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Viewpoint

Precision Chemistry in Two-Dimensional Materials: Adding, Removing, and Replacing the Atoms at Will

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1. INTRODUCTION

Two-dimensional (2D) materials are intriguing for wide applications such as electronics, optics, energy, catalysis, and so forth, which has rapidly fueled the development of nanoscience and technology in recent decades. Because of their reduced dimensionality, large specific surface area, and atomic thickness, various novel behaviors and properties emerged, though the synthesis, preparation, and modification of 2D materials remain challenging on the atomic scale. Many conventional processing approaches developed from 3D bulk materials are no longer applicable to 2D materials. For instance, the ion implantation technique favored by the semiconductor industry is incompatible with 2D materials due to the vulnerable 2D atomic structures. For a similar reason, wet chemical treatments and chemical annealing approaches are not recommended for 2D materials. Therefore, developing precision chemistry down to an atomic scale is highly desirable for 2D materials. In this viewpoint, we will address the atomicwise chemical approaches and reactions which are suitable for or even exclusive to 2D structures. These approaches work as a "lancet" which can add, remove, and replace the desired atoms in 2D materials at will, with the rest of the atoms well preserved (Figure 1). In this way, the physical and chemical properties can be finely tuned, and various new properties can emerge. We believe the rise in precision chemistry for 2D materials will enable a boom in relevant fields in materials science, physics, and engineering.

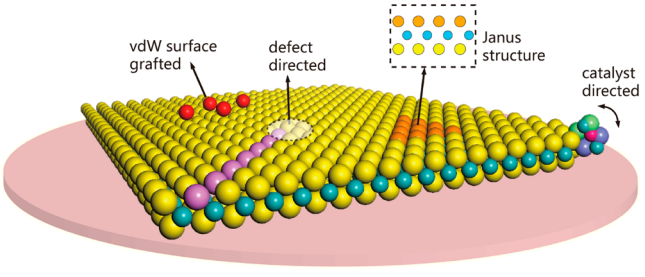

Figure 1. Scheme of some 2D structures enabled by precision chemistry.

2. APPROACHES FOR PRECISION CHEMISTRY IN 2D MATERIALS

2.a. Plasma-Assisted Methods

Plasma can be used as an effective technique to treat 2D structures, including etching, doping, phase transition, oxidation, and so forth. Beyond these, plasma can also be employed to selectively remove atoms layer by layer in those sandwiched 2D structures, such as transition-metal dichalcogenides (TMD). Taking advantage of the large absorption energy of hydrogen atoms at the vacancy sites of the MoS_2 lattice, remote hydrogen plasma can be introduced to strip off the top layer of sulfur atoms from the MoS_2 monolayer at a certain power and duration, with the top layer of sulfur atoms fully replaced by H atoms, forming an H−Mo−S sandwich.[1] Afterward, the layer of H atoms can be replaced by Se atoms after a thermal selenization process, resulting in the structurally stable Janus MoSSe monolayer in which the Mo atoms are covalently sandwiched by one layer of S atoms and another layer of Se atoms (Figure 2).

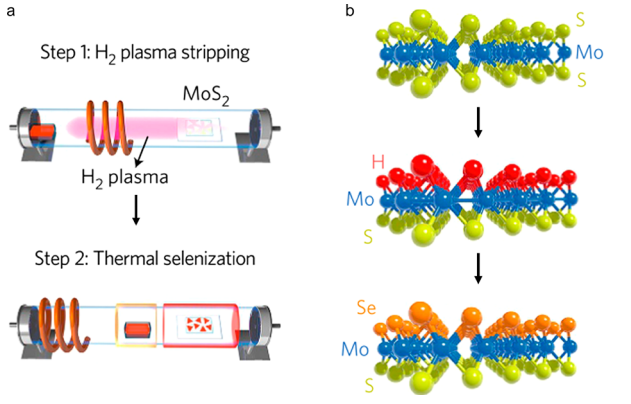

Figure 2. (a) Two-step plasma-assisted chemical vapor deposition method. (b) 3D atomic structure illustration of the MoS_2 monolayer, Janus H−Mo−S monolayer, and Janus Se−Mo−S monolayer. (Reproduced with permission from ref 1. Copyright 2017 Springer.)

