Strain engineering of two-dimensional materials: Methods, properties, and applications

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Abstract
Two-dimensional (2D) materials have attracted extensive research interests due to their excellent properties related to unique structure. Strain engineering, as an important strategy for tuning the lattice and electronic structure of 2D materials, has been widely used in the modulation of physical properties, which broadens their applications in flexible nanoelectronic and optoelectronic devices. In this review, we first summarize the methods of inducing strain to 2D materials and discuss the advantages and problems of various methods. We then introduce the strain-induced effects on optical, electrical, and magnetic properties, together with the phase transition of 2D materials. Finally, we illustrate the potential applications of strained 2D materials and further look forward to their opportunities and challenges in practical applications in the future.

KEYWORDS
2D materials, photodetector, piezoresistive effect, strain engineering, strain sensor

1 INTRODUCTION

Since graphene was successfully exfoliated in 2004, two-dimensional (2D) materials have become the hotspots over the past decade due to their excellent optical, electrical, magnetic, mechanical, and thermal properties. 2D materials usually possess ultrahigh carrier mobility and field-effect switching ratio, which make them ideal channel materials for field-effect transistors (FETs) and digital logic transistors. In addition, 2D materials with appropriate and tunable direct band gap have achieved extremely efficient photon absorption, photon emission, and photoelectric conversion and then have been widely used in the field of optical and optoelectronic devices. Based on the characteristics of flexibility and transparency, 2D materials can be used in the emerging research areas such as transparent display screen and wearable electronics. Recently, in order to further meet more demands and expand the application fields of 2D materials, various techniques have been exploited to modulate their properties, including doping, alloying, inducing defect, forming van der Waals heterostructure, electrostatic regulation, and strain engineering. Among them, strain engineering is an effective technique that changes the lattice and electronic structure and thus modulates various properties of 2D materials.

Strain engineering has been widely used in the semiconductor industry in the last few decades. For example, when the uniaxial or biaxial tensile strain is applied to the channel of a silicon transistor, the electron mobility (or hole mobility) can be greatly enhanced. However, traditional semiconductor bulk single crystals only withstand very limited strain, which largely limit the further application of strain modulation. Compared with bulk materials, 2D materials have stronger deformation capacity and they can
withstand greater elastic strain without fracture, which show great potentials in strain engineering.

For example, most graphite materials will break when the strain is up to 0.1%, while monolayer graphene can withstand 25% elastic strain.\(^{20}\) In addition, for traditional strain engineering, only during the process of in-plane epitaxial growth, it can obtain less than 1% of residual strain or prestrain. While there are a variety of deformation methods both in plane and out of plane to be designed for 2D materials due to the atomic-scale thickness, which means more various methods to induce strain to 2D materials. Moreover, 2D materials are more sensitive to strain and even small strains can change the lattice structure, which significantly modulate their physical properties. Strain-induced changes of various physical properties have been effectively achieved in 2D materials. In aspect of optical properties, strain can directly change the phonon structure and energy band structure of 2D materials, resulting in the shift of Raman and photoluminescence (PL) peaks.\(^{21,22}\) Especially, strain induces the transition from indirect to direct band gap in 2D materials, which greatly enhances luminous efficiency, making it more suitable for optoelectronic devices. As for electrical properties, proper strain amplitude can improve the performance of FETs by increasing the carrier mobility of 2D materials.\(^{23}\) The piezoresistive and piezoelectric effect appear in strained 2D materials, allowing their potential applications in the sensors, photodetectors, and nanogenerators.\(^{24}\) In addition, strain can not only modulate the magnetism of intrinsic van der Waals magnets but also induce magnetism in nonmagnetic 2D materials.\(^{25,26}\) Therefore, strain engineering of 2D materials greatly expands their application scope in the field of flexible devices, such as strain sensors, optoelectronic devices, and other wearable devices.\(^{27-29}\)

In this review, we present how to induce strain to 2D materials from the lattice mismatch—the use of different substrates, such as flexible substrates, patterned substrates, and piezoelectric substrates—to the action of atomic force microscope (AFM) tip and bubbles. Meanwhile, we simply introduce some techniques of monitoring the strain distribution in strained 2D materials, including scanning tunneling microscopy (STM), high-resolution transmission electron microscopy (HRTEM), Raman and PL spectroscopy, as well as second and third harmonic generation. Then, we discuss the modulation of various properties of 2D materials based on the strain engineering, such as phonon mode, energy band, carrier mobility, piezoresistive effect, piezoelectric effect, magnetic behaviors, and phase transition. Finally, we illustrate the potential applications of strained 2D materials, including flexible strain sensors and photodetectors, and further look forward to their opportunities and challenges in practical applications in the future.

## 2 | METHODS OF INDUCING STRAIN

### 2.1 | Lattice mismatch

In the traditional semiconductor industry, strain is induced in bulk single crystals due to the mismatch of lattice constants between single-crystal materials and substrates in the epitaxial growth. The method of lattice mismatch can also be applied to the strain inducing of 2D materials with atomic thickness. When there is a lattice mismatch between two components of 2D heterojunctions or between 2D materials and substrates, tensile or compressive strain is induced in 2D materials.

Recently, various lateral heterojunctions formed by two kinds of 2D materials with similar crystal structures but different lattice constants have been widely demonstrated, emerging large strain at the line interface. Li et al grew a monolayer WSe\(_2\)-MoS\(_2\) lateral heterojunction by the method of two-step epitaxial growth and revealed the existence of both tensile and compressive strain in the epitaxial MoS\(_2\) region through the PL measurements, due to a large mismatch of lattice constants between WSe\(_2\) and MoS\(_2\).\(^{30}\) Moreover, Shih et al imaged the moiré pattern in the MoS\(_2\) region of in-plane WSe\(_2\)-MoS\(_2\) heterojunctions by STM to further investigate the strain distributions induced by lattice mismatch.\(^{31}\) By analyzing the moiré pattern, 2D strain tensor of the central lattice point was considered \(\varepsilon_{aa} = 1.17\%, \varepsilon_{bb} = 0.69\%, \text{ and } \varepsilon_{ab} = \varepsilon_{ba} = 0.69\%\), respectively. From the scanning transmission electron microscopy (STEM) images, two types of dislocations were found at the interface of WSe\(_2\)-MoS\(_2\) lateral heterojunctions, causing the partial relief of strain and then the anisotropic strain.

Strain induced by lattice mismatch is inhomogeneous, which is the largest at the center of the mismatched interface, while it decreases in the region far away from the interface. Through high-resolution microscopic characterization methods, such as STEM and STM, the lattice distortion is clearly observed, so that the strain distribution can be investigated in detail. In addition, the moiré pattern formed by lattice mismatch not only helps quantitatively describe the strain tensor but also modifies the electronic structure of 2D materials, which has aroused wide interests.\(^{32}\)

### 2.2 | Flexible substrates

Due to the atomic-scale thickness, 2D materials tend to deform in the out-of-plane direction after being subjected to the external force. The deformation leads to the generation of strain in 2D materials. By selecting proper flexible substrates, strain can be induced and adjusted in 2D
materials by applying the external force to the substrates. There are many kinds of flexible substrates available, such as polydimethylsiloxane (PDMS), polycarbonate (PC), polystyrene (PS), polyethylene terephthalate (PET), polyethylene naphthalate (PEN), and polyimid (PI).

### 2.2.1 Mismatch of elastic modulus

Mismatch of elastic modulus between 2D materials and flexible substrates can induce strain in 2D materials by producing wrinkles. The most commonly used flexible substrate is PDMS substrate.\(^{22,33,34}\) As shown in Figure 1 (A), after transferring 2D materials to the prestretched PDMS substrate and then releasing the prestrain, periodic wrinkles of 2D materials form in the direction of initial strain axis due to the buckling-induced delamination caused by the differences in both elastic modulus and recovery ability between 2D materials and PDMS substrate.\(^{34}\) The prestrain \(\varepsilon_{\text{pre}}\) is defined as follows:\(^{35}\)

\[
\varepsilon_{\text{pre}} = \left( \frac{\Delta L}{L} \right) \times 100\%
\]

where \(L\) and \(\Delta L\) are the initial and stretched lengths of the PDMS substrate, respectively. Furthermore, if the prestrain is applied in both \(x\) and \(y\) directions of the substrate, subsequent release of the prestrain leads to the crumpling of adhered 2D materials and further inducing the formation of three-dimensional (3D) wrinkles.\(^{29}\) It is found that this 3D crumple structure of 2D materials improves the light absorption; therefore, the performance of optoelectronic devices based on this structure is also greatly enhanced. In addition to prestretching the flexible substrates to produce the prestrain, periodic wrinkles of 2D materials will also appear when transferring 2D materials to the topmost surface of the pre-bent PDMS substrate and then releasing the prestrain to yield uniaxial compression strain (Figure 1(B)).\(^{36}\) The induced maximum strain \(\varepsilon\) on the top of wrinkles can be calculated by the following formula:\(^{34,37}\)

