



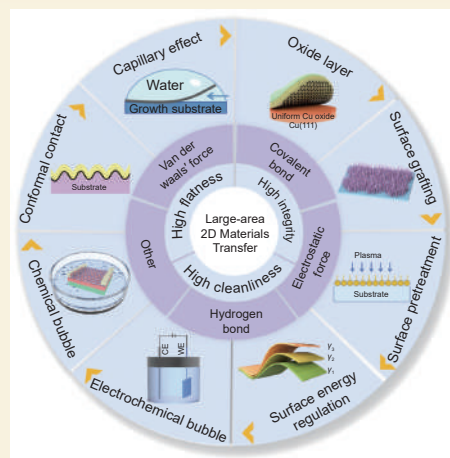
Controlling interfacial adhesion during the transfer of large-area 2D materials: mechanisms, strategies, and research advances

HU Rong[†], SONG Jia[†], HUANG Wei, ZHOU An-na, LIN Jia-long,
CAO Yang^{*}, HU Sheng^{*}

(College of Chemistry and Chemical Engineering, Xiamen University, Xiamen 361005, China)

Abstract: Large-area two-dimensional (2D) materials, such as graphene, MoS₂, WS₂, h-BN, black phosphorus, and MXenes, are a class of advanced materials with many possible applications. Different applications need different substrates, and each substrate may need a different way of transferring the 2D material onto it. Problems such as local stress concentrations, an uneven surface tension, inconsistent adhesion, mechanical damage and contamination during the transfer can adversely affect the quality and properties of the transferred material. Therefore, how to improve the integrity, flatness and cleanness of large area 2D materials is a challenge. In order to achieve high-quality transfer, the main concern is to control the interface adhesion between the substrate, the 2D material and the transfer medium. This review focuses on this topic, and finally, in order to promote the industrial use of large area 2D materials, provides a recipe for this transfer process based on the requirements of the application, and points out the current problems and directions for future development.

Key words: 2D materials; Graphene; Large-area; Interfacial adhesion modulation; High quality transfer



1 Introduction

Large-area, high-quality two-dimensional (2D) materials^[1-2] possess unique physical and chemical properties, such as atomic-scale thickness, excellent mechanical flexibility, and outstanding electrical^[3-4], magnetic^[5-6], optical^[7], mechanical^[8-9], and chemical properties^[10]. They are widely used in fields such as electronic devices^[11-13], optical devices^[14-16], optoelectronic devices^[17-19], catalysis devices^[20-21], flexible devices^[22-23] and energy storage devices^[24-26]. Compared to other synthesis methods^[27-28], Chemical vapor deposition (CVD)^[29-31] can produce high-purity, uniform, and low-defect films, with precise control over composition, thickness, and structure, making it the dominant method for 2D material growth. However, high-quality 2D material growth normally requires high temperatures above 700 °C^[32] and corrosive gases^[33]. Such high temperatures can negatively impact the target substrate in flexible devices

and optoelectronic devices, affecting the reliability and performance of the final devices. For example, for flexible electronic devices, the required substrates are flexible polymers or glass, which are not heat-resistant, making it difficult to directly grow 2D films on the substrates without any damage^[34-35]. It is reported that direct growth on oxide substrates is possible, but the choice of material substrates is rather limited^[36]. The use of precursors such as salts can lower the growth temperatures and improve quality^[37-40], but their introduction can lead to doping and contamination of the resulting 2D materials^[41-43]. Although CVD technology can currently grow 2D heterostructures directly without a transfer process^[44-45], the choice of

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materials and assembly sequence are limited by the growth temperature gradient^[46]. Mechanical transfer techniques offer unique flexibility, enabling transfer onto any substrate, allowing the integration of various nanomaterials, overcoming the limitations of lattice matching and size mismatch^[47–49]. Therefore, transfer from specialized growth substrates is required.

The transfer process involves exfoliating the 2D material from the growth substrate (e.g., copper foil, silicon wafer) while the transfer medium supports and protects the material during the process, and then it is placed onto the target substrates^[47–49]. Several challenges remain in this process^[50–52]. (1) Maintaining integrity: high-temperature growth conditions can strengthen the interfacial adhesion between the 2D material and the growth substrate, leading to defects or damage during transfer. Therefore, weakening the interfacial adhesion between these two interfaces is crucial for ensuring damage-free exfoliation. (2) Improving flatness: uneven force distribution during transfer can result in wrinkles or bonding issues that affect flatness. Thus, enhancing the interfacial adhesion between the 2D material and the target substrate is key to achieving smooth and uniform bonding. (3) Ensuring cleanliness: maintaining surface cleanliness after transfer is challenging, especially when removing transfer medium residues and surface contaminants that can negatively impact the material's electronic properties and interface characteristics. Therefore, controlling the interfacial adhesion between the 2D material and the transfer medium is essential for enabling a clean release. Although in small-scale transfer, process optimization allows effective control over localized stress, mechanical damage, and contamination^[53–54], as the transfer area increases to the centimeter scale or larger scale, the likelihood of localized damage, cracks, wrinkles, or structural failure significantly increases^[55]. Therefore, optimizing the transfer process to reduce damage, contamination and structural defects remains one of the key challenges in the practical applications of 2D materials.

The key challenges in the CVD transfer process of 2D materials lies in how to properly modulate the

interfacial adhesion forces among the 2D materials, growth substrate, target substrate, and transfer medium. These interfacial forces, including van der Waals (vdW) forces^[56–57], electrostatic forces^[58–59] and covalent bonds^[60], play a crucial role in the material transfer process. Existing studies have made significant progress in these research areas and have proposed various methods (such as using bubbling techniques to achieve as complete a transfer as possible from the growth substrate; annealing and plasma treatments to increase the adhesion between the target substrate and 2D materials; and employing low molecular weight transfer media, sublimable, and water-soluble transfer media to improve surface cleanliness)^[61–64]. Combining these mature methods from the perspective of modulating interlayer interactions would be highly beneficial for optimizing the transfer process.

This review systematically examines the interfacial adhesion forces in the context of large-area 2D material transfer, with a focus on their types, quantification methodologies, and characterization approaches. Special emphasis is focused on how to precisely control the interfacial adhesion forces between the 2D material, the growth substrate, the target substrate, and the transfer medium in the transfer process, to achieve high-quality large-area 2D material transfer. Firstly, we analyze the interfacial adhesion forces governing the transfer process through theoretical and experimental perspectives, emphasizing the 3 predominant adhesion mechanisms: vdW forces, covalent bonds, and electrostatic forces. We also introduced specific methods for measuring and calculating adhesion forces from the perspectives of experimental characterization and theoretical analysis. Building upon this foundation, the regulation of interfacial forces is systematically examined through three critical parameters: integrity, flatness and cleanliness, which collectively determine the quality of material transfer. Furthermore, this review provides practical guidelines for selecting transfer methodologies according to specific application demands. Finally, this review summarizes several innovative transfer and characterization concepts, which inspires future ex-

ploration in this field. This review aims to provide systematic insights for researchers working on the large-area transfer of 2D materials, helping them understand and regulate interfacial adhesion forces to achieve efficient, high-quality transfers, and to promote the large-scale application of 2D materials in fields such as electronics, optoelectronics and flexible electronics.

2 Interfacial adhesion forces in 2D material transfer: types, mechanisms, quantitative characterization and theoretical analysis

In the transfer process of 2D materials, interfacial adhesion forces not only affect the stability of 2D materials but also determine the transfer quality and efficiency. In addition to vdW forces, covalent bonds, and electrostatic forces, weak interactions induced by specific elements also play a crucial role in interfacial adhesion. These forces act differently at various interfaces, and their strength significantly influences the transfer quality and efficiency. This section will explore the applications and limitations of these interfacial adhesion forces in large-area transfers, laying the foundation for the discussion of interfacial force modulation strategies.

By integrating experimental methods and theoretical analysis, this section will further investigate the impact of interfacial adhesion forces on the transfer process and elaborate on their quantification and characterization methods, providing theoretical support and practical guidance for high-quality 2D heterostructure transfer. A deep understanding and modulation of these mechanisms will help address the aforementioned challenges and promote their applications in quantum devices, optoelectronic chips, and flexible electronics.

2.1 Interfacial adhesion forces during the 2D materials transfer process

During the transfer process of 2D materials, interfacial adhesion forces play a critical role in determining the success or failure of the transfer. Some stud-

ies have shown that the transfer process can significantly degrade the electronic properties and structural quality of 2D materials, thereby affecting device performance metrics and increasing process time and production costs^[65]. These detrimental effects are mainly attributed to surface wrinkles, material fractures, surface contamination, and unintentional doping during the transfer process^[66–68], all of which are closely related to interfacial forces. Therefore, understanding the types and mechanisms of forces involved in the transfer process is essential for ensuring a smooth transfer and enhancing device performance.

Interfacial adhesion forces mainly include vdW forces, covalent bonds and electrostatic forces, each of which directly influences the transfer quality. Additionally, specific metal-2D material interactions and hydrogen bonds play specific roles in certain transfer processes. A comprehensive understanding of these factors is key to optimizing the 2D material transfer process.

2.1.1 Types and mechanisms of main interfacial adhesion forces

(1) vdW forces: As a non-bonding interaction, vdW forces play a crucial role in the disassembly and reassembly of layered crystalline materials^[69], such as vdW materials. The strength of vdW interactions typically ranges from 0.1 to 10 kJ mol⁻¹, which is 2 to 3 orders of magnitude weaker than ionic or covalent bonds (approximately 100–1000 kJ mol⁻¹) (Fig. 1a). Although vdW forces are relatively weak, they are sufficient to ensure strong adhesion between materials. However, conventional 3D rigid materials, due to their surface roughness, struggle to achieve the necessary vdW distance for full activation of vdW interactions (Fig. 1b-d). In contrast, 2D materials, which possess atomic-scale smoothness and flexibility, serve as an excellent example of materials that exhibit strong vdW interactions (Fig. 1e). This weak interaction enables layered crystals to be separated into individual atomic planes and reassembled into heterostructures. As a short-range force, it is present throughout the entire 2D material transfer process, particularly in the physical adsorption between 2D materials and their