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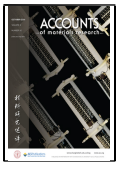

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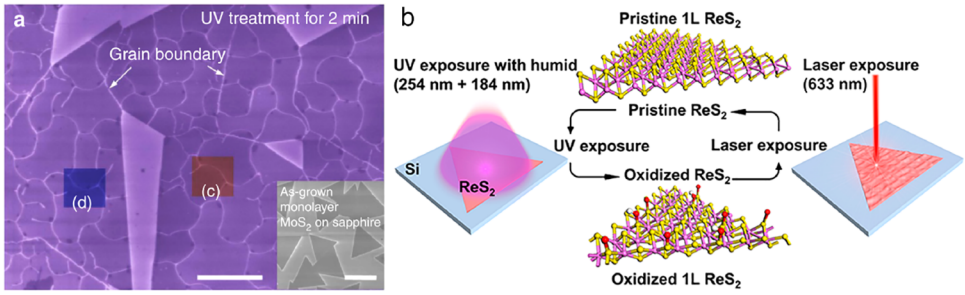

Figure 3. (a) SEM image of the crack pattern generated on MoS_2 after UV light exposure under humid conditions. (Reproduced with permission from ref 2. Copyright 2017 Springer.) (b) Schematic illustration of the reversible photochemistry method, showing the UV treatment in a moist environment and the laser irradiation recovery process. (Adapted from ref 5. Copyright 2021 Wiley.)

2.b. Mechanochemical Methods

The flexible 2D lattice enables versatile strain engineering in 2D materials, such as wrinkling, crumpling, cracking, and bending, which are the fundamentals of 2D material-based flexible functional devices. Different strain states lead to distinct chemical activities in 2D layers. For instance, the crack tip zones (highly strained zones) in 2D MoS_2 are vulnerable to attack by reactive species.[2] These crack patterns simultaneously generated by strain and a reactive chemical environment show impressive curvatures which are absent in straight cracks in vacuum (Figure 3a). The misfit interfacial strain between the 2D layers and substrates can also create various subdomain structures,[3,4] thus providing different platforms for intentional chemical doping, functionalization, and so on.

2.c. Photochemical Methods

The 2D materials show less tolerance for high-energy processing, which would cause irreversible damage to the structure or uncontrollable defects, making the doping of 2D material a big challenge. To address such an issue, another way is to graft the dopant atoms onto the van der Waals surface through a gentler procedure instead of accessing the 2D lattice. For instance, under ultraviolet light irradiation with controlled humidity, highly oxidative O_3 and oxygen-related radicals can be created and quickly react with the 2D ReS_2 or $ReSe_2$ lattice, resulting in the grafting of oxygen radicals on the van der Waals surface, which would act as a hole donor to the 2D materials, as evidenced by the p-doping behavior in the field-effect transistor device results.[5] More interestingly, a laser annealing treatment can be used to remove such covalent oxidation on the vdW surface without generating any atomic defects, and the optical and electric transport performance can simultaneously recover to the pristine status (Figure 3b). Such a reversible redox photochemistry method is a tuning nob allowing scalable doping and property modulation on industry scales.

2.d. Catalytic Methods

In general, 2D material growth either follows the Wulff construction in equilibrium or shows more stochastic kinetic morphologies. This means that except for the equilibrium polygon shapes which depends on the free energies of the edges, it is rather difficult to yield other shapes of 2D materials in a controllable manner. However, introducing a catalyst particle could be an effective way to break the Wulff construction rule. For instance, by using potassium chloride (KCl) as the catalyst and combined with a plasma pretreatment on growth substrates, high-index edges and unusual shapes of 2D MoS_2 can be obtained. A proposed vapor−liquid−adatom−solid (VLAS) growth mechanism can be used to explain such interesting behavior (Figure 4a). Briefly, the transport and anchoring of adatoms during growth can be guided by such mobile catalytic particles[6] (Figure 4b).

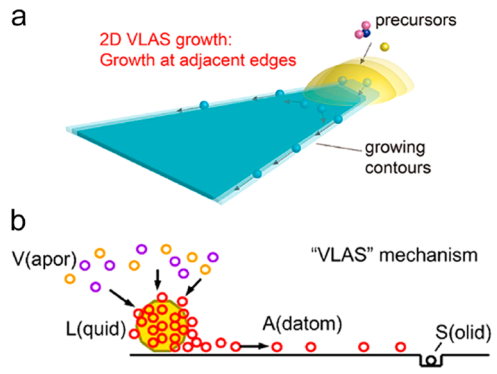

Figure 4. (a) Schematic illustration of the KCl-catalyzed growth of the 2D MoS_2 nanosheet. (b) Schematic illustration of the VLAS mechanism. (Reprinted with permission from ref 6. Copyright 2020 American Chemical Society.)