**FIGURE 1**  (A) Schematic diagram of the process for forming ReSe\(_2\) wrinkles by prestretched PDMS substrate.\(^{34}\) Adapted with permission: Copyright 2015, American Chemical Society. (B) The process for forming InSe wrinkles by pre-bent PDMS substrate.\(^{36}\) Reproduced with permission: Copyright 2019, Royal Society of Chemistry. (C) Illustration of laser-induced local heating. (D) Schematic diagram of biaxial tensile strain in monolayer MoS\(_2\).\(^{44}\) Reproduced with permission: Copyright 2015, IOP Publishing Ltd. (E) Schematic of thermally induced PS substrates to form graphene/graphite wrinkles. (F) Illustration of the original graphene device (left), texturing graphene devices of uniaxial strain (middle) and biaxial strain (right).\(^{47}\) Reproduced with permission: Copyright 2015, American Chemical Society. (G) 3D diagram of two-point bending apparatus.\(^{50}\) Copyright 2009, American Physical Society. (H) Actual photograph of four-point bending apparatus. (I) The simplified cross-section of the bending flexible substrate.\(^{51}\) Reproduced with permission: Copyright 2013, American Chemical Society
where \( h \) and \( \sigma \) are the thickness and Poisson’s ratio of 2D materials, and \( \delta \) and \( \lambda \) are the height and width of periodic wrinkles, which can be extracted from the AFM or the scanning electron microscopy (SEM) image.

However, the generation of wrinkles in 2D materials is relatively random, and it is difficult to control and repeat. Generally, the wrinkles in monolayer or bilayer 2D materials may easily collapse due to their low stiffness, while multilayer 2D materials tend to form stable wrinkles. The strain of wrinkles induced by the pre-trained flexible substrates is inhomogeneous, which is the largest at the top of wrinkle, usually in the range of \( \sim 1\% - 2\% \).

\[ \epsilon = \pi^2 h \delta (1 - \sigma^2) \lambda^2 \]  

(2)

### 2.2.2 Mismatch of thermal expansion

Choosing the substrate with a thermal expansion coefficient being different from 2D materials can induce strain to 2D materials during the synthesis process of chemical vapor deposition (CVD) or epitaxial growth and the condition of local heating. Because the degrees of expansion and contraction between the substrate and 2D materials are different when the temperature changes, there exists the strong interaction among them; subsequently, the deformation of substrate can induce tensile or compressive strain in 2D materials. Recent studies indicate that 2D materials grown on a variety of rigid substrates can generate strain, such as silicon/silicon oxide (Si/SiO\(_2\)), sapphire, and mica. However, this strain cannot be effectively transferred to 2D materials due to low Young’s modulus of rigid substrates. Therefore, using flexible substrates with high Young’s modulus can effectively induce more strain to 2D materials.

As a result of much higher thermal expansion coefficient, some polymer substrates are prone to be heavily stretched when being heated, thus inducing tensile strain in the 2D materials attached to their surfaces. For example, a PDMS substrate has a relatively large positive thermal expansion coefficient, while the thermal expansion coefficient of MoS\(_2\) is negative. When being locally heated using a laser, the stretching of the PDMS substrate will introduce tensile strain to MoS\(_2\), as shown in Figure 1(C), (D). Then, a biaxial homogenous strain of 0.23\% has been achieved by heating up the MoS\(_2\)/PDMS sample at 150°C. Furthermore, it is important to maintain a balance between thermal expansion coefficient and Young’s modulus of flexible substrates. According to the finite element analysis done by Frisenda et al, substrates with both larger thermal expansion coefficient and higher Young’s modulus can effectively transform the thermal expansion of substrates into homogenous biaxial strain of 2D materials, and the calculated strain transfer efficiency is larger than 80% when heating or cooling the PC substrate with higher Young’s modulus of 2.5 GPa. After transferring monolayer MoS\(_2\) onto the PC substrate, a variable strain in MoS\(_2\) is continuously induced from a compressive strain of \(-1.48\%\) to a tensile strain of 0.48\% by gradually changing the temperature of the PC substrate from \(-200\) to \(100°C\).

Despite the larger thermal expansion, some polymer substrates tend to shrink when being heated because of their unique mechanical properties. A PS substrate is a typical thermally induced shrinkable substrate, which is often used to introduce compressive strain to 2D materials. Nam et al realized well-defined 3D texturing of graphene by heating PS substrates above the glass transition temperature and achieved uniaxial, biaxial (uniform), and localized strains by controlling the boundary of PS substrates (Figure 1(E), (F)). It was also found that the morphology of graphene crumples was related to the deformation degree of PS substrates, which was easily tuned by changing the processing parameters.

Based on the mismatched thermal expansion between the substrate and 2D materials, biaxial and homogenous strains can be induced in a controlled manner by heating or cooling the substrate. While one problem that cannot be ignored is that the induced strain will be released slowly with the decrease of temperature or long-time storage of samples.

### 2.2.3 Bending flexible substrates

Flexible substrates with high Young’s modulus (such as PET, PEN, and PI) can be used to directly apply strain on the upper surface of 2D materials by bending with external force. Bending is typically achieved by the methods of two-point bending or four-point bending through the designed clamping tools, and the typical bending devices are shown in Figure 1(G), (H). By controlling the flexible substrates to be bent up or down, tensile or compressive strain is induced in 2D materials, corresponding to positive or negative strain values, respectively. When 2D materials are located at the center of a bent flexible substrate, the strain \( \epsilon \) can be defined as follows:

\[ \epsilon = \tau / 2R \]

(3)

where \( \tau \) is the thickness of the flexible substrate, and \( R \) is the curvature radius of bent flexible substrate (Figure 1).
(I)). It is a direct way to induce a continuously varying strain to 2D materials by changing the degree of bending point by point.

The method of bending flexible substrates to induce uniaxial and homogenous strains is easy to operate, and it has the advantages of good reproducibility, reversible regulation in a critical range, and so on. Moreover, Raman spectroscopy, PL spectroscopy, and other characterization methods have been simultaneously used to carry out the real-time measurements in the process of inducing mechanical strain, which can better explain the effects of strain on the physical properties of 2D materials.

Significantly, when strain is induced in 2D materials by bending flexible substrates, it is better to avoid using substrates with low Young's modulus such as PDMS because these substrates are more likely to have the low strain transfer efficiency. Moreover, due to the weak van der Waals interaction, there are severe slippage between 2D materials and the substrates during the bending processes, resulting in low strain transfer efficiency. To overcome the limitation, a certain thickness of metal is usually prepared on the two edges of 2D materials, which can prevent 2D materials from slipping under the action of mechanical force and simultaneously serve as the electrodes of the related FETs. In addition, Duan et al reported a new method of polymer encapsulation to achieve effective strain engineering of 2D materials. First, monolayer MoS2 was transferred onto Si/SiO2 substrate, and then polyvinyl alcohol (PVA) was spin-coated to completely encapsulate MoS2. After peeling off PVA from the Si/SiO2 substrate, monolayer MoS2 was closely attached to the flexible PVA substrate. The strong interaction between the spin-coated PVA and monolayer MoS2 ensures the effective strain transfer, while the slippage and decoupling can be neglected. Therefore, it is important to explore new approaches to induce strain by bending flexible substrates, which should satisfy three conditions: high Young's modulus of substrates, the strong interaction at the interface, and minimum slippage.

### 2.3 Patterned substrates

Transferring 2D materials to the patterned substrates is also a common way to induce strain into 2D materials due to the fluctuation of patterned surface. The surface of rigid substrates has been successfully modified through the techniques of ion-beam sputtering, photolithography, electron-beam etching, and nano self-assembly. The obtained substrates form a unique surface with a periodic ripple, nanocone, or nanopillar structure. By using patterned substrates, strain has been successfully introduced into many 2D materials, including graphene, MoS2, etc. For example, Molle et al used Au as a sacrificial layer to pattern SiO2/Si substrates by ion-beam sputtering and then transferred MoS2 nanosheets to the rippled SiO2/Si substrates to induce local strain in the obtained MoS2 ripples, introducing a one-directional anisotropy. The schematic diagram of the process for patterning the substrates and forming the MoS2 ripples is shown in Figure 2(A). Cavallo et al designed periodic Cr hard mask arrays on bulk Si and then obtained Si pillar structure by reactive ion etching. After removing the Cr mask and the process of dry thermal oxidation, the substrates with an inverted funnel Si array were formed (Figure 2(B)). MoS2 nanosheets were later transferred to the patterned Si substrate by the dry transfer method, and the off-axis SEM image showed that the conformal contact between MoS2 and the patterned Si substrate was found, as shown in Figure 2(C), (D). The induced tensile strain in MoS2 nanosheets ranged from ~3.46 to ~3.65% and presented the distribution of a single inverted funnel.