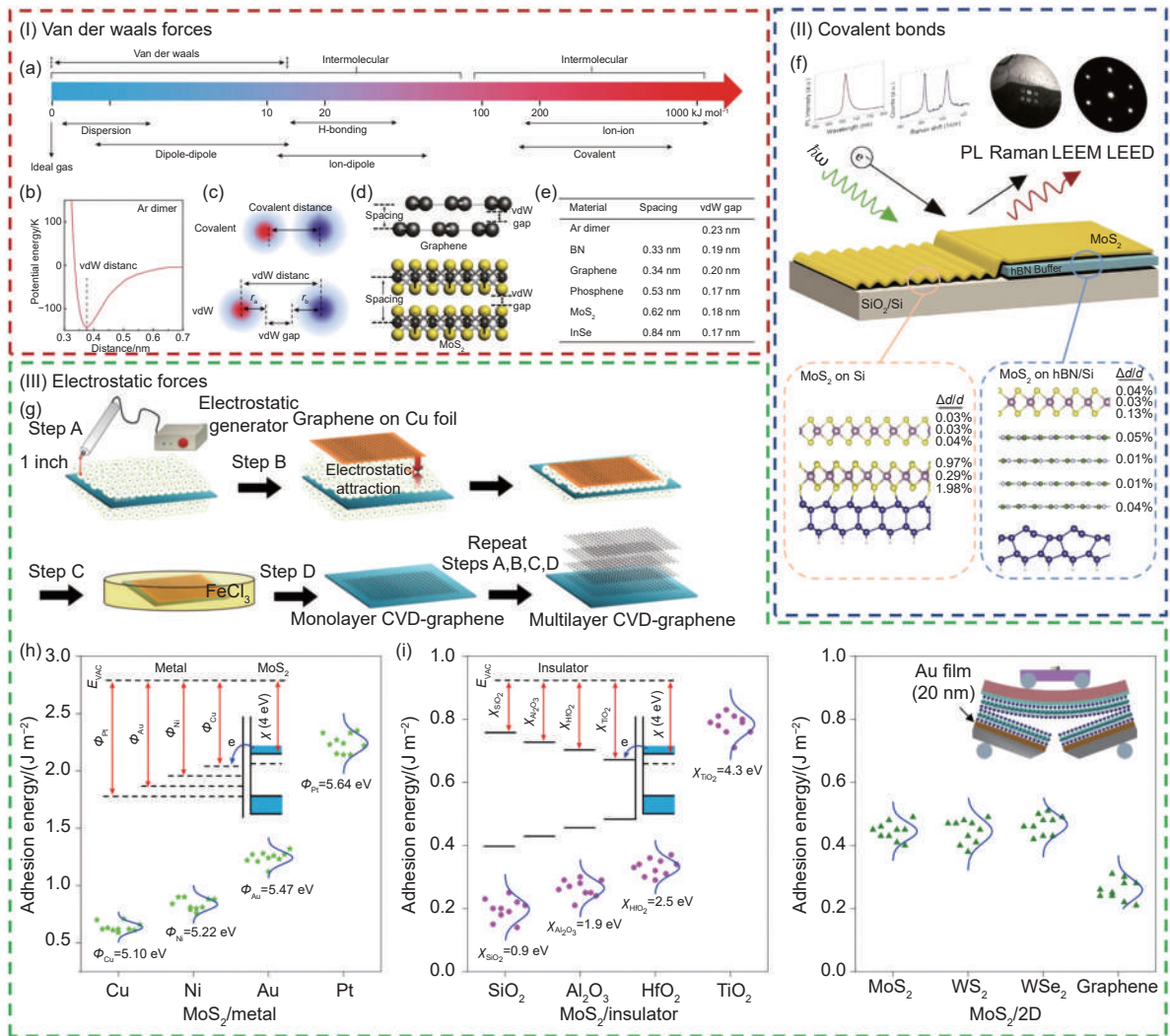


Fig. 1 Different types of forces in 2D materials. (I) Definition of vdW interaction, vdW distance, and vdW gap. (a) Energies of various molecular interactions. (b) Potential energy versus distance for an Ar dimer system. (c) Schematic illustration of the vdW gap and vdW distance in a covalent-bonded system and a vdW system. (d, e) Comparison between layer spacing and calculated vdW gap in various layered 2D materials^[69]. (II) Typical example of covalent bonds. (f) Schematic of the characterization of MoS₂ with and without an h-BN buffer on Si-based substrates^[75]. (III) Typical example of electrostatic forces. (g) Schematic of electrostatic force-assisted transfer^[78]. Measured interfacial adhesion energy of the MoS₂/metal (h), MoS₂/insulator (i) and MoS₂/2D interfaces (j)^[75]. Reprinted with permission

substrates^[47]. Specifically, during the transfer process, when a 2D material is transferred from one substrate to the target substrate, the strength of the interfacial vdW forces determines the final device quality and performance.

One of the most typical applications of vdW forces in the transfer process is using one 2D material (eg. h-BN) to pick up other 2D materials (eg. graphene)^[70]. This method has been widely used for transferring small-scale 2D materials (typically with lateral sizes less than 200 micrometers). However, as the film size increases, this weak interaction^[71] makes

the material highly susceptible to cracks and fractures during the transfer process, posing limitations for large-area and industrial applications. While interfacial defects, bubbles, and other imperfections provide valuable insights into the properties of 2D materials^[71–73], they significantly reduce the long-term stability and lifespan of devices in practical applications.

(2) Covalent bonds: As a strong type of interaction, covalent bonds play an indispensable role in the transfer process, especially in the large-area transfer of 2D materials. During the thin-film growth process,

gaseous precursors decompose at high temperatures and undergo chemical reactions on the growth substrate surface, forming a 2D thin film. Choi et al.^[74] found that after high-temperature annealing, covalent C—O bonds can form at the Gr/SiO₂ interface. Therefore, covalent bonds are essential in both the film growth process and the stabilization of devices, contributing to improved device quality. However, in the transfer process of 2D materials, these strong covalent bonds make it more difficult to separate the material from the substrate, potentially leading to film damage during transfer (Fig. 1f)^[75]. As a result, additional strategies are often required to weaken this adhesion force during the transfer process.

One common strategy is to remove the substrate by chemical etching^[76] to eliminate its strong bonding with the material. Although this method has been widely used over the past decade, its high cost and the non-reusability of the substrate limit its industrial-scale application. Additionally, the use of etchants may introduce contamination, and even though contamination-free etchants^[77] have been proposed, challenges related to implementation and cost remain. Therefore, developing more efficient and environmentally friendly transfer strategies is essential for improving transfer efficiency and reducing costs. Specific interfacial force modulation methods will be discussed in detail in later sections.

(3) Electrostatic forces: electrostatic forces arise from differences in interfacial charge distribution and can be dynamically modulated through external fields. They play a crucial role, particularly in the adhesion and separation of materials from substrates (Fig. 1g)^[78]. For example, Sumanasekera et al.^[80] utilized electrostatic attraction to transfer freshly exfoliated graphite crystals onto target substrates. However, this method is only applicable for transferring small graphene flakes (typically on the micrometer scale) under high voltage. On the other hand, Han et al.^[81] facilitated the direct transfer of graphene onto various target substrates (such as glass, PET and PDMS) using electric field forces. This process combines mechanical pressing, heating and electrostatic forces to

achieve ultraconformal contact under high temperatures and high voltage. Specifically, Na₂O and K₂O in glass decompose under an electric field to form Na⁺, K⁺ and O²⁻, promoting the formation of C—O covalent bonds between graphene and oxygen, while the negative charge on the PET surface enhances its adhesion to graphene. Despite these advancements, practical challenges remain, particularly in the precise modulation of environmental temperature and electric field strength, as excessive voltage may cause substrate deformation or damage.

Furthermore, electrostatic forces can also assist in the exfoliation process of 2D materials (Fig. 1h-j). In metal-assisted 2D material transfer, electrostatic forces also play a vital role. Shin et al. found that due to the higher work function of metals (such as Au, Cu and Ni) compared to the electron affinity of MoS₂, electron would transfer from MoS₂ to the metal^[75]. This results in a positively charged MoS₂ surface and a negatively charged metal substrate, thereby enhancing the electrostatic attraction between them and improving the stability and transfer efficiency of the heterostructure. Such interfacial charge modulation can effectively optimize material adhesion and increase transfer success rates. Overall, the critical role of electrostatic forces in 2D material transfer has been widely recognized. However, their applicability, environmental dependence, and controllability in different systems still require further optimization, and relevant modulation strategies will be discussed in later sections.

2.1.2 Specific interfacial adhesion: potential applications for transfer processes

In addition to the above forces universally applicable to 2D materials, there also exist specific forces that are unique to certain 2D materials. For instance, the metal-2D material interactions, and non-covalent forces like hydrogen bonding. These specific interactions can significantly influence the stability of the materials and the interfacial adhesion, therefore, offer potential innovative pathways for advancing 2D material transfer technologies with optimized transfer efficiency. In the following, we provide a few relev-

ant examples.

(1) Au-S bonds: the bonding interaction between gold (Au) and sulfur (S) has demonstrated unique advantages in the adhesion of metals to transition metal dichalcogenide (TMD) materials^[82]. The metal-sulfur interaction between Au and TMD materials not only enhances interfacial adhesion but also facilitates interfacial contact under appropriate pressure due to gold's exceptional mechanical flexibility. Similarly, interactions between gold and other elements, such as phosphorus (P) and chlorine (Cl), exhibit significant effects at P/Au and Cl/Au interfaces, forming covalent-like quasi-bonding^[83], which further strengthens the interfacial adhesion. Leveraging these principles, Gao et al.^[84] developed an Au-assisted mechanical exfoliation technique, successfully achieving the exfoliation of millimeter-scale 2D materials. However, fully harnessing these complex interactions between metals and 2D materials, particularly for damage-free transfer of wafer-scale single crystals, remains a significant challenge. Additionally, the strong interactions between metals and 2D materials complicate post-transfer processing. Solvents are typically required to remove the metal layer^[76], which may increase costs and introduce contaminations, thereby limiting the industrial applicability of this method. Consequently, optimizing the interactions between metals and 2D materials, minimizing post-processing requirements, and improving transfer efficiency remain critical research objectives for future studies.

(2) Hydrogen bonds: as a specific type of electrostatic interaction, hydrogen bonds exhibit relatively high binding energies (25-40 kJ mol⁻¹), significantly surpassing those of vdW forces (~ 5 kJ mol⁻¹). These interactions are limited to elements with high electronegativity, such as nitrogen (N) and oxygen (O). Additionally, the unique directionality, specificity, and reversibility of hydrogen bonds make them crucial in interfacial adhesion. For example, Yang et al.^[85] significantly enhanced the structural stability of heterostructures and promoted interfacial charge transfer by introducing —OOH groups on the surface of titanium dioxide nanotubes (TNTs), enabling hy-

drogen bond formation (N···H—O—O—Ti) between the —OOH groups on TNTs and the —NH₂ groups in Fe-MOF. As a relatively weak non-covalent force, hydrogen bonding holds potential in the assembly, transfer, and functionalization of 2D materials. By providing temporary yet stable interfacial adhesion, hydrogen bonds can mitigate mechanical stress-induced damages during transfer, thus improving the stability of materials and the quality of subsequent devices^[86]. Although the research on the direct formation of hydrogen bonds between substrates or transfer media and 2D materials remains limited, their higher binding energy compared to vdW forces enables them to provide more stable interfacial adhesion during transfer. This not only strengthens the bonding between materials and transfer media but also establishes temporary yet stable connections between materials and substrates, thereby reducing cracking, contamination, or wrinkling and enhancing transfer quality and material performance. These specific interfacial interactions may play a critical role in the transfer of certain 2D materials. Rational modulation of these forces not only helps optimize transfer quality and interfacial stability but also offers new strategies for achieving large-area, high-precision transfer of 2D materials, further advancing the fabrication and application of high-performance devices.

Thus, a deep understanding of interfacial bonding forces is crucial for optimizing the transfer processes of 2D materials, enhancing device performance, and advancing large-scale fabrication. Modulation of these bonding forces further improves material adhesion and reduces interfacial defects, thereby leading to enhanced material stability and electronic transport properties for the optimized overall performance of devices. Furthermore, the distinct mechanisms, ranges, and strengths of different types of bonding forces determine their suitability depending on application scenarios. It is necessary to further systematically explore the interfacial bonding forces for the processing capabilities of 2D materials and establishing a solid foundation for their widespread applications.

2.2 Measuring interfacial adhesion forces: characterization techniques and theoretical analysis

The interaction between 2D materials and substrates is often manifested as adhesion force. To better understand the different types of interaction forces, especially the role of adhesion forces in the transfer process of 2D materials, it is essential to quantify and characterize these forces. To accurately capture the details of these interactions, it is necessary to use appropriate theoretical and experimental methods for their quantitative analysis. As our understanding of the interfacial forces deepens, we can better grasp the magnitude and distribution of these forces, providing a theoretical foundation for optimizing the transfer process. Additionally, this lays the groundwork for subsequent strategies to modulate interfacial forces, further improving the efficiency and quality of 2D material transfer. Next, we introduce several commonly used experimental and theoretical methods for quantifying and characterizing the adhesion forces between 2D materials and substrates.

2.2.1 Characterization techniques for interfacial adhesion force

In terms of experimentation, atomic force microscopy (AFM) force spectroscopy is a commonly used method for characterizing interfacial adhesion forces. Through AFM measurements, the adhesion force between 2D materials and substrates can be directly quantified, revealing the magnitude and nature of the adhesion force. For example, Lu et al.^[87] fabricated nanoscale 2D crystals (such as graphene, h-BN and MoS₂) on nanoscale platforms, then used an AFM probe to measure interfacial adhesion forces (Fig. 2a-b). During the measurement, the AFM probe was attached to the top of the nanoscale platform, and stretching or shear was applied in either the vertical or parallel direction relative to the substrate. By analyzing the force-displacement (F-d) curve, the interfacial adhesion energy (IAE), interlayer shear strength, and peeling strength can be quantified, providing detailed data support for the mechanical properties during the 2D material transfer process. Guo et al.^[89] used graphite-coated AFM tips and measured the critical adhe-

sion forces between graphite-graphite ($P_{G/G}$), graphite-BN ($P_{BN/G}$), and graphite-MoS₂ ($P_{MoS_2/G}$), then calculated the adhesion forces at the interface between graphite and other materials using the known adhesion pressure between graphite-graphite.

The above measurements are designed for rigid substrates. For elastic substrates, due to the large deformation of polymeric materials, the adhesion forces between 2D materials and soft polymer substrates are still difficult to characterize. Traditional testing methods may not provide sufficient resolution, resulting in reduced data accuracy. For example, Zhu et al.^[92] indirectly inferred the interfacial adhesion force by measuring the sliding and buckling behavior of graphene on a polyethylene terephthalate (PET) substrate. However, AFM images might overestimate the width of the buckling ridge, leading to inaccurate measurements of adhesion energy. To address this issue, Lu et al.^[88] used the spontaneous formation of buckles and wrinkles during the delamination of MoS₂ on PDMS. By combining AFM measurements of the height and width of the buckles and wrinkles in the delaminated area (Fig. 2c-d), the adhesion force was calculated using a formula fitting method, thus avoiding the issue of measuring step heights on substrates with high elasticity. This method improved accuracy by an order of magnitude compared to traditional methods, providing a more reliable measurement tool for characterizing the adhesion force in the transfer of 2D materials on soft substrates.

