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New insights into the interactions between two-dimensional ice and two-dimensional materials

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Abstract

Water is one of the most essential substances for life on Earth and plays a vital role in both natural and technological processes. Recently, there has been growing interest in studying the behavior of water molecules in confined spaces, particularly in low-dimensional materials and structures. Regardless of whether it is in the form of gas, liquid, or solid, water can interact and form interfaces with many low-dimensional structures. Given the current controversial understanding of two-dimensional (2D) ice and the increasing interplay between water/ice and 2D materials such as graphene and transition-metal dichalcogenides, we provide a brief overview of recent progresses on the interfaces of 2D ice and 2D van der Waals layered materials. This review highlights their potential contributions to the breakthroughs in tribology, membrane technology, nanofluidic, and nanodevice applications. Of particular interest is the recent discovery of ultrahigh lubricity between 2D ice and 2D layered materials, as well as the ability to modulate the surface adhesion between layers. These findings have the potential to enable new technological advances in both electronics and various industries. Meanwhile, this rapidly evolving field presents its own challenges, and we also discuss future directions for exploiting the interactions between 2D ice and 2D layered materials.

INTRODUCTION

Two-dimensional (2D) layered materials have great significance in the field of material science and technology due to their exceptional properties and unique atomic structures.[1–4] The ultrathin thickness of these materials results in a high surface-to-volume ratio, which provides a large area for interaction with other materials or environments.[5–7] This characteristic can enable the development of various sensing[8,9] and electrocatalytic/photoelectrocatalytic applications.[10,11] Moreover, 2D layered materials have a vast range

of electronic, optical, and mechanical properties that can be tuned by altering their structures and morphologies,[6,12–14] making them highly versatile for a wide range of applications like electronics, energy, tribology, and medicine.[15,16] Despite their unique properties, the practical applications of 2D layered materials face challenges due to high surface instability.[17–19] As a result, predicting their behavior with different interfaces and engineering their surfaces with specific properties may prove challenging.

On the other hand, water, as an essential and fundamental requirement for life, has captured the attention of researchers for centuries due to its complex behaviors. In the past decade, with the growing interest in the field of 2D materials, attention has shifted towards the structure and properties of 2D ice,[20–23] which differ significantly from common bulky ice. In principle, a droplet incapsulated by a solid 2D system can be transformed into 2D ice under sufficient pressure or compression force (Figure 1a).[16,25–28] Depending on confinement conditions, the thickness can be varied from ca. 0.25 nm, equal to ultra-flattened monolayer 2D ice,[20,29,30] to a few nm (Figure 1b).[16] The most well-established hexagonal structure of 2D ice[21,31,32] (Figure 1c) consists of two planes of water molecules, where each molecule forms three hydrogen bonds (H bonds) with in-plane neighboring molecules and an H bond with molecules in the opposite plane; therefore, the layer was "interlocked" without any free hydrogen dangling bonds on the surface, similar to the surface of 2D layered materials. Due to the high-density "interlocked" H bonds, this 2D hexagonal structure can be flattened by itself on a planar surface and has a higher melting point[33,34] than the typical 3D (bulk) hexagonal (I_h) phase. Except for this, the formation of other 2D ice phases like pentagonal, square, and rhombic structures depends on pressure and temperature, which has been predicted theoretically (Figure 1c).[21,27,35–37] It is worth noting that the superionic phase of 2D ice was expected to show superconductive behavior at temperatures above 400 K.[27,38,39] However, to date, only the hexagonal and pentagonal structures of 2D ice on metal surfaces have been experimentally observed (Figure 1d,e).[24,32] Other 2D ice structures are unlikely to be verified experimentally at present due to the extreme conditions required and the difficulty in imaging atomic structures in confined spaces.

Owing to its atomically flat and robust surface,[40,41] as well as the van der Waals (vdW) interface between layers,[14,42] 2D layered materials can serve as the ideal platform to realize dimension-confined 2D ice.