In addition, one-dimensional (1D) semiconductor nanostructure arrays are applied to understand the nanopatterning of substrates, and these substrates induce periodic strain to 2D materials. Simultaneously, nanopatterned substrates and 2D materials construct the mix-dimensional hybrid heterojunctions. Zhang et al used homoepitaxial ZnO nanorod arrays (NRAs) to pattern the single-crystal ZnO substrate by hydrothermal synthesis and then transferred monolayer MoS2 to the patterned ZnO substrate by the wet transfer method, yielding the organized MoS2 wrinkles. Figure 2(E) shows the specific process of inducing strain into monolayer MoS2 by the NRA-patterned ZnO substrate. MoS2 wrinkles on the patterned ZnO substrate suggest the formation of periodic deformation, further inducing periodically localized biaxial strain. The strain in monolayer MoS2 induced by the patterned ZnO substrate is inhomogeneous, and the strain distribution can be divided into three parts, including the unstrained region, the strained region, and the most strained region, which is shown in Figure 2(F).

It is worth noting that not only rigid substrates can be modified, patterned flexible substrates can also be obtained through a series of different methods. For example, Stafford et al produced nanopatterned PS films, where PS solution was first spin-casted on the nanostructured Si template and the obtained PS film was then transferred to a prestrained PDMS film, after releasing the prestrain of PDMS film, patterned PS films with periodic wrinkles were formed. The wavelength and morphology of wrinkles were related to the geometric parameters of the nanostructured Si template and the prestrain direction of PDMS film. Compared with modified rigid substrates, the wrinkle-patterned flexible substrates have better scalability, which can be used in
various flexible electronic devices. Chen et al fabricated suspended wavy-structured electrode arrays by transferring the graphene oxide (GO) microribbons to the pretrained tripod-structured PDMS substrate and then releasing the prestrain, which were used in stretchable supercapacitors (Figure 2(G)). These unique electrode arrays reduced the strain concentration and kept constant distance of the electrode fingers in the stretching/releasing process. It was proved that the stretchable supercapacitors showed stable electrochemical performance even under 100% strain and 5000 cycles.

Compared with the method of using flexible substrates to induce strain, using patterned substrates has a greater advantage that it keeps 2D materials in a constant local strained state, which is convenient for subsequent research. Moreover, the state of strain is closely related to the morphology of patterned substrates; thus, the strain and strain-induced properties can be designed by changing the nanostructure patterns of substrates.

### 2.4 Piezoelectric substrates

Piezoelectric ceramic has the ability of generating mechanical deformation when applying an excited electric field; that is, it has the inverse piezoelectric effect, which can be used as a substrate to induce strain into 2D materials. When the electric field is applied in the direction of electric polarization of piezoelectric substrates, the substrates deform in the plane perpendicular to the electric field and thus induce isotropic strain to the connected 2D materials. The positive or negative voltage determines whether the piezoelectric substrates are stretched or contracted. Therefore, whether the induced strain in 2D materials is tensile or compressive depends on the direction of electric field.

Louca et al introduced the variable tensile strain to a ribbon-like MoTe2 single crystal by applying an external electric field to a piezoelectric stack, resulting in an anisotropic magnetoresistance being dependent on the relative direction of tensile strain and crystal orientation. In order to improve the strain transfer efficiency, the MoTe2 single crystal was glued on the surface of piezoelectric stack, instead of relying on the van der Waals interaction. However, the provided strain was still relatively small, with the maximum value being only 0.05%.

Lau et al induced a uniform biaxial compressive strain to trilayer MoS2 by applying a bias voltage to a piezoelectric substrate ([Pb(Mg1/3Nb2/3)O3]0.7-[PbTiO3]0.3, PMN-PT) (Figure 2(H)). Due to ultrathin thickness and superelasticity of the graphene top electrode, the polarization-
induced strain from the PMN-PT substrate was better transferred to trilayer MoS₂, and the induced strain was up to 0.2% under the voltage of 500 V.

Compared with uniaxial strain induced by bending or prestraining flexible substrates, biaxial strain induced by using piezoelectric substrates can be modulated more precisely by controlling the applied voltage. Although this method of inducing strain is relatively simple and straightforward, there are potential safety problems due to the excessive applied voltage, which does not meet the energy-saving requirements as well. Moreover, it still remains unknown that if the interfacial interaction between piezoelectric substrate and 2D materials is strong enough to completely transfer the polarization-induced strain from the deformed substrate to the target 2D materials. How to obtain larger strain amplitude and higher strain transfer efficiency still remains a problem when inducing strain by piezoelectric substrates.

2.5 | AFM tip

The AFM is an important tool to characterize the physical properties of 2D materials. The high-resolution AFM can not only accurately obtain the surface morphology information of 2D materials but also apply the strain to 2D materials through direct contact between the tip and the sample, thus inducing strain into 2D materials due to the local deformation caused by the tip.⁶⁸,⁶⁹

Wang et al induced isotropic compressive and tensile strains to monolayer MoS₂ by applying forces at different positions of MoS₂ flake through AFM tip.⁷⁰ As shown in Figure 3(A), (B), the monolayer MoS₂ flake was concave when the tip was placed in the central region, corresponding to compressive strain, whereas it was convex when the tip was located near the edges, corresponding to tensile strain. The local strain ε can be expressed as follows:

$$\varepsilon = F / (A \cdot E)$$  \hspace{1cm} (3)

where F and A are the cross-sectional area and the applied force of AFM tip, and E is Young's modulus of monolayer MoS₂. However, due to the physical limitation of Si substrate, the maximum of applied force can only be up to 25 mN, which means the AFM tip causes only small deformations of monolayer MoS₂.

In order to avoid the limitation of rigid substrates, Kis et al placed the AFM tip in the middle of the suspended MoS₂ membrane and applied force to deform it while applying the source–drain bias voltage, thus inducing mechanical strain and achieving simultaneously the electrical measurement, as shown in Figure 3(C).⁷¹ The deflection depth of the MoS₂ membrane in the center of the tip reached 33 nm, which reflected on relatively large degree of strain. Bayer et al utilized the AFM tip to apply strain to the suspended few-layer graphene (FLG) and simultaneously performed in situ hyperspectral Raman mapping to observe the strain distribution by analyzing the frequency shifts of G and 2D Raman peaks under strain.⁷² When applying a force of ~1300 nN on the FLG through the tip, there was a small and roughly circular region with small downshifts of G and 2D peaks under the position of AFM tip. As the applied force is increased, both the diameter of the circular region and the frequency shifts of G and 2D peaks became larger. The strain caused by the deformation of the suspended graphene was inhomogeneous, where the areas close to the AFM tip were more deformed, meaning a greater strain.

In addition, tip-enhanced Raman spectroscopy (TERS), combined with AFM and Raman measurements, have been widely used to monitor the strain distribution in strained 2D materials. In TERS, the tip scans the surface of 2D materials with subnanometer accuracy; when the tip metal is excited by the laser to produce localized surface plasmon resonance, a strong electromagnetic field will be generated between the tip and the surface of sample, thus greatly improving the intensity of Raman signals and the image resolution.⁷³ Recently, the detection of spatially localized strain can reach nanoscale resolution in the system of strained monolayer MoS₂ (~2.3 nm),⁷⁴ trilayer MoS₂ (~25 nm),⁷⁵ and monolayer WS₂ (~15 nm).⁷⁶

2.6 | Bubble

Due to the van der Waals force between 2D materials and the substrate, water and hydrocarbon adsorbed in 2D materials will aggregate together during the assemble process, resulting in bubbles at the interface.⁷⁶ The bubbles cause the surface fluctuation of 2D materials, which induces strain to 2D materials at the interface. Grigorieva et al transferred monolayer MoS₂ to various substrates by wet transfer technique and obtained bubbles at the corresponding interface, thus inducing a smooth and gradient strain of about 2% into monolayer MoS₂.⁷⁷ The optical microscopy (OM) image and AFM topography of the bubbles at the MoS₂/h-BN interface are shown in Figure 3(D), (E). The bubble shape is described by two parameters, R and h (Figure 3(F)), which represent the radius and the maximum height of bubble, respectively. The aspect ratio h/R is defined using the following formula:
where $\gamma$ is the adhesion energy of 2D materials to substrate, $C_1$ is a numerical coefficient, and $Y$ is Young's modulus of 2D materials. Therefore, the aspect ratio $h/R$ of bubble is only relevant to 2D materials and substrate, while it is independent of the bubble itself. The local strain $\varepsilon$ of monolayer MoS$_2$ enclosing a bubble is proportional to the aspect ratio $h/R$, which can be described as follows:

$$\varepsilon \sim (h/R)^2$$  \hspace{1cm} (5)$$

It is obvious that the bubbles naturally presented at the van der Waals interface induce universal and uniform strain.