Here, we have compiled the adhesion energies between different films as well as the film-substrate interfaces (Table 1), to provide a clear understanding of the interfacial adhesion forces, thereby offering guidance for optimizing the subsequent interfacial transfer processes.

2.2.2 Theoretical analysis for interfacial adhesion force

Density functional theory (DFT) calculations have been widely used to study the interfacial energy between 2D materials and substrates. Through DFT calculations, the interaction forces between different materials can be accurately simulated and predicted,

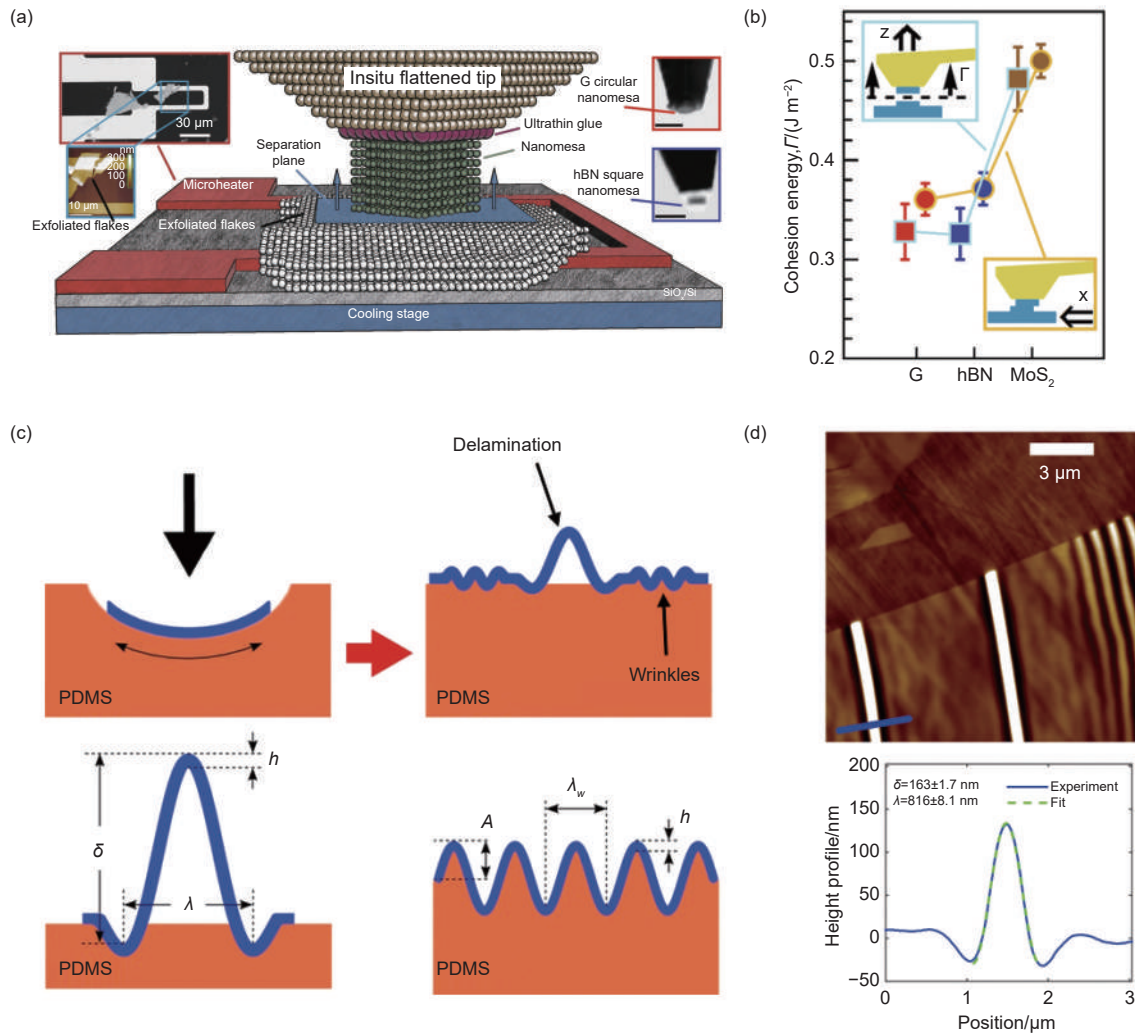


Fig. 2 (a) Schematic diagram of AFM system for interfacial adhesion measurements. (b) Cohesive energy of graphene (G), h-BN, and MoS₂ crystals obtained by normal force measurements (cyan-bordered squares) and shear force measurements (orange-bordered circles) at room temperature^[87]. (c) Schematics of the process to create spontaneous wrinkles and buckle delaminations in few layer MoS₂ on PDMS substrate. (d) AFM image of MoS₂ flakes on PDMS^[88]. Reprinted with permission

helping us to better understand the interfacial energy and its impact on the transfer process. For example, Chiesa et al.^[90] used DFT calculations to determine the vdW energy between graphene and substrates for different graphene layer numbers (Fig. 3a-c). The calculations showed that as the number of graphene layers increased, both the adhesion force and Hamaker coefficient increased significantly, which was consistent with experimental results. This indicates that the number of layers in 2D materials has a significant impact on interfacial adhesion forces, with more layers leading to stronger adhesion, which could affect the transfer efficiency and stability of the 2D material.

Additionally, charge density difference analysis

can be used to study the transfer of electrons during the transfer process, revealing the types of interactions at the interface. For instance, Righi et al.^[91] found that the charge density at the interface is directly related to the interfacial adhesion force (Fig. 3d-i). They pointed out that, typically, in non-ideal stacking configurations, the charge density at the interface is lower than that in the minimum configuration. This finding suggests that the distribution of interface charges has an important impact on the strength of the interfacial adhesion force, and changes in charge density may directly lead to variations in adhesion force. This means that by analyzing interfacial energy and electronic structure, we can provide a the-

Table 1 The adhesion energy between films and the film-substrate interface

Interface	Adhesion energy/(mJ m ⁻²)	Ref.
MoS ₂ -SiO ₂	200±30	[75]
Graphene-SiO ₂	453±6	[93]
h-BN-SiO ₂	208±47	[94]
MoSe ₂ -SiO ₂	270±30	[95]
MoS ₂ -TiO ₂	780±30	[75]
MoS ₂ -Al ₂ O ₃	260±20	[75]
MoS ₂ -HfO ₂	330±20	[75]
MoS ₂ -SiN	429	[96]
Graphene-SiN	3282	[96]
Graphene-PMMA	84	[97]
Graphene-PDMS	7	[98]
Graphene-PET	0.5	[92]
MoS ₂ -PDMS	18±2	[88]
Graphene-Cu	740±130	[99]
Graphene-Au	450±100	[100]
Graphene-Ni	6775±556	[101]
Graphene-Pt	4021	[96]
MoS ₂ -Cu	640±30	[75]
MoS ₂ -Au	1250±50	[75]
MoS ₂ -Pt	2240±60	[75]
MoS ₂ -Ni	840±40	[75]
MoS ₂ -Ti	140±30	[102]
MoS ₂ -Cr	110±50	[102]
Graphene-Graphene	86±16	[103]
Graphene-MoS ₂	140±26	[103]
Graphene-h-BN	126±20	[103]
MoS ₂ -MoS ₂	174±18	[103]
MoS ₂ -h-BN	136±11	[103]
H-BN-h-BN	129±4	[103]
MoSe ₂ -MoSe ₂	355	[104]
WS ₂ -WS ₂	269	[104]
WSe ₂ -WSe ₂	59	[104]
WSe ₂ -WSe ₂	104	[104]
WTe ₂ -WTe ₂	27	[104]

oretical basis for optimizing the interfacial adhesion force in practical transfer processes.

Interfacial roughness is a critical factor influencing the interaction forces between two interfaces. As mentioned above, surface interactions such as vdW forces are significantly altered due to the presence of roughness. A rough surface reduces the actual contact area and introduces localized non-uniform contact states, thereby weakening the effective interfacial forces. Moreover, roughness changes the distribution of separation distances between surfaces, further affecting the strength of short-range forces such as vdW interactions. Therefore, interfacial roughness is closely related to surface adhesion and cannot be ig-

nored when analyzing adhesion behavior. The Maugis-Dugdale model uses a simplified Dugdale model, which has adjustable parameters that can be modified for different scenarios. The adhesion energy between the tip and the sample is calculated using the Maugis-Dugdale theory^[105,153]:

$$W_{\text{adh}} = \frac{F_{\text{adh}}}{\lambda\pi R_{\text{tip}}} \quad (1)$$

where W_{adh} represents the adhesion energy per unit area, F_{adh} is the measured adhesion force, R_{tip} is the tip radius, and λ is an effective coefficient with a range of $1.5 \leq \lambda \leq 2.0$, determined by the characteristics of the contact pair^[106]. This model is based on the following assumptions: the substrate strain is small, the material is a homogeneous, isotropic linear elastic body, and the tip has a perfectly smooth parabolic geometry (*i.e.*, a three-dimensional rotational paraboloid).

$$z_{\text{tip}}(r) = z_{\text{paraboloid}}(r) + z_{\text{roughness}}(r) = \frac{r^2}{2R_{\text{tip}}} - A \cos\left(\frac{2\pi r}{\lambda}\right) \quad (2)$$

Here, r and z are polar coordinates, where R , defines the radius of the overall parabolic shape, and A and λ represent the amplitude and wavelength of the sine wave, respectively. The resulting tip is considered rigid and comes into contact with a rigid plane.

Considering the influence of surface roughness on the measured adhesion energy, the true adhesion energy is calculated using a modified Rumpf model, which reproduces the fundamental trend as a function of roughness. The definition of an axisymmetric tips the superposition of a paraboloid (for the overall tip shape) and a sine wave (to represent roughness).

Therefore, the transfer process of high-quality 2D materials depends on the modulation of interfacial adhesion forces. By understanding and modulating vdW forces, covalent bonds, electrostatic forces, and other interfacial interactions, we can lay the theoretical foundation and practical guidance for realizing a “modulated adhesion-damage-free release” transfer process. With the continuous development of theoretical calculation methods and experimental techniques, it is hopeful that we can resolve the challenges related to adhesion force modulation in the large-scale preparation and application of 2D heterostructures, thereby advancing the widespread application of 2D