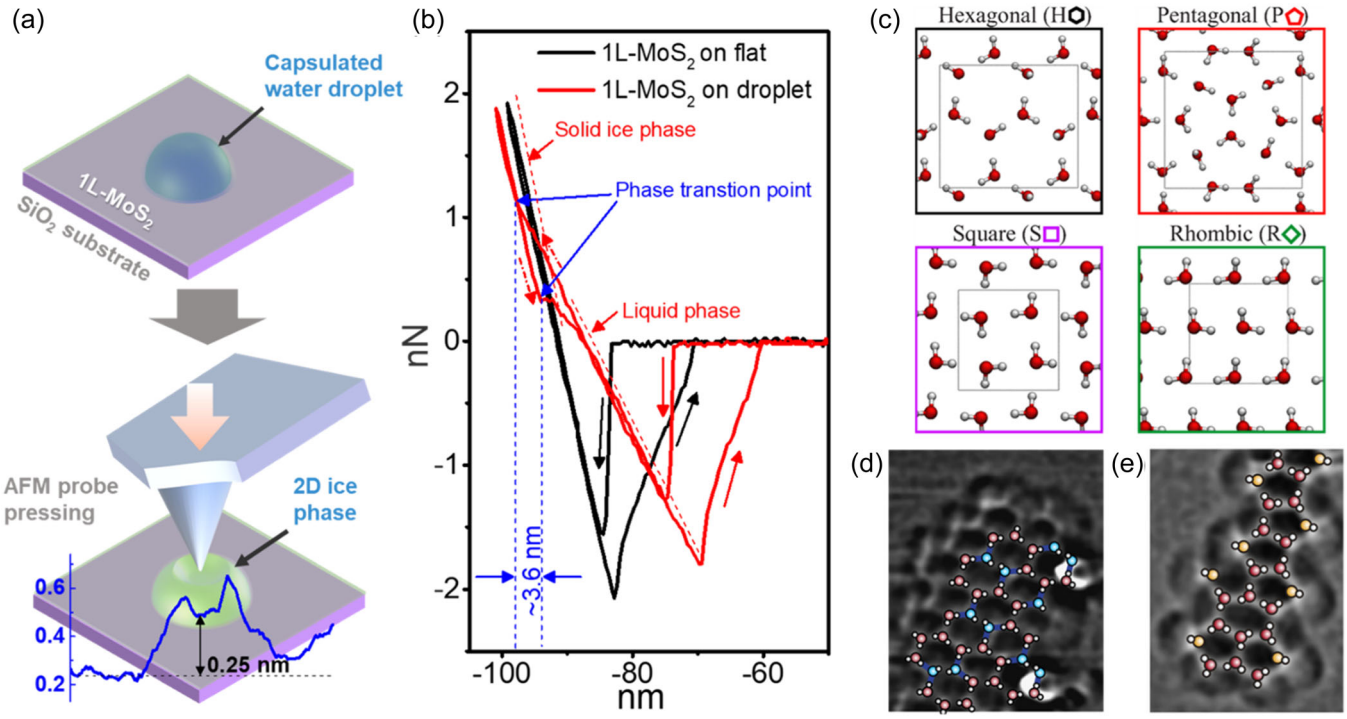

FIGURE 1 Phase transition under compression and phase structures of confined two-dimensional (2D) ice. (a) The schematic illustrates how a water droplet was confined under single-layer (1L) MoS_2. When an AFM probe scanned over the droplet, it enabled water-to-ice transition under the compressive force. The line profile shows that the lowest point in the middle of the water droplet is about 0.25 nm, which is equal to the 2D ice single-layer thickness. (b) Force curves of 1L-MoS_2 on the substrate and on the water droplet. The smaller slope in the red line corresponds to the compression stage on the liquid-phase water encapsulated in the bump, while the high slope corresponds to the solid 2D ice phase, almost identical to the force slope on flat MoS_2 (black line). Adapted with permission.[16] Copyright 2023, American Chemical Society. (c) Different computational phase structures of 2D ice including hexagonal ice, pentagonal ice, square ice, and rhombic ice. Adapted with permission.[18] Copyright 2016, American Physical Society. (d, e) AFM images of hexagonal and pentagonal structure of 2D ice obtained on a metal surface. Adapted with permission.[24] Copyright 2017, Springer Nature. AFM, atomic force microscope.