In addition to the stable bubbles discussed above, the bubbles can also be generated by adjusting the internal and external pressure difference of 2D materials, such as blowing gas into the microcavity. Bunch et al transferred CVD-grown and mechanically exfoliated MoS$_2$ monolayer membranes onto the substrates with periodic cylindrical microcavities and blew N$_2$ to the cavities to produce bubbles across MoS$_2$ membranes and cavities, as shown in Figure 3(G), (H). The suspended MoS$_2$ membranes bulged up or down, which depended on the pressure difference between internal and external environment, corresponding to tensile or compressive biaxial strain. The strain $\varepsilon$ at the center of a suspended MoS$_2$ membrane is defined as follows:

$$\varepsilon = c(\nu) \times (\delta/a)^2$$  \hspace{1cm} (6)$$

where $c(\nu)$ is a constant determined by the Poisson's ratio of monolayer MoS$_2$, and $\delta$ and $a$ represent the bending degrees of suspended MoS$_2$ membranes (Figure 3(I)), which can be measured by AFM. The pressure is varied with the amount of N$_2$, so that $\delta$ can be continuously changed, further achieving continuous and reversible adjustment of strain.

Furthermore, the sudden changes of internal and external pressures can also be achieved by placing 2D materials in different pressure environments. Li et al transferred monolayer graphene onto Si/SiO$_2$ substrates with circular holes and placed the sample in an autoclave for several days to reach equilibrium under high pressure. When taking the sample back to the ambient conditions, graphene bulged up due to the sudden pressure

FIGURE 3  Schematic diagram of (A) compressive and (B) tensile strain generation when applying forces at the middle or edge of monolayer MoS$_2$.\(^\text{70}\) Adapted with permission: Copyright 2015, Springer Nature. (C) Diagram of the setup for the electrical measurement of suspended MoS$_2$ membrane under compressive strain caused by AFM tip.\(^\text{71}\) Reproduced with permission: Copyright 2015, American Chemical Society. (D) OM image of bubbles at the interface of MoS$_2$ and h-BN (The scale bar was 10 $\mu$m). (E) AFM image (The scale bar was 2 $\mu$m) and (F) cross-sectional height profile of a typical bubble formed at the interface.\(^\text{77}\) Reproduced with permission: Copyright 2019, American Chemical Society. (G) Device schematic of a suspended MoS$_2$ membrane on a cylindrical cavity. (H) CVD-grown MoS$_2$ monolayers transferred on the substrate with cylindrical microcavities (The scale bar was 20 $\mu$m). (I) AFM cross-sections of the device under different external pressures.\(^\text{78}\) Reproduced with permission: Copyright 2016, American Chemical Society
changes across the circular holes. The limitation of this method is that the induced strain in sample keeps a constant value, and more samples with different strains need to be prepared by adjusting the pressure difference.

### 2.7 Comparison of different methods of inducing strain

In the above content, we systematically described and classified the experimental methods of inducing strain to 2D materials and discussed the advantages and disadvantages of each method. In order to compare different methods of inducing strain more intuitively, we have listed the ranges and types of strain that can be achieved in various strained 2D material systems in Table 1. This summary may allow us to find the most appropriate methods for different research demands.

### 3 MODULATION OF PROPERTIES BASED ON STRAIN ENGINEERING

#### 3.1 Phonon mode

The phonon structure of 2D materials will be changed due to the distortion of lattice structure caused by uniaxial or biaxial strain. Generally, the tensile strain leads to the softening of phonon mode, while compressive strain is the opposite. Raman spectroscopy is an effective method to investigate the change of phonon mode, which

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**Table 1** The ranges and types of strain in various strained 2D material systems by different inducing methods

| Straining methods          | Substrates                     | Materials/layers | Ranges of strain                  | Types of strain          | References |
|----------------------------|--------------------------------|-----------------|-----------------------------------|--------------------------|------------|
| Lattice mismatch           | Si/SiO$_2$                    | WSe$_2$-MoS$_2$/1 | $-1.1 \pm 0.18\%$ to $1.59 \pm 0.25\%$ | Local inhomogeneous      | [30]       |
| SrTiO$_3$                  | SnSe$_2$/1–3                  | Equivalent pressure: 11–23 GPa | Local inhomogeneous               | [80]       |
| Prestrained flexible substrates | PDMS                          | WS$_2$/2–4       | 0–2%                              | Local inhomogeneous      | [33]       |
| Gel-PAK film               | ReSe$_2$/mutilayers           | $\sim 1.64\%$   | Local inhomogeneous               | [34]       |
| PDSM                       | Graphene/mutilayers           | Below 0.3%       | Local inhomogeneous               | [35]       |
| Bending flexible substrates | PET                            | MoS$_2$/1–2      | 0–0.8%                            | Uniaxial homogeneous     | [21]       |
|                            | PVA                            | MoS$_2$/1        | 0–1.49%                           | Uniaxial homogeneous     | [52]       |
|                            | PP                             | InSe/4–8         | 0–1.15%                           | Uniaxial homogeneous     | [81]       |
|                            | PI                             | MoS$_2$/1–2      | 0–0.32%                           | Uniaxial homogeneous     | [82]       |
|                            | PEN                            | BP/mutilayers    | $-0.22$ to $0.15\%$               | Uniaxial homogeneous     | [83]       |
| Heating or cooling substrates | Silica, etc.                | WSe$_2$/1        | $-0.2$ to $1\%$                   | Biaxial homogeneous      | [84]       |
|                            | PC                             | MoS$_2$/1        | $-1.58$ to $0.48\%$               | Biaxial homogeneous      | [45]       |
|                            | PS                             | Graphene/mutilayers | $\sim 0.27\%$                  | Uniaxial/biaxial homogeneous | [47]       |
| Patterned substrates       | Rippled Si/SiO$_2$            | MoS$_2$/4        | $\sim 0.5\%$                      | Local inhomogeneous (anisotropic) | [53]       |
|                            | Patterned PDMS                | Graphene/mutilayers | 1–2%                         | Local inhomogeneous      | [54]       |
|                            | Line patterned Si             | Graphene/mutilayers | $\sim 0.5\%$                  | Biaxial inhomogeneous (anisotropic) | [56]       |
| Piezoelectric substrates   | PMN-PT                         | MoS$_2$/3        | $-0.04\%/100$ V                   | Biaxial homogeneous      | [67]       |
|                            | PMN-PT                         | $\alpha$-In$_2$Se$_3$ | $-0.011\%/100$ V             | Biaxial homogeneous      | [85]       |
| AFM tip                    | Si/SiO$_2$                    | MoS$_2$/1        | $4.7 \times 10^{-3}$F             | Local inhomogeneous (isotropic) | [70]       |
|                            | Si/SiO$_2$                    | Suspended MoS$_2$/1–3 | $\delta_{mem}$: $\sim 33$ nm  | Local inhomogeneous (isotropic) | [71]       |
| Bubbles                    | h-BN, etc.                    | MoS$_2$/1        | $\sim 2\%$                        | Gradient homogeneous     | [77]       |
|                            | Si/SiO$_2$ with cavities      | MoS$_2$/mutilayers | $-0.8$ to $0.15\%$             | Biaxial homogeneous      | [78]       |
can be shown in the splitting and shifting of Raman peaks.

Ferrari et al investigated the optical phonon changes of monolayer graphene under uniaxial strain by Raman spectroscopy and found that uniaxial tensile strain resulted in the red shifts of G and 2D peaks of graphene.\textsuperscript{50} Furthermore, the G peak split into G\textsuperscript{+} and G\textsuperscript{−} sub-bands due to the split of doubly degenerate E\textsubscript{2g} phonons when the applied strain was over 0.6%, which represented the vibration modes perpendicular and parallel to the strain direction, respectively (Figure 4(A)). In other studies, the 2D peak of monolayer graphene also splits into two peaks, 2D\textsuperscript{+} and 2D\textsuperscript{−}, both of them undergo the red shift with increasing strain, and the red shift value depends on the strain direction.\textsuperscript{86} The first-principles calculations show that the reason for the splitting of 2D double-resonance Raman scattering peak is the combine effect of distorted Dirac cones and TO phonon dispersion anisotropy caused by uniaxial strain.