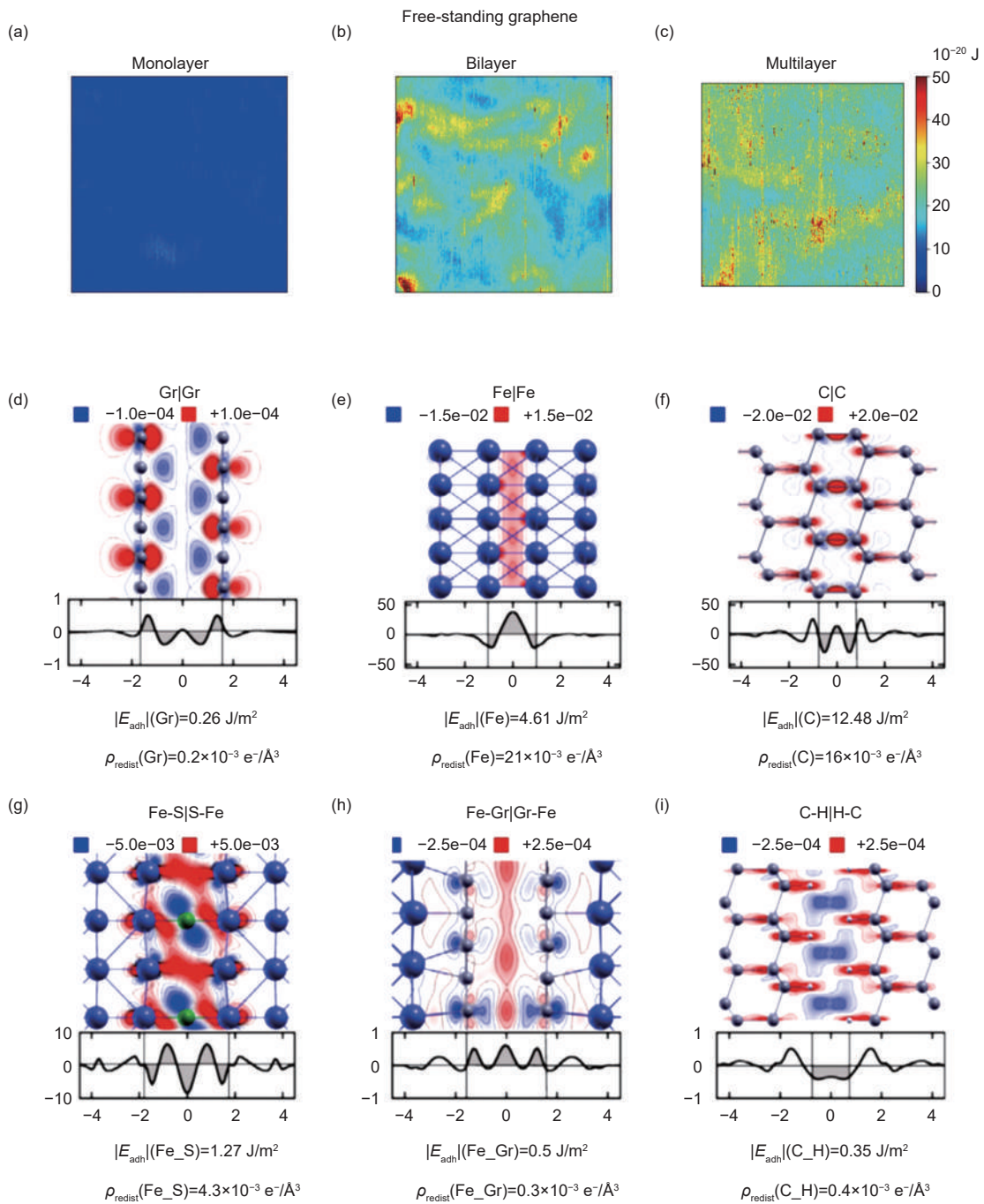


Fig. 3 (a-c) Hamaker coefficient maps for free-standing monolayer, bilayer and multilayer graphene, respectively^[90]. (d-i) Charge density differences ρ_{diff} of Gr, Fe, C, Fe-S, Fe-Gr and C-H, respectively and their connection to adhesion values^[91]. Reprinted with permission

materials in fields such as electronics, optoelectronics, and flexible electronics.

3 Interfacial adhesion modulation for high quality transfer of large-area 2D materials

The key goal of high quality 2D materials in

transfer process, involve maintaining structural integrity, surface flatness, and interface cleanliness^[79,107-112]. Therefore, this section summarizes quality improvement strategies for large-area 2D material transfer from three dimensions: integrity, flatness, and cleanliness (Fig. 4). These strategies include enhancing integrity by modulating interfacial adhesion, as well as improving flatness and cleanli-

ness through interfacial adhesion modulate.

3.1 Integrity enhancement by weakening growing substrate adhesion

The atomic-scale thickness of 2D materials during mechanical exfoliation makes them prone to cracks and damage during transfer processes due to excessively strong interfacial adhesion between the growth substrate and 2D materials, leading to compromised structural integrity^[113]. To mitigate material cracking during transfer, it is essential to weaken the interfacial adhesion at the growth substrate/2D material interface. This section systematically reviews the strategies for reducing interfacial adhesion between growth substrates and 2D materials, thereby minimizing crack formation and enhancing material integrity.

3.1.1 Crack reduction via weakening growth substrate adhesion

(1) Chemical bubbling method

The bubble method is an innovative approach for modulating the interfacial adhesion between materials and growth substrates. It is a non-contact modulation technique that leverages the gradual formation of a gas layer at the interface through bubbles, thereby creating a thin gas gap between the material and the substrate. The resulting local stress field weakens the interfacial adhesion between the growth substrate and the 2D material, enabling uniform and damage-free separation. This method prevents tearing caused by mechanical stretching, achieving rapid, complete and high-quality exfoliation of 2D materials^[114–116]. Based on the bubble generation mechanism, the bubble method can be categorized into chemical and electrochemical approaches.

The chemical bubbling method is a technique that generates bubbles through chemical reactions. Rummeli and his colleagues successfully achieved the crack-free, large-area transfer of graphene using this method. As shown in Fig. 5a, the researchers employed a mixed solution of NH_4OH , H_2O_2 and H_2O in a volume ratio of 1 : 1 : 3, heating it to 80 °C. Under the action of NH_4OH , H_2O_2 rapidly decomposed to produce O_2 gas. Subsequently, a polymethyl methacrylate (PMMA)/graphene/growth substrate stack was

then immersed in the mixed solution. During this process, bubbles penetrated the interface between graphene and the growth substrate and gradually diffused across the entire interface. The localized compressive stress generated by the bubbles weakened the interfacial adhesion between the growth substrate and the 2D material, ultimately facilitating the complete delamination of the PMMA/graphene from the substrate^[117].

(2) Electrochemical bubbling method

The electrochemical bubbling method utilizes a 2D material/growth substrate as the cathode, forming a dual-electrode system with the anode. By applying a constant voltage or constant current between the cathode and anode, an electrochemical reaction occurs at the cathode interface, generating gas bubbles. In a typical electrochemical bubble-induced stress modulation process, a PMMA layer is first spin-coated onto the graphene/copper sample as a protective layer. Then, an aqueous solution of $0.05 \text{ mmol L}^{-1} \text{ K}_2\text{S}_2\text{O}_8$ is used as the electrolyte, and a DC voltage is applied between the PMMA/graphene/Cu cathode and a glassy carbon anode in the electrolytic cell (Fig. 5b). During voltage application, a hydrogen evolution reaction occurs at the graphene/Cu cathode electrode. H_2 bubbles form at the graphene/Cu interface, providing a mild yet persistent compressive stress that weakens the interfacial adhesion between graphene and the Cu substrate. Ultimately, the 2D graphene material was completely and crack-free delaminated from the copper substrate^[114]. Ren et al. discovered that when bubbles are used to modulate the adhesion between the growth substrate and the 2D material, they primarily modulate the stress state of the 2D material at the interface, inducing compressive stress and corresponding deformation (Fig. 5c). This approach effectively weakens the adhesion between the growth substrate and the 2D material. Notably, this method has been successfully extended to enable continuous synthesis of 2m-long graphene/epoxy resin/PET (Fig. 5d)^[116]. Cheng and his colleagues successfully exfoliated graphene with high crystallinity, no cracks, and high carrier mobility using the electrochemical

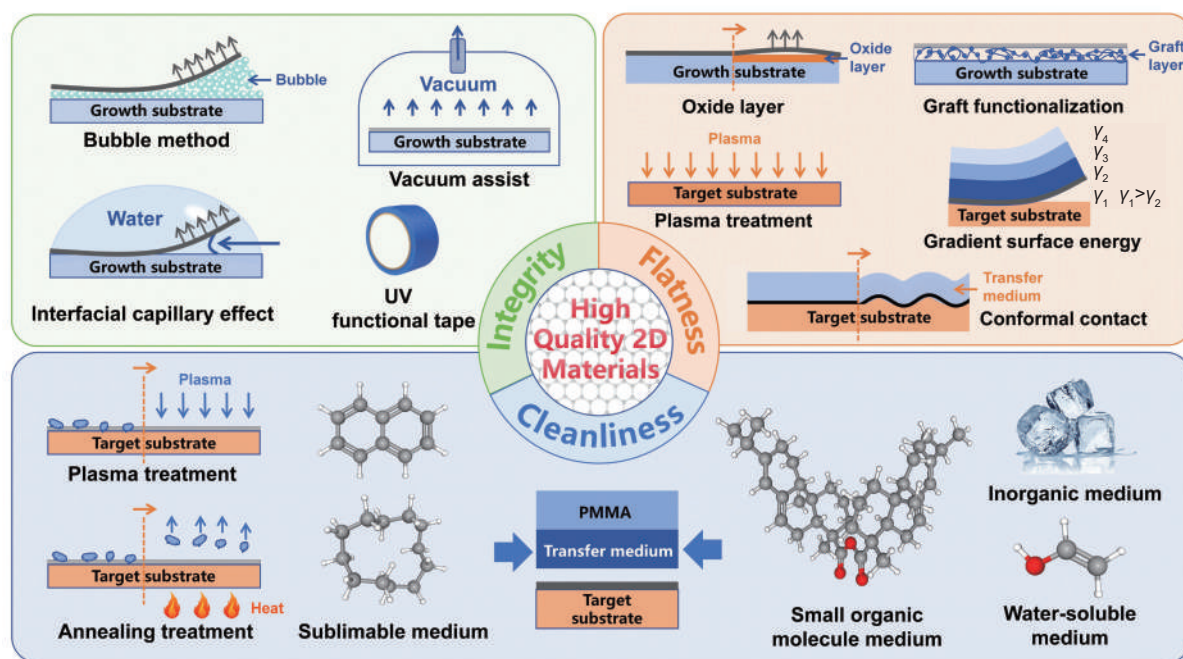


Fig. 4 Strategies for improving the quality of large-area 2D material transfer in terms of integrity, flatness and cleanliness

gas bubbling method. After growing graphene by CVD, they used a PMMA/graphene/Pt structure as the cathode in a constant-current electrolysis cell immersed in a NaOH electrolyte. Subsequently, a water reduction reaction occurred at the cathode, generating H_2 bubbles. The compressive stress introduced by these bubbles effectively weakened the interfacial adhesion between the Pt substrate and the graphene, allowing the PMMA/graphene layer to be successfully exfoliated within tens of seconds. This process avoided the stress-induced tearing issue caused by excessive interfacial adhesion in traditional mechanical exfoliation^[115].

(3) Interfacial capillary effect

The interfacial capillary effect is the core physical mechanism in liquid-assisted transfer processes. When a hydrophilic growth substrate and a hydrophobic 2D material come into contact with an aqueous solution, differences in surface energy drive water molecules to infiltrate the interface^[118]. Under the action of capillary forces, the pressure gradient generated by liquid infiltration can partially weaken the vdW forces and electrostatic interactions at the interface, thereby reducing the difficulty of mechanical exfoliation. This helps to avoid stress-induced tearing

caused by excessive interfacial adhesion in traditional mechanical exfoliation, thereby improving the integrity of 2D materials^[119–120]. At present, capillary-driven exfoliation techniques using water, organic solvents, and their mixtures have become an advanced, non-destructive, and efficient approach for exfoliating 2D materials.

The research conducted by Cao et al. reveals the hydrophobic properties of MoS_2 , in contrast to the hydrophilicity of the growth substrate. As shown in Fig. 5e, this difference in surface energy facilitates the penetration of water molecules beneath the 2D MoS_2 material. Once water molecules infiltrate the interface between the material and the substrate, their lubricating effect weakens the vdW forces and electrostatic interactions at the interface. Additionally, the polarity of water can alter the interfacial energy, reducing the adhesion between the material and the substrate. Furthermore, water molecules disrupt the pre-existing hydrogen bond network or chemisorption on the MoS_2 surface and form a dynamic hydration layer at the interface, further isolating the direct contact between the material and the substrate. This reduction in binding force allows the 2D MoS_2 material to be more easily detached from the growth substrate, thereby minimizing material damage and tearing while enhancing its