When combining 2D ice and 2D vdW layered materials, new phenomena emerge at the interface and unlock the potential applications of both materials. For instance, water vapor can be condensed into hexagonal 2D ice on the hydrophobic surface of graphite and hexagonal boron nitride (h-BN).[32,43] Humidity can also be trapped at the vdW interface between graphene and single-layer molybdenum disulfides (MoS_2).[34,44,45] It can accumulate as liquid droplets that are sealed between the 2D layers[25] or transform into different phases of 2D ice through continuous or discontinuous transitions (Figure 1a).[16,26,46] On the other hand, atomically thin self-flattened 2D ice can serve as a supportive layer to reduce the instability of 2D layered materials, thereby improving their intrinsic properties. In this context, recent studies have revealed intriguing findings on confined 2D ice, such as its ability to enhance lubricity on the surface of 2D layered materials.[16] Furthermore, novel approaches have emerged for the clean transfer[47] and tribological applications of 2D layered materials, leveraging the unique properties of confined 2D ice.

LIMITATIONS IN EXISTING THEORETICAL AND EXPERIMENTAL STUDIES

In principle, when molecules are confined to a thickness of less than 4–6 molecular layers, a transition to a solid state can occur. This is because the transverse density profile of the confinement plates induces planar order to the confined molecules, resulting in solidification. This phenomenon was initially observed in the 2D ice confined in a mercury/water/mercury junction.[48] The transition from liquid water to 2D ice was recognized by an increase in the out-of-plane elastic modulus as the confined space decreased (see Figure 1b).[16,48] Later, Zangi and Mark conducted molecular dynamics (MD) simulations to explore the ordered structures of 2D monolayer and bilayer ice in confined spaces.[20] They observed that the transition from water to ice was indicated by a four- to fivefold reduction in the lateral diffusion coefficient of water, when the out-of-plane dimension was confined to the range of 0.51–0.55 nm, regardless of external pressure.[20,49] However, these early theoretical studies did not account for the characteristics of materials acting as confinement walls or wall-to-wall interactions at the nanoscale level, limiting their correlation with experimental studies.

Moreover, MD simulations have been developed to predict the properties of 2D ice/water confined within specific 2D layered materials such as graphene,[50] MoS_2,[33] and h-BN.[51] These simulations have considered various external conditions and vdW interactions at the respective interfaces. For example, water molecules confined between two single-layered graphene sheets can withstand an internal pressure of approximately ~1 GPa due to the vdW force between the layers, resulting in the formation of 2D cubic ice.[50,52] Although the theoretical prediction of 2D cubic ice structure existed, doubts were raised about whether the experimental results genuinely represented ice confined between two monolayers of graphene. Such suspicions were reasonable, given that the choice of water force field models could alter the 2D ice phases obtained from MD simulations.[50,53] While introducing density functional theory (DFT) to predict the structural relaxation and energy profile of the initial system can improve accuracy,[54–56] it comes with a trade-off of increased computational cost and restrictions under certain temperature and pressure conditions.

Besides abundant theoretical studies, experimental studies of confined 2D ice have been limited by the difficulty in characterizing the dynamics of water using available equipment. Transmission electron microscopy (TEM), for instance, has difficulties in identifying the exact structures of confined 2D ice.[52] The main challenge stems from the fact that hydrogen atoms in water molecules are almost undetectable by TEM or any other atomistic-scale imaging system. Additionally, 2D ice is extremely vulnerable to irradiation and damaging effects from high-energy incident beams.