In addition, for anisotropic 2D materials, uniaxial strains along different crystal orientations have different effects on their Raman peaks. Taking black phosphorus (BP) for example, Lau et al discovered that uniaxial tensile strain along its zigzag (ZZ) direction caused the red shifts of A\textsubscript{2g} and B\textsubscript{2g} Raman peaks, but it had no effect on the A\textsubscript{1g} peak, yet the A\textsuperscript{1}g and B\textsubscript{2g} Raman peaks got red-shifted, while the A\textsubscript{2g} peak barely shifted when the strain was applied along the armchair (AC) direction.\textsuperscript{88} According to the density functional perturbation theory calculations, the anisotropic Raman responses originated from the changes of both bond lengths and bond angles in the strained BP flakes. This proves that the uniaxial strain can selectively adjust the out-of-plane and in-plane vibration modes of BP, which provides guidance for the application of anisotropic flexible electronic devices.\textsuperscript{89}

Based on the strain-induced phonon mode of 2D materials, Raman spectroscopy has been widely used in monitoring the magnitude and distribution of strain by measuring the strain-sensitive and Raman-active modes of strained 2D materials. The frequency of Raman peaks changes linearly with the strain, which means the

**FIGURE 4** (A) The red shift and splitting of G Raman peak of monolayer graphene under uniaxial strain.\textsuperscript{50} Reproduced with permission: Copyright 2009, American Physical Society. (B) The splitting of E\textsuperscript{′} Raman peak of monolayer MoS\textsubscript{2} under uniaxial strain.\textsuperscript{51} Reproduced with permission: Copyright 2013, American Chemical Society. (C) OM image of a wrinkled MoS\textsubscript{2} flake. (D) Raman mapping of the E\textsubscript{1g} mode of the MoS\textsubscript{2} flake in (C).\textsuperscript{87} Reproduced with permission: Copyright 2019, Royal Society of Chemistry. (E) PL spectroscopy of bilayer WSe\textsubscript{2} under different strains. Inset was the OM image of bilayer WSe\textsubscript{2} flake.\textsuperscript{90} Reproduced with permission: Copyright 2014, American Chemical Society. (F) Schematic diagram of the “funnel effect” in a MoS\textsubscript{2} wrinkle.\textsuperscript{22} Reproduced with permission: Copyright 2013, American Chemical Society.
magnitude of strain in 2D materials can be quantified through the shifts of Raman peaks of a certain vibration mode. Moreover, Raman mapping offers a straightforward way to monitor the inhomogeneous strain distribution.

3.2 | Energy band

Strain can significantly change the electronic structure of 2D materials and thus increases or decreases the band gap. Javey et al demonstrated that there was a drastic enhancement of PL intensity and a slight red shift of PL peak position for the bilayer WSe$_2$ under uniaxial homogeneous tensile strain (Figure 4(E)). Density functional theory (DFT) calculations showed that the conduction band minima changed from K to Σ points in the Brillouin zone, indicating a transition from an indirect band gap to a direct band gap when applying uniaxial strain, which was consistent with the experimental results. The strain-induced transition between indirect gap and direct gap is also observed in other 2D materials both in experiment and calculation, including MoS$_2$, WS$_2$, antimonene, BP, and so on.

In addition, for nonhomogeneous strain caused by wrinkles or bubbles, compared with the unstrained region, the PL spectroscopy in the strained region shows increased intensity and red-shifted peak position, which originates from the “funnel effect”. As shown in Figure 4(F), when the laser irradiates on a wrinkled MoS$_2$ flake, photogenerated excitons drift to the wrinkle region with the maximum strain which has the narrowest band gap and then recombine here, resulting in the red shift of PL peak position.

Beyond that, the modulation of band gap by strain is related to the layer number of 2D materials. Bolotin et al reported the band gap structure changes of monolayer and bilayer MoS$_2$ under uniaxial tensile strain and found that when applying 1% tensile strain, the band gap of monolayer MoS$_2$ decreased by ~45 meV, whereas that of bilayer MoS$_2$ decreased by ~120 meV. Yan et al studied the evolution of energy band structure of few-layer BP (2–10 layers) with in-plane biaxial strain by infrared (IR) spectroscopy. The results showed that the shift of IR peak position was linear with the applied strain, and the rate of shift was obviously exponential and dependent on the number of layers; that is, less layers corresponded to the smaller rate of IR peak shifts, which was completely consistent with the calculation results. This dependence was closely related to the interlayer van der Waals interaction caused by the strain, indicating that the in-plane tensile strain weakened the interlayer interaction due to the in-plane puckered crystal structure of BP.

3.3 | Nonlinear optical response

Recently, 2D materials have been proved to exhibit various interesting nonlinear optical properties, including second-harmonic generation (SHG) and third-harmonic generation (THG). SHG is a nonlinear optical process described by the second-order susceptibility tensor, usually observed in materials with broken inversion symmetry, such as monolayer or odd-layer transition metal dichalcogenides (TMDs), and CrOCl. SHG is sensitive to the lattice symmetry of materials, and the intensity can be greatly influenced by strain because strain will distort the lattice, thus changing the optical susceptibility of 2D materials. Meanwhile, the direction of applied strain can also be indicated by the polarization-resolved SHG.

Lu et al transferred monolayer MoS$_2$ to the Si/SiO$_2$ substrate spin-coated with TiO$_2$ nanowire (NW) suspension, thus inducing strain in MoS$_2$ through the formation of wrinkles. By analyzing the polarization-resolved SHG signal, it was found that there existed a strong anisotropic enhancement of SHG in the hybrid structure between monolayer MoS$_2$ and 1D TiO$_2$ NW, and the anisotropy mainly depended on the relatively direction between MoS$_2$ and TiO$_2$ NW. When TiO$_2$ NW was along the AC direction of MoS$_2$, the strain-induced deformation of AC axis was larger than that of ZZ axis, which led to the enhancement of SHG perpendicular to NW being less than that parallel to NW, resulting in a large anisotropic SHG signal finally.

Liu et al investigated the evolution of SHG intensity of monolayer MoSe$_2$ under different uniaxial tensile strains generated by bending the mica substrate. Because monolayer MoSe$_2$ belongs to the D$_{3h}$ point group, it has strong second-order nonlinear optical response. The intensity of SHG signal decreases linearly with the tensile strain, implying that the strain amplitude can be directly monitored by the SHG intensity. Moreover, the relative change of SHG intensity with strain is up to 0.49 ± 0.05/e, which is nearly one order of magnitude larger than that of PL peak intensity, showing that SHG is a more effective and sensitive technique to monitor the strain in 2D materials. In addition, the strain direction can be displayed through the evolution of polarization-resolved SHG pattern. Without strain, the intensity of SHG shows six same petals when the polarization of SHG is parallel or perpendicular to the incident polarization, thus the total SHG intensity has a circular shape. When the strain is applied, the SHG intensity decreases in each petal, which leads to the asymmetric shape of six petals, resulting in a strong angular dependence of the total SHG intensity, and thus the minor axis of polarization-resolved SHG intensity can indicate the strain direction.
In addition to simply describing the strain amplitude, polarization-resolved SHG can also be used to accurately map the full strain tensor in strained 2D materials.\textsuperscript{100} This method is based on the strain-modified nonlinear susceptibility tensor caused by the photoelastic effect. The strain-modified second-order nonlinear susceptibility tensor can be described using the following formula:

$$
\chi^{(2)}_{ijk} = \chi^{(2,0)}_{ijk} + P_{ijklm}u_{lm}
$$

where \(\chi^{(2)}_{ijk}\) and \(\chi^{(2,0)}_{ijk}\) are the second-order nonlinear susceptibility tensor of unstrained and strained materials, and \(P_{ijklm}\) is the photoelastic tensor. Obviously, the nonlinear susceptibility corresponds with the strain tensor one by one through a photoelastic tensor. Furchi et al successfully mapped the strain distribution in inhomogeneously strained monolayer MoS\textsubscript{2} by the method of polarization-resolved SHG and obtained the spatial resolution of 280 nm. Compared with the linear optical strain monitoring methods, such as Raman and PL mapping, the nonlinear SHG measurement to map the strain has a higher spatial resolution.

However, SHG process usually only exists in the materials with noncentrosymmetric structure, the related strain mapping technique cannot be well utilized in centrosymmetric 2D materials, such as even-layered TMDs. Different from the second-order susceptibility, the third-order nonlinear optical susceptibility is a common parameter in any 2D materials. Therefore, a more universal technique by applying polarization-dependent THG is designed to image the full strain tensor of strained 2D materials.\textsuperscript{103} After determining the photoelastic tensor of noncentrosymmetric monolayer WS\textsubscript{2} and centrosymmetric bilayer WS\textsubscript{2}, the full strain tensor can be extracted by fitting the THG pattern, and thus the strain distribution in homogeneously strained and randomly strained monolayer WS\textsubscript{2} is successfully mapped.

### 3.4 Electrical properties

#### 3.4.1 Carrier mobility

When 2D materials are used in FETs, two conditions need to be satisfied, that is, proper band gap and high carrier mobility. Theoretical studies have demonstrated that strain in 2D materials can not only influence the band gap but also change the carrier mobility.\textsuperscript{102,103} In some cases, strain can significantly increase the carrier mobility of 2D materials. Berry et al obtained a few-layer MoS\textsubscript{2} flake with wrinkles by micromechanical exfoliation and fabricated the MoS\textsubscript{2} FETs in the wrinkled and flat regions, respectively.\textsuperscript{87} The transfer curves of corresponding devices at different temperatures are shown in Figure 5(A), (B). It is observed that the carrier mobility of the wrinkled device (\(\mu_w = 5.55 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}\)) is about three times that of the flat device (\(\mu_f = 1.42 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}\)) at 30 K because the tensile strain in MoS\textsubscript{2} wrinkles suppresses the electron–phonon coupling and reduces the lattice scattering and thus increases the carrier mobility. Garaj et al fabricated MoS\textsubscript{2} FETs on the Si substrate with a crest-SiN\textsubscript{x} (c-SiN\textsubscript{x}) dielectric layer (Figure 5(C)) and found that the mobility of c-SiN\textsubscript{x} MoS\textsubscript{2} FETs was much higher than that of ordinary MoS\textsubscript{2} FETs with a SiO\textsubscript{2} dielectric layer (Figure 5(D)), reflecting the roughly positive correlation between the carrier mobility and the substrate roughness.\textsuperscript{104} Theoretical calculations indicated that the strain fluctuations of the rough c-SiN\textsubscript{x} dielectric layer reduced the band gap and carrier effective mass of MoS\textsubscript{2}, thus significantly improving the electrical properties and the carrier mobility.