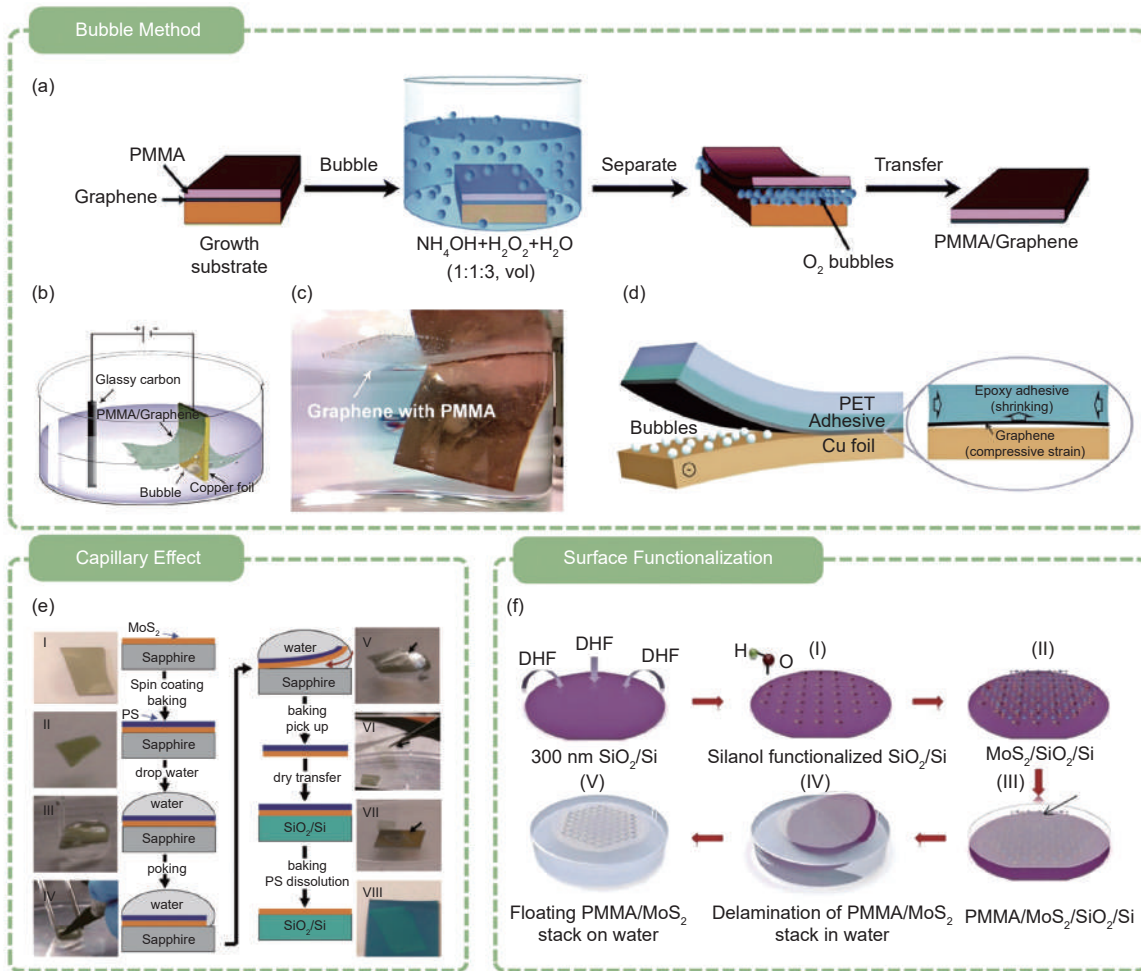


Fig. 5 Weakening the interfacial bonding strength between growth substrates and materials by diverse strategies to enhance transfer-process integrity. (a) Schematic diagram of the stripping process in which the chemical bubble method weakens the growth substrate and the bond between materials to improve the integrity of 2D materials^[117]. (b) Schematic diagram of the principle of the electrochemical bubble method to weaken the growth substrate and the bond between materials to improve the integrity of 2D materials and (c) optical picture of the stripping process^[114]. (d) Schematic diagram of the principle of reducing the binding force between growing substrates and materials by electrochemical bubble method^[116]. (e) Flow chart of the transfer of the capillary effect of water to weaken the growth substrate and the bond between materials to improve the integrity of molybdenum disulfide^[120]. (f) Process flow chart of the growth substrate surface functionalization to weaken the bonding force between the growth substrate and materials and improve the integrity of molybdenum disulfide^[121]. Reprinted with permission

structural integrity^[120]. Although deionized water is commonly used as the primary medium for polymer-supported wet transfer, its high surface tension and the low wettability of the substrate make it difficult for water molecules to penetrate the interface between the 2D material and the growth substrate. As a result, using deionized water as the transfer medium typically requires a prolonged period to achieve complete separation. To address this challenge, Zhou et al. proposed an innovative solution: mixing water with a low-polarity ethanol solution to reduce the surface tension of the transfer medium and enhance the wettability of the substrate. By introducing an ethanol-

water solution, the mixed solution successfully infiltrates the non-contact interface between the 2D material and the growth substrate, naturally generating a micro-scale capillary effect. This method not only enables rapid, damage-free delamination of the 2D material within 1 min, but also achieves the large-area transfer of a monolayer molybdenum disulfide (MoS_2) film with a diameter of ~ 5 cm ensuring a transfer integrity of up to 99%^[119].

(4) Oxide layer

Currently, introducing a uniform oxide layer between the growth substrate and the 2D material has proven to be an effective strategy to address the tear-

ing and defects caused by mechanical exfoliation. The formation of an oxide layer on the surface of the growth substrate effectively weakens the interfacial adhesion, shifting the binding force from electrostatic interactions to weaker vdW forces. Additionally, the smooth metal interface transforms into a rough metal oxide interface, ultimately reducing the adhesion between the 2D material and the growth substrate. This facilitates the complete exfoliation of the 2D material from the substrate surface, yielding high-quality, crack-free 2D materials. Liu et al. successfully constructed a uniform copper oxide layer covering the surface of a copper growth substrate using an ethanol/water mixed solution as a surface oxidant. This copper oxide layer significantly weakened the $d-\pi$ interactions between graphene and the copper interface, enabling the crack-free and intact transfer of a 10 cm graphene 2D material onto a silicon wafer^[113].

(5) Growth substrate surface functionalization

The surface functionalization strategy of the growth substrate is an interface adhesion modulation method for obtaining intact and crack-free 2D materials. By chemically modifying or physically treating the surface of the growth substrate, its surface energy, roughness, or hydrophilicity/hydrophobicity can be uniformly altered. This effectively weakens the adhesion between the 2D material and the substrate, enabling the acquisition of complete and crack-free 2D materials. Ahn et al. reported a hydroxyl-functionalized SiO_2/Si growth substrate, whose modified surface exhibited superhydrophilicity. This characteristic allowed the solvent to effectively penetrate the interface between the polymer-covered MoS_2 and the growth substrate with minimal mechanical force, thereby weakening the interfacial adhesion between the SiO_2/Si growth substrate and MoS_2 . As a result, an intact, crack-free MoS_2 thin film was obtained (Fig. 5f). Moreover, this method facilitates the transfer of continuous, large-area MoS_2 thin films onto conventional polymer films, which act as convenient substrates. This significantly simplifies the fabrication process of large-area ultrathin flexible devices

based on MoS_2 ^[121].

3.1.2 Tunable transfer medium for enhanced adhesion and easy release of 2D Materials

The adhesion and removal processes have opposite adhesion requirements for the transfer medium. When the medium adheres to the 2D material, a high adhesion force between the polymer and the 2D material is needed to prevent damage caused by wrinkling, misalignment or detachment. However, if the adhesion is too strong, the removal of the transfer medium becomes challenging, often leading to damage and tearing of the 2D material^[117]. Therefore, employing a strategy that segmentally modulates the interfacial bonding force between the transfer medium and the 2D material can effectively enhance the integrity of the 2D material, preventing cracks caused by either excessively weak or strong adhesion forces.

Ago et al. have recently developed a novel UV-light-responsive functional adhesive tape whose adhesion can be dynamically and intelligently controlled. This UV-controllable adhesive tape enables the transfer of high-quality, large-area monolayer graphene films (with carrier mobility exceeding $5000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at room temperature and sizes up to $50 \text{ mm} \times 50 \text{ mm}$) with 99% integrity, as well as other 2D materials such as MoS_2 , WS_2 and h-BN. During the removal of the transfer medium from the 2D materials, UV irradiation induces further crosslinking of polymer chains within the functional adhesive tape, significantly weakening the adhesion between the tape and the 2D material. This process allows the tape to be easily detached from the material surface, ensuring the integrity of the transferred materials^[65].

In summary, reducing the interfacial adhesion between the growth substrate and the material is an effective way to decrease the risk of cracks in 2D materials and improve their integrity. Currently, strategies to achieve this goal include: (1) Chemical/electrochemical bubble method: generating bubbles at the interface through chemical reactions or electrolysis, and using the local compressive stress produced by the gas layer to weaken the adhesion between the growth substrate and the material, thereby achieving rapid and

non-destructive exfoliation. (2) Interfacial capillary effect: Using the surface tension difference of water or ethanol mixtures, and through capillary penetration to weaken vdW forces and electrostatic interactions, thereby achieving efficient and non-destructive transfer of 2D materials; (3) Oxide layer modulation: Constructing a uniform oxide layer on the surface of the growth substrate to convert strong electrostatic interactions into weak vdW forces, promoting the complete exfoliation of 2D materials; (4) Surface functionalization of the substrate: changing the surface energy or wettability through chemical modification (such as hydroxylation) to reduce the interfacial adhesion; (5) Controllable transfer medium: Using UV-responsive adhesives to dynamically adjust the adhesion strength, maintaining high adhesion during transfer to prevent misalignment, and weakening the adhesion through UV crosslinking during removal to ensure material integrity. These strategies, by precisely controlling the interfacial adhesion between the growth substrate and the material, effectively reduce the cracks in 2D materials and significantly improve the transfer quality of 2D materials, laying a solid foundation for the preparation of high-performance devices.

3.2 Flatness enhancement via enhanced target substrate adhesion

The transfer of 2D materials from the transfer medium to the target substrate is often accompanied by interfacial bubbles, wrinkles and defects, which cause non-uniform adhesion at the 2D material/target substrate interface. This results in poor conformal contact and reduced flatness of the 2D material^[86,122–123]. To address the issue of non-uniform interfacial bonding, enhancing the interfacial adhesion between the 2D material and target substrate is critical, it can effectively improve the flatness of 2D materials.

3.2.1 Interfacial conformability improvement via enhanced target substrate adhesion

(1) Plasma treatment

In the transfer process of 2D materials, surface modification of the target substrate plays a crucial

role. Its primary purpose is to enhance the bonding strength between the 2D material and the substrate, thereby improving the flatness of the transferred material. Given that 2D materials typically have atomic-scale thickness, their interactions with the substrate (such as vdW forces and electrostatic forces) are relatively weak, making them susceptible to various factors such as surface roughness, chemical contamination, and surface energy. Therefore, appropriate surface modification of the target substrate before transfer is essential. This process optimizes interfacial properties, enhances the adhesion of 2D materials, reduces the formation of bubbles and wrinkles, and improves the interfacial flatness of the 2D material.

Lukosius and his colleagues investigated the effects of oxygen plasma treatment on the surfaces of different target substrates, including SiO₂ and Si₃N₄. They found that oxygen plasma treatment significantly enhanced the adhesion of graphene to these target substrates by removing carbon contamination and increasing surface activity. The possible interactions and modification models of SiO₂ and Si₃N₄ surfaces after oxygen plasma treatment are illustrated in Fig. 6a. Specifically, the oxygen plasma-generated reactive radicals and particles first bombard the grown target substrates, removing partial carbon contamination from the surface. As plasma treatment progresses, the process transitions from surface cleaning to surface activation, leading to an increase in surface energy. With the removal of carbon atoms from the surface, the bonding force between graphene carbon atoms and the target wafer atoms is enhanced, thereby improving the adhesion quality and smoothness of graphene. Experimental results indicate that the treated target substrates enable the transfer of continuous and wrinkle-free graphene, which is of great significance for the large-scale manufacturing of graphene-based electronic devices in the future^[124].

(2) Annealing

In the interface between 2D materials and the target substrate, 2D materials primarily adhere to the substrate through relatively weak vdW forces. This weak bonding force is insufficient to ensure the stable

and uniform fixation of large-scale few-layer 2D materials on the target substrate surface, which is essential for extensive micro/nano fabrication processes. Therefore, certain physical or chemical methods must be employed to enhance the interfacial adhesion energy and improve the material's flatness. High-temperature annealing is a widely used technique to enhance interfacial adhesion energy. Choi et al. significantly increased the adhesion energy between graphene and the SiO₂ substrate through rapid thermal annealing at 450 °C. At high temperatures, new bonds form at the interface, thereby increasing the bonding energy between graphene and the substrate, resulting in a smoother graphene surface^[125]. Similarly, research demonstrated that annealing h-BN/SiO₂ in the temperature range of 300 to 400 °C can significantly enhance the adhesion between the 2D material and the target substrate, achieving highly flatness h-BN. This is because, after annealing, h-BN undergoes slight deformation in response to the roughness of the substrate, leading to a tighter connection and improved conformity between h-BN and SiO₂^[122].

