Mechanics from the Institute of Mechanics in 2016. In 2020, he completed his Ph.D. in Solid Mechanics from the University of Texas at Austin. Before joining PKU in March 2022, he worked as a Marie Curie Fellow at the Mathematical Institute, University of Oxford. His research interests center around the mechanics of thin films, including their surfaces and interfaces.



Luqi Liu is currently a full professor at the National Center of Nanoscience and Technology, China. She obtained her bachelor's and master's degrees from Beijing Normal University in 1994 and 2000, respectively, and her Ph.D. degree from the Institute of Chemistry at the Chinese Academy of Sciences in 2003. After that, she undertook postdoctoral research in the Department of Materials and Interfaces at the Weizmann Institute of Science (Israel). In 2007, she joined NCNST as an associate professor and was promoted to professor in 2018. Her research interests focus on the mechanics of low-dimensional nanomaterials and multifunctional polymer-based nanocomposites.



Zhong Zhang is a full professor at the University of Science and Technology of China. He obtained his bachelor, master, and Ph.D. degrees from the University of Science and Technology of China in 1990, 1996, and 1999, respectively. In 2001, he was granted the Sofja Kovalevskaja Award by the Alexander von Humboldt Foundation, which supported him to establish an independent research group at the Institute for Composite Materials, University of Kaiserslautern, Germany. He was recruited by NCNST in 2005 and joined the University of Science and Technology of China in 2022. His major research interests are multifunctional polymer nanocomposites, nanomechanics, nanostructured surfaces and coatings, nanomanufacturing, and their applications.