However, strain also sometimes has negative impact on the carrier mobility of 2D materials. For example, few-layer SnSe has a large anisotropic ratio of carrier mobility, and monolayer SnSe is a nearly direct band gap semiconductor with high electron mobility of \(10^3 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}\) at zero strain.\textsuperscript{105,106} Under small tensile strain, the mobility of monolayer SnSe reaches up to \(10^4 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}\), whereas it will reduce to \(300 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}\) once the strain is over 8%. This phenomenon is attributed to the combine effect of strain on the effective mass and the elastic modulus. Small strain effectively decreases the effective mass and increases the elastic modulus, leading to an obvious enhancement of carrier mobility, but the opposite often occurs for the larger strain.

In addition, strain can achieve the modulation of anisotropic free-carrier mobility of 2D materials. Yang et al found that the preferred conducting direction of monolayer BP was rotated by 90° under 3–6% uniaxial or biaxial strain according to the first-principles simulations.\textsuperscript{107} For intrinsic monolayer BP, the electron mobility along the AC direction is as high as \(10^7 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}\), while that along the ZZ direction is very low. When applying uniaxial strain (5–6%) or biaxial strain (3–4%), the electron mobility along the corresponding direction decreases (AC) or increases (ZZ) dramatically. The change in electrical conductance anisotropy is due to the energy order change of two lowest-energy conduction bands under strain.

#### 3.4.2 Piezoresistive effect

When uniaxial or biaxial strain is introduced to 2D materials, their resistance will be changed, which we call the piezoresistive effect. Current research has shown that the
piezoresistive effect of 2D materials mainly come from the band gap change caused by strain. Zhang et al constructed BP FETs on flexible PEN substrates and observed a large room-temperature piezoresistive effect by the four-probe method. Figure 5(E), (F) shows the OM image of a flexible BP FET and the schematic diagram of how to achieve its compressive and tensile strain, respectively. As shown in Figure 5(G), when tensile (0.15%) or compressive (−0.18%) strain is induced in BP, its resistance is significantly changed especially at the charge neutral point, reflecting on a larger piezoresistive effect. Through the analysis of IR spectroscopy and thermal activation behavior, it was found that the piezoresistive effect started from the strain-induced change of band gap and carrier concentration. Tensile strain increased the band gap and reduced the carrier transition, leading to the increased resistance, while the band gap was reduced by compressive strain, which resulted in more carrier transition and the decreased resistance.

Furthermore, resistance can also be tuned by changing the carrier scattering in locally strained 2D materials. When strain is applied to 2D materials, the internal carrier scattering may increase or decrease, resulting in opposite resistance changes. Generally, for most CVD-grown or mechanical exfoliated intrinsic unstrained 2D materials, the band gap plays a more dominant role in resistance, while the carrier scattering is often neglected under strain. However, in some cases, the influence of carrier scattering on resistance cannot be ignored, especially for 2D materials with inherent highly localized strain. Zhang et al prepared periodically wrinkled graphene by the method of prestraining the PDMS substrate, which suffered from larger strain than flat graphene. The resistance decreased when applying strain to the wrinkled graphene because the strain flattening process reduced the carrier scattering.

Specifically, the charge tunneling model is usually used to explain the piezoresistive effect in polycrystalline 2D material nanosheets. The 2D material film is made...
up of densely packed nanosheets islands, and the charges are transferred from one island to another island through the tunneling effect. The tunneling distance of different islands increases when applying uniaxial tensile strain, thus reducing the tunneling of charges and causing the increase of resistance.

### 3.4.3 Piezoelectric effect

When some asymmetric crystal materials deform under the action of external force in a certain direction, the positive and negative polarization charges will be generated on two opposite surfaces, which is called the piezoelectric effect. To a certain extent, due to the reduction of dimensionality, 2D materials display the 3D symmetry breaking compared to bulk crystal structures, so some nonpiezoelectric bulk materials show intrinsic piezoelectric effect in the case of monolayer forms.

Wang et al first discovered the piezoelectric effect of mechanically exfoliated monolayer MoS2 under uniaxial strain by bending flexible PET substrates in experiments.\(^{109}\) It was found that odd-layer (1, 3, 5) MoS2 without centrosymmetry showed stronger piezoelectric response as the number of layers decreased, while even-layer (2, 4, 6) MoS2 with centrosymmetry had no piezoelectric response. As shown in Figure 5(H), strain-induced polarization charges generated along the ZZ edge of the monolayer MoS2 device when stretching the substrate, which drove electrons flow over the external circuit. However, when the strain was released, electrons flowed in the opposite direction. The polarization charges tune the Schottky barrier of the interface between metal electrode and MoS2 and thus show asymmetric modulation of electrical transport properties by strain. Furthermore, the piezoelectric effect of CVD-grown triangular monolayer MoS2 is observed under homogenous strain induced by the AFM tip.\(^{70}\) Compared with the irregular-shaped exfoliated monolayer MoS2, the polarization charges generated at three ZZ edges of triangular MoS2, which showed better modulation ability on the Schottky barrier and the carrier transport. The polarization charges and piezoelectric potential can change the carrier distribution in piezoelectric 2D materials and modify the Schottky barrier height between different materials, which can greatly improve the performance of electronic and optoelectronic devices. The related device application will be discussed in the next section.

### 3.5 Magnetic properties

The emerging of 2D magnetic materials has provided a new platform for the spintronics. However, most of the 2D materials are nonmagnetic, so it is of great value to exploit new method to induce or modulate magnetism to achieve their applications in spintronics. Recently, in many theoretical studies, local strain is proved to be a promising way to induce and modulate the magnetic properties of 2D materials.

On one hand, strain can induce magnetism in nonmagnetic 2D materials. The strain-induced magnetism of nonmagnetic 2D materials can be divided into two categories. One is to directly induce magnetism in 2D materials by applying strain, including TaS2, TaSe2,\(^{110}\) NbS2, NbSe2,\(^{111}\) SnSe2,\(^{112}\) and M2C (M = Hf, Nb, Sc, Ta, V) monolayers,\(^{113}\) which are nonmagnetic and become ferromagnetic under biaxial tensile strain. Gao et al investigated strain-induced magnetic properties of layered NbS2 and NbSe2 and found that these two materials exhibited the ferromagnetic characteristics under tensile strain.\(^{111}\) As shown in Figure 6(A), (B), spin-polarized states appeared when applying tensile strain due to the different effects of strain on the magnetic moment of S (Se) atoms and Nb atoms. It was confirmed that the ferromagnetic characteristics resulted from the competitive behaviors of through-bond and through-space interactions under strain. Geng et al reported that SnSe2 AC nanoribbons were magnetized under 10% compressive strain, and the appeared magnetic moment increased linearly with increasing strain.\(^{112}\) In addition, for some nonmagnetic 2D materials, such as graphene and MoS2, it is difficult to directly induce magnetism only by applying strain. The combination of strain engineering and doping, or inducing defects or vacancies, has proved to be an effective way to modulate the magnetism of these 2D materials. Nonmagnetic 2D materials can exhibit magnetism by doping or inducing defects or vacancies, and then their magnetism can be further enhanced by applying strain.\(^{114,115}\) It has been shown that the magnetism of ZZ MoS2 nanoribbons,\(^{116}\) monolayer MoS2 with defects,\(^{117}\) and transition metal-doped monolayer MoS2\(^{118}\) can be greatly enhanced by uniaxial and biaxial tensile strains.