2.e. Defect-Directed Methods

There are diverse kinds of defects existing in 2D materials, including vacancy, step, twinning, grain boundary, and so forth. Normally, defects are undesired, but they can act as agents to aid the synthesis or processing of 2D materials, especially in atomic precision. For example, attributed to the lattice mismatch between MoS_2 and WSe_2, many interfacial misfit dislocations are formed during the synthesis of the WSe_2/MoS_2 lateral junction.[7] During the second step of growth, MoS_2 prefers the higher reactive dislocation cores, thus pushing the dislocations away from the original interface, forming 1D MoS_2 subnanometer channels in the WSe_2 lattice. Furthermore, the artificial dislocation in the parent WSe_2 lattice can benefit the formation of 1D nanostripes with a high density of impurity atoms since the dislocation cores can either climb or migrate under the driving force to fill the vacancy lines, which enables an effective doping strategy for 2D crystals[8] (Figure 4c). Defects from the substrate can also be employed for precision chemistry control. Because of the 6-fold symmetry of Cu(111), in most cases the van der Waals growth of hBN on Cu(111) will produce two sets of configurations with different orientations but almost degenerate binding energies. Such characteristics are fundamental obstacles in achieving wafer-scale single-crystal hBN.[9] However, this serious problem can be well solved by the atomic step edges on Cu(111), which help to separate the binding energy of those two kinds of configuration by 0.23 eV, ensuring the mono-orientated

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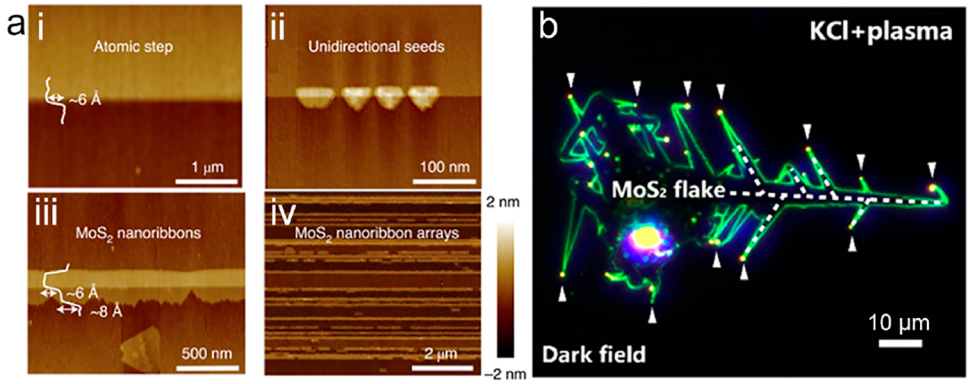

Figure 5. (a) AFM images showing the synthesis evolution of a well-defined MoS_2 nanoribbon along the steps on the β-Ga_2O_3 substrate: (i) atomic step on the β-Ga_2O_3 substrate, (ii) nucleation of MoS_2 seeds along the edge, (iii) a continuous MoS_2 nanoribbon, and (iv) a dense array of aligned MoS_2 nanoribbons. (Reprinted with permission from ref 12. Copyright 2020 Springer.) (b) Dark-field OM image of MoS_2 spikes. (Reprinted with permission from ref 6. Copyright 2020 American Chemical Society.).

growth of hBN on Cu(111) at the wafer scale. This achievement is an important milestone toward the wafer-scale integration of 2D electronics.

3. EMERGENT 2D STRUCTURES ENABLED BY PRECISION CHEMISTRY

Precision chemistry in 2D materials can result in unprecedented 2D structures, expand the family of 2D materials, bring about new functionalities, and enhance the quality, homogeneity, and purity of 2D materials. Herein we give a few recent examples.