(3) Gradient surface energy (GSE)

In the transfer process of 2D materials, the surface energy of the target substrate significantly affects material adhesion, interface quality, and device performance. By modulating the surface energy of the target substrate, adhesion can be enhanced, interface conformity improved, and the formation of bubbles and wrinkles reduced, thereby increasing interface smoothness. According to thin-film adhesion theory, the interlayer transfer of thin films is primarily governed by the differences in surface energy among layers^[123]. When the target substrate exhibits higher surface energy, it demonstrates better wettability and stronger adhesion at the interface, making it a more effective “receptor” for the thin film. Therefore, to ensure the key functionality of wafer-scale 2D material integration and achieve reliable adhesion and non-destructive release, it is essential to carefully design the surface energy of the transfer medium and the target substrate. Peng et al. designed a multifunctional 3-layer transfer medium with a progressively decreasing

surface energy from the target SiO₂/Si wafer to the top polydimethylsiloxane (PDMS) layer (Fig. 6b). The intrinsic relationship between GSE design and interface adhesion modulate lies in precisely tuning the surface energy difference between the transfer medium and the target substrate. This enables reliable adhesion and non-destructive release of graphene from the growth substrate to the target substrate. Specifically, the surface energy of the 3-layer transfer medium gradually decreases from the target SiO₂/Si wafer to the top PDMS layer. This design allows the high-surface-energy target substrate to better wet and firmly adhere to the graphene, while the low-surface-energy PDMS layer ensures that the final release does not damage the graphene. According to thin-film adhesion theory, this gradient distribution of surface energy directly influences the spreading coefficient and fracture strength of the interface, thereby enhancing interface adhesion. This approach avoids the wrinkling issues commonly observed in conventional wet-transfer processes and enables the transfer of high-quality, high-flatness 2D materials^[126].

(4) Conformal contact

In the 2D material transfer process, achieving conformal contact between the material and the target substrate is an effective strategy to enhance the adhesion of the target substrate. Conformal contact ensures that the 2D material closely adheres to the target substrate, preventing the formation of bubbles, wrinkles, or inclusions. This improves the interfacial adhesion, enhances the flatness of the 2D material, and helps maintain its excellent properties. Liu et al. demonstrated that introducing oxygen-containing hydroxyl volatile molecules or polymers with a low glass transition temperature (T_g), such as polypropylene carbonate (PPC), into PMMA can induce deformation of the supporting film during the heating process. This enables conformal contact between graphene and target substrates with different surface morphologies (Fig. 6c-d). This controllable conformal contact enhances the interfacial adhesion between graphene and the target substrate, allowing the direct transfer of 2D materials without wrinkles and achiev-

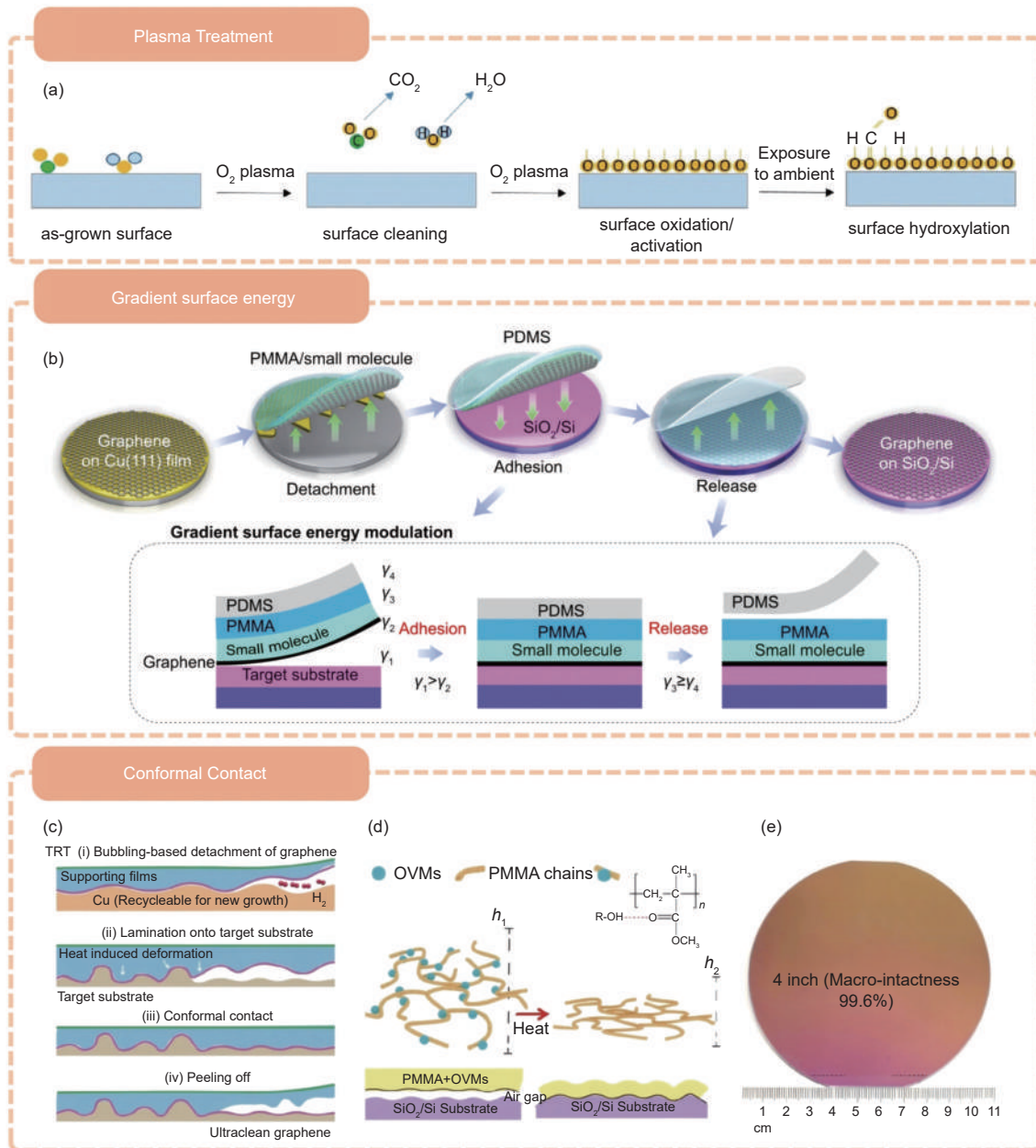


Fig. 6 Enhancing substrate-material interfacial adhesion via multimodal strategies for surface flatness optimization. (a) Schematic diagram of 2D material transfer process in which plasma treatment enhances the binding force of target substrate and material to improve flatness^[124]. (b) Schematic diagram of graphene material laminations where gradient surface energy is modulated to enhance the target substrate and material bonding to improve flatness^[126]. (c) Schematic diagram of a process in which the conformal contact strategy enhances the binding force of the target substrate and material to improve flatness, and (d) schematic diagram of the effect and (e) the optical image of a 4-inch 2D material with 99.6% integrity obtained by the preparation^[86]. Reprinted with permission

ing a 99.6% integrity transfer of a 4-inch 2D material (Fig. 6e). The optimization of interfacial adhesion benefits from the steric hindrance effect of OVMs, which increases the chain spacing of PMMA. During the heating process, the embedded volatile molecules evaporate, leading to the rearrangement of PMMA chains. This, in turn, drives polymer deformation, en-

abling conformal contact between graphene and the SiO₂/Si target substrate, ultimately achieving a high level of flatness^[86].

3.2.2 Interfacial contact improvement via stress relief

The release of material stress is crucial for optimizing interfacial contact. Stress typically arises from

mechanical stretching, thermal expansion, or interfacial mismatch during the transfer process. If not released in a timely manner, it may lead to wrinkles, cracks, or poor interfacial contact, all of which can affect the flatness and intrinsic properties of the 2D material^[127–128]. Effectively releasing stress can enhance the interfacial contact between the 2D material and the substrate, ensuring high flatness after transfer.

Stress may be introduced in the material surface during the transfer process, which can affect the flatness and intrinsic properties of the 2D materials. Annealing treatment is a commonly used optimization method aimed at releasing stress, improving interfacial contact, and enhancing material flatness, thereby restoring or enhancing the physical and chemical properties of 2D materials. Choi et al. conducted an in-depth study on the effects of different annealing methods on the adhesion energy between graphene and the target SiO_x/Si substrate. They compared rapid thermal annealing (RTA) and vacuum annealing (VA) techniques and found that the adhesion energy of untreated graphene on the SiO_x/Si substrate was approximately 2.978 J m⁻². After RTA treatment, the adhesion energy significantly increased to 10.09 J m⁻², while VA treatment further enhanced the adhesion energy to 20.64 J m⁻². These findings reveal that the adhesion strength between graphene and the target substrate can be significantly improved through appropriate annealing processes, effectively enhancing interfacial contact and improving the flatness of 2D materials^[74]. Peng et al. utilized thermal annealing techniques to release residual stress in MoS₂ 2D materials, successfully achieving close contact between graphene and the TEM hole substrate. This substantially improved interfacial adhesion while maintaining the high quality and intrinsic properties of the material^[127].

In summary, when 2D materials are transferred to the target substrate, the problems of non-uniform adhesion and surface flatness degradation caused by interface bubbles, wrinkles and contaminants can be significantly improved by enhancing the adhesion strength between the target substrate and the 2D ma-

terials and adopting cleaning strategies. Currently, the adopted strategies include: (1) Plasma treatment: utilizing oxygen plasma to remove contaminants on the substrate surface and enhance surface activity, thereby strengthening the adhesion between the target substrate and the 2D materials and achieving wrinkle-free transfer. (2) High-temperature annealing: enhancing the adhesion between the target substrate and the 2D materials through the annealing process, thereby improving the material's flatness. (3) GSE design: constructing a three-layer transfer medium with gradually decreasing surface energy, using a high surface energy substrate to enhance wettability, while a low surface energy layer ensures damage-free release, achieving high-quality transfer. (4) Conformal contact optimization: Introducing a low glass transition temperature polymer (eg. PPC) into the transfer medium, inducing deformation of the transfer medium through heating to make the 2D materials closely adhere to the target substrate, thereby improving its flatness. (5) Stress release optimization: using rapid thermal annealing or vacuum annealing to release residual stress during the transfer process, further enhancing the bonding strength between the target substrate and the 2D materials. These strategies systematically improve the flatness of large-area 2D materials on the target substrate surface, laying a solid foundation for their large-scale application.

3.3 Cleanliness enhancement strategies

The detachment of 2D materials from the transfer medium surface often leaves polymer residues on the material. Simultaneously, incomplete removal of the transfer medium during cleaning or dissolution can also lead to residual contamination^[61,109]. Furthermore, metal/metal oxide particles are introduced during the etching of metal substrates, and metal ion contaminants are incorporated from the etching agents^[111]. These pollutants significantly degrade the cleanliness of 2D materials. To address residue contamination and enhance material cleanliness, this section first reviews the direct removal of residual polymers through annealing or plasma treatment, and then discusses strategies to prevent polymer residues by optimizing

transfer media. These approaches effectively mitigate surface residue contamination and improve the cleanliness of 2D materials.

3.3.1 Residual polymer ablation

For 2D materials with polymer residues on the surface after removing the photoresist, physical cleaning can be an effective supplementary cleaning process that efficiently removes residual polymers and improves the cleanliness of the material. Physical cleaning after transfer typically requires extreme conditions to decompose and evaporate polymer residues, such as annealing, plasma treatment, electron beam treatment and Joule heating^[64,125,129–130]. The most commonly used physical cleaning method is high-temperature annealing. Fang et al. removed the photoresist residues on the graphene surface after annealing in a vacuum at temperatures above 300 °C. After annealing at 300 °C, the surface roughness of the graphene material shows only 0.4 nm, even lower than that of any freshly transferred monolayer graphene before fabrication. After increasing the annealing temperature to 400 °C, the roughness further decreased to 0.15 nm, and the photoresist contamination on the graphene surface was completely removed^[64]. Geim et al. successfully removed surface contaminants by annealing in a H₂ atmosphere at 250 °C^[131]. Chiu et al. reported a systematic study on the effect of annealing on PMMA residues. The study found that PMMA residues on graphene undergo 2-step decomposition: PMMA residues exposed to air (PMMA-A) require a lower decomposition temperature, while PMMA in contact with the graphene (PMMA-G) requires a higher decomposition temperature. This suggests that the interaction with graphene may affect the decomposition of PMMA chain segments. Stronger interfacial interactions typically require higher decomposition temperatures^[132].

In addition to annealing, plasma treatment is also widely used to remove polymer residues after transfer. Duesberg et al. reported a cleaning method for removing PMMA residues from graphene surfaces using O₂ and H₂ plasma treatment. Plasma effectively removes PMMA residues and significantly enhances the elec-

trical conductivity of graphene, as well as the electron and hole field-effect mobility^[129]. Besides the traditional O₂ and H₂ plasma treatments, O. Renault and others developed a mild plasma treatment method using high-density H₂ and H₂-N₂ plasma. By precisely modulating the ion energy within the range of 11–14 eV, high-density H₂ and H₂-N₂ plasma selectively removes polymer residues from monolayer graphene without causing significant damage to the graphene surface^[133].