On the other hand, the magnetic properties of intrinsic 2D magnets, such as Curie temperature, magnetic anisotropy, and magnetic exchange coupling, can be efficiently tuned by strain.\(^{119}\) Su et al observed that the Curie temperature of 2D Cr2Ge2Se6 was greatly increased up to room temperature (from 144 K to 326 K) under 3% strain, due to the reduction of energy difference between the 3d orbitals of Cr atoms and the 4p orbitals of Se atoms.\(^{120}\) Morell et al investigated the magnetic-phase transition of antiferromagnetically coupled bilayer CrI3 under strain. It was found that tensile strain could stabilize the antiferromagnetic phase, while compressive strain made the system transform into ferromagnetic phase, which was related to the strain-induced horizontal shift of interlayer.\(^{121}\)
In addition, the effect of strain on the magnetic properties of 2D materials has also been experimentally proven. Buckling-induced strain, such as wrinkles or web buckles, plays an important role in inducing the magnetism to nonmagnetic 2D materials. Tongay et al prepared ReSe$_2$ wrinkles by prestrained Gel-PAK film and confirmed that magnetic properties were induced at the wrinkled region of monolayer ReSe$_2$ by analyzing the relative changes of phase and amplitude of magnetic force microscope (MFM), which is shown in Figure 6(C). DFT calculations indicated that the generation of magnetic moment at the wrinkled region was due to the spin polarization caused by local strain. Xiang et al fabricated MoS$_2$ films by the method of polymer-assisted deposition, and the residual compressive strain was induced in MoS$_2$ due to thermal expansion mismatch. When the flat MoS$_2$ films with residual strain were disturbed by temperature and humidity, it would spontaneously transform to the web buckles of MoS$_2$. It was proved that room-temperature ferromagnetism was induced in the buckled MoS$_2$, and the saturated magnetization was increased by 7.5 times compared with that of flat MoS$_2$. Similar phenomenon has also been observed in the buckled ReS$_2$ system, indicating wrinkles or web buckles are the universal methods to induce magnetism in nonmagnetic 2D TMDs, which may provide an approach for the study of strain-related spintronic devices.

### 3.6 Phase transition

The phase transition of 2D materials involves the overall movement of atomic positions, which can be achieved by applying strain. For 2D TMDs, especially for some MX$_2$-type monolayers (where M is W or Mo, and X can be S, Se, or Te), an important feature is that they usually have multiple possible phase structures, including semiconducting 2H phase with trigonal prismatic structure and...
metallic 1T and 1T' phases with octahedral and distorted octahedral structures, as shown in Figure 6(D). Reed et al predicted theoretically that monolayer Mo- and W-TMDs were likely to achieve the phase transition from 2H to 1T' under uniaxial tensile strain. Among them, MoTe2 was the easiest to achieve this phase transition by the application of tensile strain (0.3–3%). Figure 6(E) shows the gradual process of phase transition when monolayer TMDs are progressively tensioned. First, the 2H phase undergoes elastic deformation without phase transition when applying small strain, and then it reaches a coexistence regime where 2H and 1T' phases coexist in the equilibrium as the strain increases. Finally, the 2H phase completely transforms into 1T' phase. Lee et al applied tensile strain to a MoTe2 flake by AFM tip and achieved experimentally its phase transition from 2H to 1T' under 0.2% tensile strain.125 The structures of 2H and 1T' phases and the diagram of phase transition under in-plane tensile strain are shown in the Figure 6(F). When the strain is released after some time, MoTe2 in the 1T' phase returns to the 2H phase again. Before and after the 2H–1T' phase transition, the conductivity of MoTe2 is changed by 10 000 times (Figure 6(G)), which presents a great application potential in the field of ultrasensitive strain sensors. In addition, Akinwande et al applied hydrostatic pressure using a diamond anvil cell to study the effect of pressure on the structural evolution of monolayer, bilayer, trilayer, and bulk 2H-phase MoS2.126 It was predicted that monolayer 2H-phase MoS2 achieved the phase transition to the 1T' metallic state under the pressure of 67 Gpa. Also, MoS2 with more layers was easier to reach the metallic state, especially for the bulk forms, due to more interlayer interactions.

Recently, the strain-induced phase transition behaviors of 2D materials have become more abundant. The emerging of intrinsic 2D magnets, which may exhibit the combination of atomic-phase transition and magnetic-phase transition under uniaxial and biaxial strains, has bring out more interesting and outstanding physics properties and applications. For example, CrI3 has a unique lattice structure with ferromagnetic ordering in layers and antiferromagnetic coupling between layers. By applying a certain magnitude and direction of in-plane strain, the ferromagnetic phase of CrI3 can be transformed into either the Néel antiferromagnetic or ferrimagnetic phase.127 In addition, similar synchronous phase transition induced by strain can also be observed in Re-doped MoTe2 materials.128 Ma et al found that the structural phase transition from 2H to 1T' occurred under the Re-doped concentration of ~11%, and nonmagnetic Mo1−xRexTe2 monolayer transformed into the ferromagnetism state simultaneously under biaxial strain. The synchronous emergence of the structural and magnetic transition provides great potentials in designing novel spintronic devices and phase-switching devices. In addition, the strain-induced phase transition from semimetal phase to topological insulator phase is theoretically predicted in 1T'-WTe2.129 Monolayer 1T'-WTe2 can convert from a semimetal to an insulator under the compressive or tensile strain along a-axis or b-axis, and the nontrivial Z2 topology phase is still maintained.

4 | APPLICATIONS OF STRAIN ENGINEERING

4.1 | Flexible strain sensors

The piezoresistive effect of 2D materials makes them have a widespread application in flexible strain sensors, which arouse potentials in human health monitoring, motion detection, and electronic skins. The gauge factor (GF) is an important parameter for evaluating the performance of strain sensors, and it indicates the relationship between the resistance variation and the applied strain, which can be defined as

$$\text{GF} = \frac{R - R_0}{R_0} / \varepsilon$$  (8)

where $\varepsilon$ is the applied strain, and $R$ and $R_0$ are the resistance of 2D materials with and without strain, respectively. A larger GF value shows that a smaller strain can cause a larger resistance change, which means higher sensitivity of strain sensors.

As a typical representative of 2D materials, graphene-based strain sensors have been widely studied due to excellent mechanical and electrical properties of graphene. Some graphene strain sensors tend to have low GF due to the minimal impact of strain on the band gap. Huang et al found that the band gap of suspended graphene was almost unchanged when the strain was applied to it by the wedge tip, and its piezoresistive GF was only 1.9.130 Simultaneously, theoretical calculations suggested that only larger strain (more than 26.5% uniaxial strain) can open the band gap of graphene.131

However, other studies obtain higher GF in graphene strain sensors, which seems to disagree with the theoretical prediction. Yu et al transferred monolayer graphene to flexible PDMS substrate to fabricate a graphene strain sensor (Figure 7(A)), where the obtained GF was up to 151.132 As shown in Figure 7(B), the resistance of the sensor first decreased slightly under uniaxial tensile strain below 2.47% and then linearly increased when the strain was over 2.47%. The strain resulted in the lattice distortion of graphene, which not only caused the changes of
band gap but also increased the carrier scattering, reduced the carrier mobility, and thus increased the resistance.

Moreover, initial graphene sheets tend to be connected with each other to form a network during the growing process of graphene film, and therefore strain not only affects single graphene sheet but also changes the overlapping areas between adjacent graphene sheets, which in return modifies the resistance of the entire sensor. As shown in Figure 7(C), when graphene film is subjected to compressive strain or tensile strain, the overlapping areas between adjacent graphene sheets become smaller or larger, which increases or decreases the resistance, respectively. Based on this mechanism, Xu et al used partially overlapped graphene sheets in the slide rheostat for stretchable strain sensors, and the piezoresistive sensitivity of these sensors was much higher than that of stacked graphene strain sensors. In Figure 7(D), the adjacent graphene sheets slipped relatively to each other under tensile strain, resulting in the decrease of contact area and the increase of resistance. It was proved that this graphene slide rheostat had a high tensile rate (70%), excellent cyclic stability (300 000 cycles), as well as stable response in the frequency range of 0.1–5 Hz and the temperature range of −45 to 180 °C, thus providing full-scale human motion monitoring (Figure 7(E), (F)).

Making use of the piezoresistive effect, many other 2D-material-based strain sensors have also been investigated, which show great potentials in practical application as well. Kis et al demonstrated that the piezoresistive effect in MoS2 originated from the change of band gap at the action of strain, and the GF of monolayer, bilayer, and trilayer MoS2 was $-148 \pm 19$, $-224 \pm 19$, and $-43.5 \pm 11$, respectively. The piezoresistive effect is associated with the crystal structure, leading to an anisotropic piezoresistance sensitivity along different crystal orientations in low-symmetry 2D materials. Zhang et al observed a room-temperature piezoresistive effect in BP with the GF being up to 185 along the ZZ direction. Particularly, the opposite anisotropic piezoresistive effect was observed in ReS2 under uniaxial strain, and the GF along a- and b-axes was 50.14 and $-60.49$, respectively, ascribing to the
opposite strain effect along different axes on the band gap.\textsuperscript{135} In addition, Liu et al. fabricated a SnS\textsubscript{Se} alloy nanosheet-based strain sensor with a GF being up to 69.7 and indicated that it was possible to conceive tunable strain sensitivity by changing the elemental constituents owing to its composition-dependent tunable band gap of 2D alloy material.\textsuperscript{136} Furthermore, the GF can be tuned by the gate voltage, which significantly increases the sensitivity of strain sensors. Vogel et al. fabricated strain sensors based on uniform wafer-scale trilayer MoS\textsubscript{2} and found that the modulation of the Fermi level by a gate bias increased the GF by an order of magnitude.\textsuperscript{137} The gate-tunable GF is convenient for practical sensing applications, as the higher strain sensitivity can be obtained by directly changing the gate bias in FET devices.