3.a. Chemical Grafting on van der Waals (vdW) Surfaces

Chemical grafting is an effective approach for doping 2D materials, without introducing distinct lattice defects. The dopants for grafting can vary from atoms to molecules or functional groups, thus allowing plenty of room to tailor the properties of 2D host materials. For example, oxygen radicals can be generated onto the surface of ReS_2 during UV treatment in a humid environment. Such grafted oxygen can act as a hole donor to ReS_2, resulting in p-doping behavior.[5] Similar chemical grafting was also observed in bilayer graphene after exposure to XeF_2, as evidenced by cross-sectional HRTEM, and F atoms were found to be bound to each graphene layer from both sides after fluorination.[10]

3.b. Chemical Intercalation

Owing to the weak vdW interaction between adjacent 2D layers, exotic or native atoms can be intercalated in between the vdW layers and modulate the phases and properties. Recently, Zhao et al. demonstrated the self-intercalation of native atoms into bilayer TMDs via molecular beam epitaxy. Moreover, the intercalation can be achieved not only by the vapor phase but is also applicable to solution-based approaches.[11]

3.c. Janus 2D Layers

Janus 2D structures, in which the transition-metal layer is sandwiched by two different kinds of chalcogenides, create vertical inversion asymmetry. The Janus 2D structures are suitable for investigating symmetry-sensitive properties and applications. For instance, strong second-harmonic generation and piezo responses were demonstrated in the Janus MoSSe structure.[1]

3.d. Directed Ribbons and Spikes

Shape engineering is also a crucial topic in 2D material research. Two-dimensional nanoribbons with a high aspect ratio could offer unpredictable opportunities for investigating quantum confinement physics since there are two dimensions of such structures being well confined. Very recently, well-aligned ultralong MoS_2 nanoribbons were obtained by taking advantage of the atomic step on the β-Ga_2O_3 substrate (Figure 5a), and the obtained structure is highly crystallized and has good electronic transport resonance.[12] With the help of a catalyst, abnormal 2D MoS_2 spikes can also be obtained[6] (Figure 5b).

3.e. Directed Alloys

An alloy is an efficient way through which extra atoms can be doped into the 2D in-plane lattice, resulting in the modulation of electronic and optical properties. For the case of WSe_2, selenium vacancies can be created after annealing the sample with hydrogen, and then extra atoms can be bound at these vacant sites, thus completing the doping procedure and forming the 2D alloy structures. Interestingly, such extra dopants are not distributed homogeneously but form a doping stripe line in the host lattice. Such unique behavior can be explained by the climbing and migration of the dislocated cores due to the driving force to fill the vacancy lines.[8] Such directed alloys may complement traditionally doped alloys such as specific metal atoms (e.g., vanadium) alloyed with TMD crystals through a mild solution mixing and thermal annealing process.[13] Enhanced electrocatalytic[14] and ferromagnetism[15] behavior would emerge in some intentionally doped 2D alloys.

4. OPPORTUNITIES AND APPLICATIONS FOR 2D STRUCTURES DESIGNED BY PRECISION CHEMISTRY

4.a. Electronics

Electronic doping can be facilely achieved via vdW surface grafting, such as oxygen radicals grafted onto the surface of ReS_2 via UV treatment donating holes to the n-type device channel, thus decreasing the concentration of the majority charges, the source-drain current, and the carrier mobility. While these oxygen radicals are removed by laser annealing, the current and mobility show reliable recovery. The hole-doping effect is evidenced by the observation of ambipolar charge-transport behavior in the FET configuration devices.[5]

4.b. Thermoelectrics

The thermoelectric performance can be improved by increasing the electrical conductivity and decreasing the thermal conductivity, but these two factors cannot be tuned

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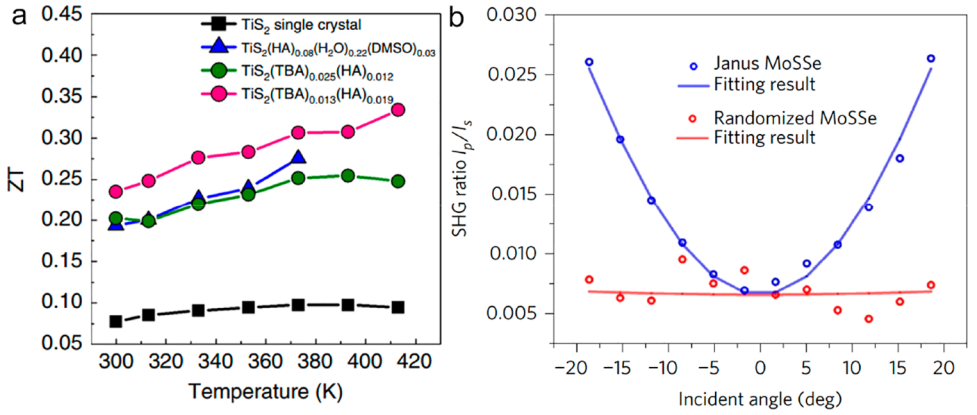