3.3.2 Residual polymer prevention by transfer medium optimization

High molecular weight polymer transfer media have long polymer chains with strong intermolecular vdW adsorption, which reduces the chain mobility and increases entanglement between overlapping chains, making complete depolymerization of high molecular weight polymers difficult^[79]. Therefore, transfer media with shorter chains and lower molecular weight have been studied in recent years. The lower molecular weight weakens the vdW forces within the polymer, facilitating depolymerization. Sunae Seo et al. reported PMMA transfer media with different average molecular weights. Graphene surfaces transferred by PMMA with a lower average molecular weight showed no polymer residue, with clean surfaces of the 2D material, resulting in enhanced field-effect mobility and reduced hole doping, without the need for post-annealing of the lower average molecular weight PMMA^[61].

Compared to polymers, organic small molecules have significantly lower sublimation temperatures and higher solubility in organic solvents, making them easier to remove. They can be used as a clean transfer medium in the transfer of high-purity large-area 2D materials^[76,134]. For example, pentylbenzene (C₂₂H₁₄), a polycyclic aromatic hydrocarbon, can be used as a clean transfer medium for graphene. After transferring graphene to the target substrate surface, pentylbenzene can be removed through thermal annealing at 250 °C or by washing with an organic solvent such as tetrahydrofuran (THF). Due to its low sublimation temperature, pentylbenzene is completely removed,

resulting in high-purity graphene^[76].

Cyclododecane (CD) is a cyclic hydrocarbon that is also used as an organic support material for graphene transfer. This non-toxic and environmentally friendly organic compound is solid at room temperature, and its vapor pressure reaches 1.33 kPa at 100 °C. Therefore, it can sublime completely when exposed to air, leaving no polymer residue on the surface of 2D materials (Fig. 7a-d), making it suitable for the clean transfer of 2D materials. Furthermore, after recrystallization, cyclododecane exhibits stronger interconnectivity, forming a more robust protective layer that effectively prevents cracking of the 2D materials during the transfer process^[62]. Similarly, Chen et al. developed a clean transfer method based on the small polycyclic hydrocarbon naphthalene as a transfer medium, which can be easily removed by sublimation under atmospheric pressure, resulting in high cleanliness of the obtained graphene surface^[63].

Due to the exclusion of organic solvents, water-soluble polymer media show great potential in the clean transfer of 2D materials. Booth et al. used water-soluble polyvinyl alcohol (PVA) as a transfer medium to achieve the clean transfer of 2D materials. They first oxidized copper foil with graphene in deionized water to decouple the graphene from the copper substrate. Then, a thermal roll laminator was used to laminate the PVA thin film on top of the graphene/copper foil, and the graphene was mechanically exfoliated from the copper onto the PVA film. After laminating the PVA/graphene film onto the target substrate surface, deionized water was used to wash away the PVA, resulting in clean graphene 2D material. As no chemical etchants or organic solvents are required, these water-soluble transfer media are highly environmentally friendly and cost-effective for the clean transfer of 2D materials^[135].

Due to their smaller molecular weight, organic molecules have weaker adhesion to materials compared to polymers, which makes them more easily removed by organic solvents, resulting in a cleaner transfer. However, because the transfer medium causes disturbances and provides insufficient tensile

protection to the 2D materials during the transfer process, the weak adhesion often leads to the formation of more cracks. Therefore, Ren et al. used rosin organic molecules as a buffer layer, inserting them between the PMMA polymer film and the 2D material, creating a bilayer transfer medium. The rosin layer at the bottom isolates the polymer from the WS₂, achieving a clean transfer without polymer residue, while the PMMA layer at the top enhances the strength of the transfer layer. Unlike the use of PMMA alone as a transfer medium, which results in polymer residues, the PMMA/rosin bilayer transfer medium leaves no polymer residue (Fig. 7e-f). Based on this bilayer transfer medium strategy, they successfully achieved high-purity and high-integrity transfer of 2D materials^[136]. Zheng et al. successfully achieved the clean transfer of 2D materials using polyphthalaldehyde (PPA) and hydrophobic polystyrene (PS) as polymer support layers. As shown in Fig. 7g, the PS layer can directly peel the 2D materials from a hydrophilic substrate through a water intercalation technique, while the PPA layer serves to protect the 2D materials from solution interference and ensures the structural integrity of the materials during the removal of the PS layer. Subsequently, the PPA layer can be completely removed by thermal annealing at 180 °C^[137].

Kong et al. proposed a clean transfer technique using paraffin as the transfer medium. Compared to PMMA transfer medium, the residue left by paraffin on the graphene surface is significantly reduced because it does not contain carbonyl functional groups that can react with electrophilic or nucleophilic reagents. In addition, under low-pressure conditions, the wax layer can evaporate during annealing treatment at 140-200 °C, enabling clean transfer of the 2D material without the need for organic solvents. Furthermore, during the heating process, paraffin, due to its high coefficient of thermal expansion, undergoes thermal expansion. This expansion can stretch the underlying graphene film, effectively reducing wrinkles in the graphene during the transfer process^[128].

Ly et al. developed an auxiliary transfer technique that uses ice as the sole medium. In this process,

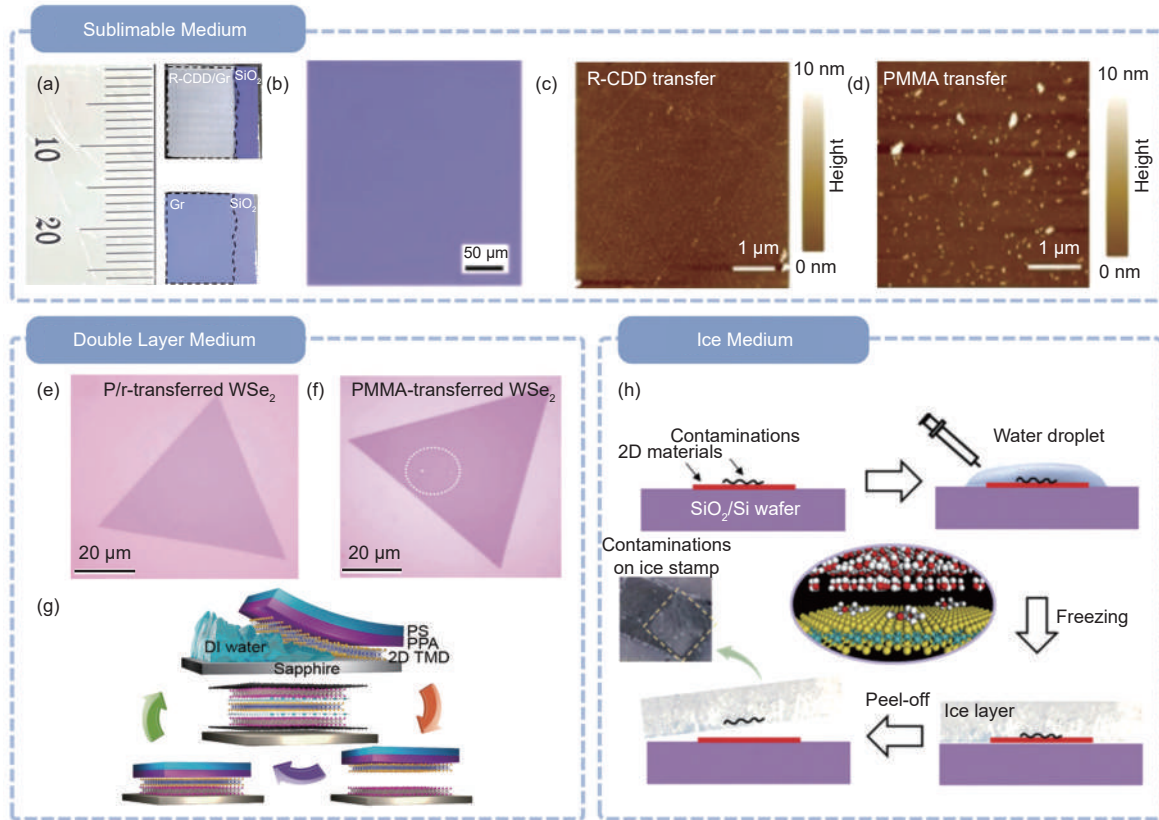


Fig. 7 Transfer media for enhanced interfacial cleanliness. (a-b) Optical images of graphene cleanliness improved with a sublimable cyclododecane transfer medium and atomic force microscope images of (c) cyclododecane as transfer medium and (d) PMMA as transfer medium^[63]. (e) Optical images of improving the cleanliness of WS₂ materials using PMMA/rosin double layer transfer medium and (f) PMMA single layer transfer medium^[136]. (g) Transfer process flow chart of using double-layer transfer medium to improve the cleanliness of WS₂ materials^[137]. (h) Schematic diagram of the process of treating MoS₂ surface residues with ice to improve cleanliness^[112]. Reprinted with permission

ice demonstrates significant adhesion to the surface of 2D materials through vdW forces and hydrogen bonds. The study reveals that through adjusting the environmental temperature, the bonding strength between ice and the 2D materials can be effectively modulated, making it possible to separate the materials from the growth substrate. Subsequently, the ice layer carrying the 2D materials can form a stable connection with the target substrate, enabling the successful transfer of the material. After the transfer is completed, the ice layer can be easily removed by heating, leaving no residue. Since the entire transfer process completely avoids the use of polymers, chemical etchants, and organic solvents, the 2D materials obtained through the ice transfer technique maintain high quality and excellent cleanliness. Furthermore, ice can serve as a non-damaging residue removal method. As shown in Fig. 7h, the modulated adhesion force of ice successfully removed residuals from the

MoS₂ surface^[112].

3.3.3 2D materials-2D materials interlayer cleanliness enhancement

The clean interface between 2D materials is crucial for device performance. A clean interface ensures good contact between materials, reduces interface defects and impurities, and optimizes carrier separation and migration. However, the strong adhesive forces between 2D materials can lead to the accumulation of interlayer contaminants, forming composite bubbles composed of air, water and hydrocarbons, which may cause phenomena such as impurity scattering^[138–141]. For example, the interaction between bilayer graphene and residual interlayer polymers can suppress phonon transport in graphene, significantly reducing the thermal conductivity of graphene films^[142]. Additionally, during the fabrication of multilayer 2D materials using the stacking transfer method, the number of residual polymers rapidly increases due to the stacking

effect, which has a more severe impact on the transport properties of the multilayer materials^[143].

Programmed thermal annealing is a commonly used method to eliminate interlayer bubbles in 2D materials. Deng et al. developed a thermal annealing process that effectively reduces the formation of composite bubbles. Using AFM and high-resolution surface current imaging techniques, they systematically studied the formation and evolution of bubbles at the interfaces of transferred 2D materials and their vdW heterostructures. After annealing, the number of bubbles between the heterojunction layers was significantly reduced. During the annealing process, bubbles tend to migrate across the interface in flat and clean areas or move from the center to the edges, thus eliminating the interface bubbles and significantly improving the cleanliness of the heterojunction interface^[144]. Park et al. used programmed vacuum stack (PVS) technology to achieve a significant improvement in interlayer cleanliness of 4-layer MoS₂ films. When transferring MoS₂ films in a vacuum environment, the cleanliness of the material surface is higher, which helps improve the interface cleanliness when stacking multilayer materials^[145].

In summary, by eliminating polymer residues, metal particle contamination, and interlayer impurities during the transfer process of 2D materials, the surface and interlayer cleanliness of the materials can be significantly improved. The key strategies for enhancing cleanliness include: (1) Removing residual polymers: Decomposing the residual transfer medium through high-temperature annealing or plasma treatment to improve the surface cleanliness of 2D materials. (2) Optimizing the transfer medium: Selecting low-molecular-weight materials (easily depolymerizable), easily sublimable organic small molecules, or water-soluble polymers, and achieving residue-free transfer through thermal annealing or solvent cleaning; innovatively designing double-layer transfer media that combine mechanical strength with easy removability to avoid the polymer residue problem of single-layer media. (3) Ice-assisted transfer technology: Utilizing the reversible adhesion properties of

hydrogen bonds and vdW forces of ice, without using chemical reagents throughout the process, to achieve large-area, non-destructive clean transfer of 2D materials. (4) Strengthening interlayer cleaning: Using programmed thermal annealing to promote the migration and ablation of bubbles at the heterojunction interface, or employing PVS to reduce interlayer contaminants, significantly enhancing the interlayer cleanliness of 2D materials. These strategies, through the synergistic effects of physics and chemistry, effectively reduce surface and interlayer contaminants of 2D materials, thereby improving their surface and interlayer cleanliness.