Recently, strain sensors based on the composite structure of polymers and 2D materials are designed, which usually display a high GF. The piezoresistive effect of composite materials is affected by the combine effect between the resistance of filled particles and the change of inter-particle tunneling resistance. Coleman et al. prepared composite materials of polyethylene oxide and liquid-exfoliated MoS\textsubscript{2} nanosheets and found that dynamic strain sensors based on the composite structure significantly broadened the sensing capabilities.\textsuperscript{138} The addition of MoS\textsubscript{2} nanosheets not only greatly increased the conductivity but also resulted in the reinforcement of composite materials, which meant better stress transfer between polymers and nanosheets. When applying a relatively small tensile strain, the resistance of entire strain sensors decreased, which mainly depended on the resistance reduction of MoS\textsubscript{2} nanosheets caused by the strain-induced change of band gap. However, when a larger tensile strain was applied, the resistance of composite materials distinctly increased due to the increased contact resistance of adjacent nanosheets under strain. Then, the so-prepared strain sensors with higher GF were used to measure the periodic deformation at a small strain.

In addition, the piezoelectric effect of 2D materials, which modifies the interface Schottky barrier through the strain-induced polarization charges, can also be used in the design of strain sensors. Yu et al. prepared a graphene/MoS\textsubscript{2} junction and revealed that the strain-induced polarization charges in MoS\textsubscript{2} adjusted the Fermi level of graphene and further modified the Schottky barrier in graphene/MoS\textsubscript{2} junction.\textsuperscript{139} The GF of strain sensors based on graphene/MoS\textsubscript{2} junction is as high as 575 294.

In particular, when strain sensors are integrated into an array, the strain sensitivity becomes more precise, further providing their applications in electronic skin and other wearable devices. Ahn et al. fabricated an ultrathin MoS\textsubscript{2} tactile sensor array with a patterned graphene electrode (Figure 7(G)).\textsuperscript{28} This tactile sensor was transformed on a fingertip (Figure 7(H)), and then the strain distribution was mapped by pressing the stylus pen, where the position of fast-moving pen was well tracked. This sensor array presented high light transmittance (over 80%), sensitivity (GFs \(\approx\) 50–70), and good linear response characteristics even after 10 000 cycles below 1.98% strain. The thickness of sensor array was only 75 nm, which was very suitable for the application of electronic skin.

### 4.2 Flexible photodetectors

Graphene and other 2D materials have been extensively investigated in the application of advanced photodetectors due to their intriguing optical and mechanical properties. However, the atomically thin thickness of 2D materials leads to weak light adsorption, which further limits the photoelectric conversion efficiency. In order to improve light absorption ability, periodic wrinkles of 2D materials are designed by the methods of strain engineering, and thus the performance of photodetector is significantly improved. For example, the light absorption of graphene is weak in the IR to visible range, which severely limits its application in photodetectors. Nam et al. prepared crumpled graphene by the method of prestretching the flexible substrate (Figure 8(A)) and found that the optical extinction was increased by over an order of magnitude compared to flat graphene (Figure 8(B)).\textsuperscript{29} It was demonstrated that the photoresponsivity of crumpled graphene photodetectors was higher than that of flat graphene photodetectors, and it exhibited the advantages of strain tunability, wavelength selectivity, well scalability, and good stability. Particularly, the Fermi level of the strained region of wrinkles shifts, which leads to the difference in electrical potential between the localized strained region and unstrained region.\textsuperscript{140} Therefore, a quantity of back-to-back built-in electric field forms in the wrinkled nanosheet, resulting in the enhanced separation of photogenerated carriers and the decreased dark current, thus improving the performance of photodetectors.

The strain-induced changes of lattice structure and band gap can also affect the performance of photodetectors. The narrowed band gap caused by tensile strain can broaden the wavelengths of absorbed light, then the photoresponsivity is also increased. Ho et al. grew wafer-scale monolayer WS\textsubscript{2} by enhanced CVD and fabricated its photodetectors on rigid sapphire substrates and flexible PI substrates.\textsuperscript{141} It was reported that the photoresponsivity of flexible WS\textsubscript{2} photodetectors was nearly 10 times that of rigid WS\textsubscript{2} photodetectors, attributing to the enhanced light absorption of monolayer WS\textsubscript{2} by the rough surface of flexible PI substrates, as well as the reduced recombination of photogenerated
carriers caused by the carrier traps at the interface defects. In addition, the photocurrent of flexible photodetectors increased with decreasing the bending radius, meaning that it increased with increasing uniaxial tensile strain. On one hand, the tensile strain reduced the band gap of monolayer WS2 and caused the red shift of absorption spectrum, which enhanced the absorption of light below 532 nm and thus improved the internal quantum efficiency. On the other hand, the tensile strain caused the lattice structure to be distorted and the photogenerated carriers to be trapped by the defects, thus reducing their recombination probability and leading to the increased photocurrent.

In addition, the strain-induced piezoelectric effect can be used to adjust the Schottky barrier at the interface of heterojunction and thus greatly improves the performance of flexible photodetectors. Wang et al utilized the piezophototronic effect of monolayer MoS2 to build a flexible photodetector, and its photoresponsivity reached to the maximum of 2.3 × 10 A W⁻¹ under a uniaxial compressive strain of −0.38%. As shown in Figure 8(C), in the absence of strain, both the external electric field and the built-in electric field drove the photogenerated carriers to generate photocurrent (Iph). When tensile strain was applied, positive and negative polarization charges were generated at the drain/MoS2 and MoS2/source interfaces, respectively, and the Schottky barrier at the corresponding interface was increased and decreased, which prevented the separation of photogenerated carriers, resulting in the reduction of Iph. When applying a relatively small compressive strain, the polarization charges caused the energy band to tilt reverse, which promoted the separation of photogenerated carriers, resulting in an increase of Iph. However, when applying a larger compressive strain, a new energy barrier formed at the drain/MoS2 interface due to the high-density polarization charges, which in turn caused Iph to decrease again. In addition, Wang et al prepared WSe2/CdS mixed-dimensional heterojunction flexible photodetectors and demonstrated that there was a great enhancement of photoresponse under strain due to the piezoelectric effect. When applying compressive strain of −0.73%, the polarization charges at the WSe2/CdS interface caused the tilting of local energy band, thus promoting the transport of photoexcited carriers, which gave rise to the increased photocurrent (0.65 nA) and photoresponsivity (33.4 A W⁻¹).

Moreover, the modulation mechanism of strain-induced band gap change and Schottky barrier change on the performance of photodetectors is deeply investigated. For CVD-grown monolayer MoS2 photodetectors, when both the strain and channel were along the ZZ direction, only the piezoresistive effect caused by the change of band gap was observed, and the light–dark current ratio, self-powered current, and photoresponse speed were all significantly improved. However, when both of them were along the AC direction, the piezoelectric effect by the change of Schottky barrier was more dominant, and yet the photoresponsivity was increased. The optoelectronic performance of 2D materials can be improved by both the piezoelectric effect and piezoresistive effect, which is of great importance for exploring the further application of photodetectors.

5 Conclusion and Outlook

Strain engineering, as an important strategy for tuning the electronic structure and properties of 2D materials, has attracted extensive research interests. There are a variety of methods to induce strain to 2D materials due to their
atomic-scale thicknesses and strong in-plane and out-of-plane deformation capacity. Furthermore, 2D materials are more sensitive to strain and even small strains can change their lattice structure, which significantly modulate their physical properties. Consequently, the introduction of many excellent properties largely expands the application of 2D materials in the fields of flexible strain sensors, flexible photodetectors, and other wearable optoelectronic devices. However, although strain engineering of 2D materials has made great progress, there are still many problems expected to be solved. Despite the strong deformation capacity of 2D materials, the strain in 2D materials can hardly reach the theoretically predicted value. One of the important reasons is that the interfacial interaction strength between the substrate and 2D materials is not clear, resulting that the strain in the deformed substrate cannot be effectively transferred to 2D materials. As a result, it is particularly important to study the interface mechanical behavior between 2D materials and the substrate, especially the interface adhesion and friction behaviors. Then, exploring new methods or optimizing existing methods to induce strain to 2D materials are also of great importance to achieve a larger range of strain engineering in future. In addition, strain engineering has some limitations on the modulation of some properties of 2D materials. For example, the strain modulation of magnetism of 2D materials mainly focuses on the theoretical calculation level, and it is difficult to be achieved in experiment, which greatly limits further application in spintronics. The applied strain cannot reach the critical value necessary to change the physical properties of some 2D materials. The next step requires to further understand the intrinsic relationship between structure and property of 2D materials under the action of strain, so as to make more breakthrough in the strain-induced property modulation. In the application of flexible devices, due to weak adhesion between the metal electrode and flexible substrate, it is easy to fall off from flexible substrate, which greatly influences the stability and the working life of devices. Therefore, further improvement of integration technology is needed for the better practical application of various flexible devices.

CONFLICT OF INTEREST
The authors declare no conflict of interest.

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