Figure 6. (a) In-plane thermoelectric figure of merit of TiS_2-based inorganic/organic superlattices. (Adapted from ref 14. Copyright 2017 Springer.) (b) Angle-dependent SHG intensity ratio between p and s polarization in the Janus MoSSe and randomized alloy samples. (Adapted from ref 1. Copyright 2017 Springer.)

independently and are limited by the Wiedemann−Franz law. However, this issue can be solved by intercalating organic cations into the inorganic 2D layered structure, which acts as an ideal electron-transmitting but phonon-blocking structure to enhance the thermoelectric figure of merit value.[16] For the case of tetrabutylammonium (TBA) and hexylammonium (HA) double organic cations intercalated into 2D TiS_2, one can modulate the carrier concentration through simply heating the structure to an intermediate temperature, and the lower-boiling-point HA molecules can be selectively deintercalated. Therefore, the ZT value can be improved to 0.33 at 413 K for the $TiS_2(TBA)_{0.025}(HA)_{0.012}$ sample.[17]

4.c. Optics

Except for the in-plane asymmetry, the Janus 2D MoSSe monolayer also has out-of-plane inversion asymmetry, which originates from the imbalance of the electronic wave function over the S and Se atoms, resulting in an out-of-plane optical dipole transition. Under an angle-resolved polarization-selective SHG measurement setup, obvious angle-dependent evolution of the SHG intensity can be observed in the Janus 2D MoSSe (Figure 6b), while no detectable SHG signal can be seen in the randomized MoSSe alloy, indicating the existence of such vertical dipoles.[1]

4.d. Magnetics

Through a self-intercalation procedure, additional Ta atoms can be inserted into the gap of the TaS_2 host material. For the Ta_7S_{12} composition, the intercalated Ta atoms would introduce additional spin-split bands across the Fermi level, thus forming a magnetic ground state, as evidenced by the observation of linear magnetoresistance of up to 9 T at low temperature.[11]

4.e. Catalysts

Two-dimensional materials offer extensive possibilities for active-site (usually metal atoms) and substrate engineering for high-efficiency catalysts. For instance, under microwave irradiation, Pt atomic sites can be anchored on aniline-stacked graphene. This catalyst shows excellent HER activity and superior stability due to the high electrical conductivity of graphene and the improved electronic structure and hydrophilicity of Pt by aniline.[18]

5. CONCLUSIONS AND OUTLOOK

The precision chemistry in particular means well-controlled chemical reactions, which selectively add, remove, or replace designated atoms or a designated group of atoms in two-dimensional materials. This viewpoint is mainly motivated by the critical challenges in 2D material preparation/modification currently. Because of the large specific surfaces and atomic thickness, the conventional chemical processes are genuinely hard to control on 2D materials. The problems associated with reproducibility, inhomogeneity, defects, and purity remain the most significant barriers for the field of 2D materials chemistry. These challenges are inherent for 2D materials and brought about by basic thermodynamic principles. In this instance, here we propose that the chemistry in 2D materials should go beyond normal requirements/standards and normal routes to overcoming these inherent challenges. Fortunately, more and more "lancets" which can precisely "cut" or "graft" the chemical bondings on 2D surfaces or 2D edges have been invented, and empowered by this, a brighter future for 2D materials can be foreseen.

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Notes

The authors declare no competing financial interest.

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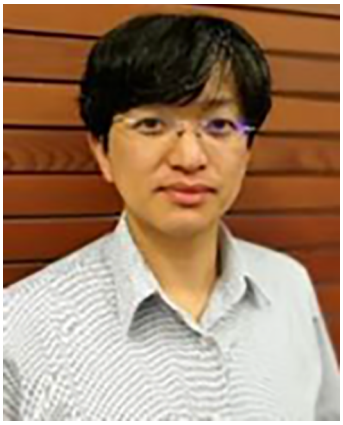

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