Furthermore, to assist researchers in better selecting appropriate transfer techniques, we have summarized the advantages and limitations of adhesion control technologies in large-area 2D material transfer (Table 2), with the aim of facilitating the construction of large-area 2D materials characterized by high integrity, flatness, and cleanliness.

4 Recipe of large-area 2D material transfer for application requirements

In previous discussions, we delved into the roles of various interfacial binding forces during the transfer process of large-area 2D materials and elucidated the critical impact of these forces on the quality and stability of material interfaces, as well as the ultimate performance of devices^[146]. Additionally, we summarized strategies for modulating specific interfacial binding forces to achieve high-quality, damage-free material transfer. However, the core of optimizing the transfer process lies in adjusting and modulating process parameters according to different application requirements. Without meticulous design of these parameters, it would be challenging to meet the application demands in terms of efficiency, quality, uniformity, and controllability.

This section focuses on the design of transfer process parameters based on application requirements, aiming to optimize the transfer methods, growth substrates, target substrates, transfer media and parameter configurations of process conditions to achieve effi-

Table 2 Advantages and limitations of various adhesion regulation strategies

	Strategy	Advantage	Limitation
Integrity enhancement	Chemical bubbling method	<ul style="list-style-type: none"> • Simple operation • High universality • Large-area scalability 	<ul style="list-style-type: none"> • Chemical damage risk • Solvent residue
	Electrochemical bubbling method	<ul style="list-style-type: none"> • High controllability • Low damage 	<ul style="list-style-type: none"> • Restricted to electrically conductive substrates • Complex equipment
	Interfacial capillary effect	<ul style="list-style-type: none"> • Mechanical stress-free transfer • Clean interface 	<ul style="list-style-type: none"> • Non-uniform capillary force distribution • Requires precise control of wettability
	Oxide layer	<ul style="list-style-type: none"> • Suitable for large-area transfer 	<ul style="list-style-type: none"> • Residual oxide layer • Complex process
	Tunable transfer medium	<ul style="list-style-type: none"> • High integrity 	<ul style="list-style-type: none"> • Challenge in large-area uniformity • Difficulty in controlling adhesion force
Flatness enhancement	Plasma treatment	<ul style="list-style-type: none"> • Suitable for large-area transfer • Rapid and efficient • High uniformity 	<ul style="list-style-type: none"> • Substrate damage risk
	Annealing	<ul style="list-style-type: none"> • High universality 	<ul style="list-style-type: none"> • High temperature damage risk • Difficulty in gradient control
	Gradient surface energy	<ul style="list-style-type: none"> • No mechanical stress 	<ul style="list-style-type: none"> • Environmental sensitivity • Media cannot be reused
	Conformal contact	<ul style="list-style-type: none"> • High flatness • Suitable for non-destructive transfer of complex micro nano structures 	<ul style="list-style-type: none"> • Large-area limitations • Media cannot be reused
Cleanliness enhancement	Residual polymer ablation	<ul style="list-style-type: none"> • High universality • Efficient 	<ul style="list-style-type: none"> • Strict atmosphere • High temperature damage risk • Thermal stress causes structural defects
	Transfer medium optimization	<ul style="list-style-type: none"> • Reduces residual risks 	<ul style="list-style-type: none"> • Scalability and efficiency limitations

cient transfer and interface quality modulate of large-area 2D materials for different performance requirements of devices in various application scenarios (such as high conductivity, high optical quality, high specific surface area, etc.)^[147]. For instance, in the field of electronic devices, high cleanliness and precise alignment are core requirements, making dry transfer methods (eg. PDMS/graphene stamps) the preferred choice due to their solvent-free contact and high-precision alignment capabilities. In contrast, for energy storage devices, low cost and large-area fabrication are key, giving direct coating techniques an advantage. Moreover, for the special requirements of heterostructure material stacking and flexible devices, techniques such as programmed vacuum stacking and roll-to-roll transfer have shown unique application potential^[148]. This section systematically summarizes the selection of process parameters for different application scenarios (Table 3), aiming to provide theoretical guidance and practical references for the scalable preparation of 2D materials in various application contexts.

5 Conclusion and outlooks

This paper provides a comprehensive review of the types, quantification methods, and characteriza-

tion techniques of interfacial adhesion forces in the large-area transfer of 2D materials. It delves into how to precisely modulate the interfacial adhesion forces between 2D materials, the growth substrate, the target substrate, and the transfer medium during the transfer process. The types of interfacial forces involved in the transfer of 2D materials include vdW forces, covalent bonds, electrostatic forces and hydrogen bonds. Achieving efficient, damage-free and high-quality transfer of large-area 2D materials is a key objective. By precisely modulating the interfacial adhesion forces between the 2D material and the growth substrate, target substrate, and transfer medium, while also modulating the stress between the interfaces, it is possible to effectively improve the integrity, flatness, and cleanliness of the material during the transfer process. This helps to prevent wrinkles, tears, and defects in the material, thereby promoting the widespread application of 2D materials in electronics, optoelectronics, flexible electronics, and quantum devices.

The continuous exploration of the unique properties of 2D heterostructures has highlighted the need for their large-scale application in fields such as electronics and optoelectronics. This requires the development of transfer techniques that achieve high cleanli-

Table 3 Transfer recipe of large-area 2D materials based on application requirements

Application	Technical requirements	Technology	Growth substrate	Target substrate	Medium	Modulation strategies
Flexible electronic devices	<ul style="list-style-type: none"> • Low-temperature transfer • Flexible substrate • Without impurities or residues 	Roll-to-roll	Flexible polymer	Flexible polymer	EVA	Substrate surface functionalization
Optoelectronic devices	<ul style="list-style-type: none"> • High purity • High flatness 	Wet transfer	h-BN	Glass	PMMA	Electrochemical bubble/Substrate surface modification
Electronic devices	<ul style="list-style-type: none"> • High cleanliness • Multilayer stacking • Ultra-smooth interface 	Dry transfer	SiO ₂ /Si	h-BN/SiO ₂ /Si	PDMS/Graphene	Substrate surface functionalization
Optical devices	<ul style="list-style-type: none"> • High optical quality • High interface cleanliness • Large-area uniformity 	Dry transfer	Sapphire (Al ₂ O ₃)	Si	PDMS	Substrate surface functionalization
Catalysis	<ul style="list-style-type: none"> • High integrity • High cleanliness • Scalability and simplicity 	Wet transfer	SiO ₂ /Si	Conductive substrate	PMMA	Interfacial capillary effect/Substrate surface modification

ness, precision, structural integrity, repeatability, and fully automated large-area processing, which are essential for advancing their practical use and performance in devices.

(1) Integration of theoretical modeling and large-scale transfer: One promising method for large-scale 2D material transfer is the roll-to-roll (R2R) process^[149]. However, this technique is often associated with mechanical contact between graphene and the carrier film, or between graphene and the flexible substrate. During the rolling transfer process, mechanical compression can lead to localized contact pressure, which may cause material damage, impairing the material's integrity and consequently reducing its performance in application. By refining interfacial adhesion force modulate, optimizing peeling strategies, and implementing intelligent detection and feedback modulate, the uniformity and yield of large-area transfer can be further improved. In terms of large-scale production, intelligence and automation will be key directions.

Machine learning (ML) holds great potential in bridging the knowledge gap in CVD growth and transfer of 2D materials. For instance, it can be used to predict the impact of substrates on 2D material nucleation and growth, control the growth of monolayer and multilayer structures, and understand grain boundary formation and defect introduction. By integrating machine learning with experimental and simulation data, researchers can gain deeper insights into these complex processes, facilitating the optimization

of growth and transfer strategies. Liu et al. employed reactive molecular dynamics (MD) simulations to investigate the CVD growth of MoS₂ from MoO₃ and S precursors, followed by the use of a machine-learning approach based on feedforward neural networks to identify key reaction mechanisms^[150]. Compared to traditional ab initio calculations, ML-driven predictions not only offer lower computational costs and faster processing speeds but also enable the rapid extraction of valuable insights from large datasets, providing an efficient tool for advancing CVD growth and transfer technologies. To address defects arising during the growth and transfer processes, Sergei V. Kalinin et al^[151]. developed a deep neural network-based workflow that achieved excellent results. They obtained high-quality atomically resolved STEM images of graphene and constructed three types of training datasets: defect-free lattices, vacancies and dopant atoms. Using a deep learning model, they effectively classified and localized defects in graphene and further analyzed their chemical structures and evolution processes, providing value for improving both the quality of the material and the model construction.

Although research in this field has been steadily increasing in recent years, machine learning-driven studies still offer vast opportunities for exploration, particularly in CVD growth and subsequent 2D material transfer processes. The integration of machine learning algorithms for parameter optimization and the development of intelligent modulation systems will enable efficient and stable continuous transfer. In

the future, with continuous improvements in interfacial adhesion regulation strategies and advancements in intelligent transfer technologies, the R2R transfer process is expected to overcome material damage caused by mechanical contact, achieving high-quality, large-area 2D material transfer. This technology holds tremendous potential to further drive the applications of 2D materials in flexible electronics, optoelectronic devices, and renewable energy. Additionally, it will contribute to the large-scale manufacturing of next-generation high-performance devices and lay a solid foundation for future technological innovations.

(2) Automated transfer system and interfacial adhesion optimization: automated transfer equipment can significantly improve transfer efficiency and quality. Through high-precision optical imaging and automated operations, human error and material damage are minimized. Automated transfer systems, using high-precision robotic arms and optical imaging systems, enable precise transfer of 2D materials. For example, the massive transfer technology developed by Fudan University utilizes PDMS molds with micropillar patterns, enabling high-density, high-yield transfer of 2D materials at the wafer level^[119]. Additionally, some systems are equipped with high-magnification microscopes and high-definition CCD cameras, which use computer-assisted positioning to ensure precise alignment of the materials. Automated transfer equipment can operate in modulated environments, such as performing dry transfer in high-humidity conditions, minimizing material damage and wrinkle formation. This environmental modulate not only improves transfer success rates but also enhances the electrical performance of the materials.

Accurate force field descriptions are crucial for revealing the mechanisms of material growth in the CVD synthesis and transfer of 2D materials. The introduction of machine learning provides new solutions to these challenges. For example, Vashishta et al. developed a deep neural network model to generate the force fields required for MD simulations. The model significantly improved prediction speed and was validated against true values (test set) generated

from DFT simulations, demonstrating its accuracy. This neural network-based force field enables the study of CVD growth involving multiple competing chemical reactions and mechanisms^[152,156]. This approach can not only be used to study existing material systems but can also be extended to other heterostructure studies that are computationally challenging^[153,157]. For different materials and target substrates, machine learning can analyze data from previous transfers to assess interfacial adhesion forces, peeling effects, and defect distribution under different transfer conditions. By doing so, it can help identify the optimal process parameters and enable adaptive modulate, improving transfer quality and consistency. This data-driven optimization strategy reduces trial-and-error costs and clarifies the intertwined process conditions and device design parameters during the transfer and device fabrication processes. By integrating different interfacial adhesion force mechanisms and parameters, the modules through machine learning method can distinguish the impact of each step on the subsequent one, enabling the selection and optimization of frameworks to evaluate the effects of each process step or design feature^[154]. This accelerates the application of different materials on various substrates.

(3) Fast and efficient characterization: for the characterization of transferred samples, although AFM morphological measurement is a non-destructive, versatile, and relatively simple method that can effectively evaluate the surface cleanliness and morphological features of the sample, this technique still has some limitations. First, the scanning range of AFM is small, meaning it can only analyze localized areas, making it difficult to reflect the overall uniformity of large-area samples. Additionally, AFM primarily provides surface morphology information, which makes it challenging to directly detect internal structural defects, atomic-level point defects, or adsorbed impurities. Furthermore, AFM measurements can be influenced by factors such as probe contamination, scanning modes, and operational conditions, which may introduce errors.

Therefore, to comprehensively assess the quality and cleanliness of post-transfer samples, it is necessary to combine multiple characterization techniques^[155]. For instance, Raman spectroscopy can be used to analyze the doping state, crystal quality, and potential structural defects of 2D materials; transmission electron microscopy (TEM) offers higher-resolution structural information and can detect internal crystal quality and defect distribution. Additionally, scanning electron microscopy (SEM) is useful for observing larger surface areas, while contact angle measurement can indirectly assess the surface cleanliness and hydrophilicity/hydrophobicity changes of the sample. Moreover, there is a need to design large-area, high-speed characterization techniques for 2D materials to accommodate the large-scale production and device fabrication of these materials.

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