An ice-like 2D structure was also observed at the hydrophobic graphene–MoS_2 interface at room temperature using an atomic force microscope (AFM).[25,34] The 2D hexagonal structure of 2D confined ice was evidenced by the distribution of the angles between two adjacent island edges that showed the dominant peak at 120°.[25] The 2D hexagonal ice had a maximum thickness of 0.56 ± 0.02 nm,[25,34] which agreed with the critical confined space theoretically determined by Zangi and Mark.[20] However, while existing studies have established the novel structure and properties of various 2D ice phases under certain initial conditions,[54] it is not yet clear whether the intrinsic properties of 2D layered materials, such as tribology, chemistry, and electrochemistry, can be tuned by their interface with confined 2D ice. In particular, the mutual effect between 2D ice and 2D layered materials may prove to be the key in overcoming the surface instability of 2D layered materials, unlocking potential applications in the future.[16,47,57]

RECENT BREAKTHROUGHS IN THE INTERACTIONS BETWEEN 2D CONFINED ICE AND 2D MATERIALS

To explore the interplay between 2D ice and 2D layered materials, attention must be paid to the formation mechanism of 2D ice within the confinement space of 2D layered materials. Changes in the phase or thickness of confined 2D ice can be attributed to the spacing and vdW interaction between the layers of 2D materials. Conversely, the unique vdW interaction can also impact the behavior of confined 2D ice. Meanwhile, the 2D water–2D ice phase transition can either be discontinuous (first order) or continuous (second order) depending on the confined water density.[46,58] The density of molecules is an uncertain factor in the open system, as opposed to the finite number of molecules in simulation work. Therefore, intercalating vapor or liquid water into 2D layered materials may not necessarily result in the formation of 2D ice because the density of intercalating water molecules is difficult to control.[34,44,59,60] In addition to this, 2D layered materials exposed to humid or wet environments may also undergo oxidation,[19,57,61] hindering the true interplay between 2D ice and 2D layered materials, particularly when characterizing their

tribological properties. This is because oxidation or water absorption on the surface tends to increase the overall friction of 2D layered materials.[44,57,61]

Recently, our new study of Thi et al.[16] demonstrated the novel approach of 3D-to-2D ice transition under the confinement of 2D layered materials at cryogenic temperatures. This approach used in situ AFM to generate the 2D ice confined between the CVD-grown transition-metal dichalcogenides (TMDCs), MoS_2 in particular, and the rigid substrate, and investigate its lubricity enhanced by the presence of the confined 2D ice at the same time (Figure 2a).[16] First, bulky 3D ice was formed between two rigid substrates, one of which contained CVD-grown MoS_2 (Figure 2b).[16] After detaching from the growth substrate, MoS_2 remained steadily on the ice layer through hydrogen bonds at temperatures below −20°C (Figure 2c).[16] Finally, the bulky ice was slowly sublimed, and the part covered by MoS_2 was spontaneously converted into 2D ice (Figure 2d).[16] The experimental topographic image of 2D confined ice with a thickness of ~3.8 nm under 2D MoS_2 is shown in Figure 2e.[16] In comparison with previous studies, the advantages of this new approach for the generation of confined 2D ice under 2D layered materials can be summarized as follows:

1. Confined 2D ice was spontaneously formed by thinning the bulky 3D ice at cryogenic temperatures and did not undergo a liquid–solid phase transition. The 3D-to-2D solid phase transition facilitated the establishment and stabilization of the structure of confined 2D ice much more easily than the uncertain conversion from the liquid phase.
2. The experiment was conducted at cryogenic temperatures, and the water molecules were present in solid ice form, minimizing the oxidation or corrosion of 2D layered materials by humidity or water.
3. The surface of the 2D layered materials was freshly detached from the growth substrate at cryogenic temperatures, thereby preventing humidity absorption. Moreover, the intrinsic lubricity of the surface was guaranteed to be affected only by the underlying 2D confined ice.

By eliminating all extraneous factors, the pure interplay between 2D layered materials and confined 2D ice enhanced the lubricity of the surface of the 2D layered materials. This resulted in the confined 2D ice sliding away due to the motion of the AFM probe (Figure 3a).[16] Friction force microscopy (FFM) was then used to

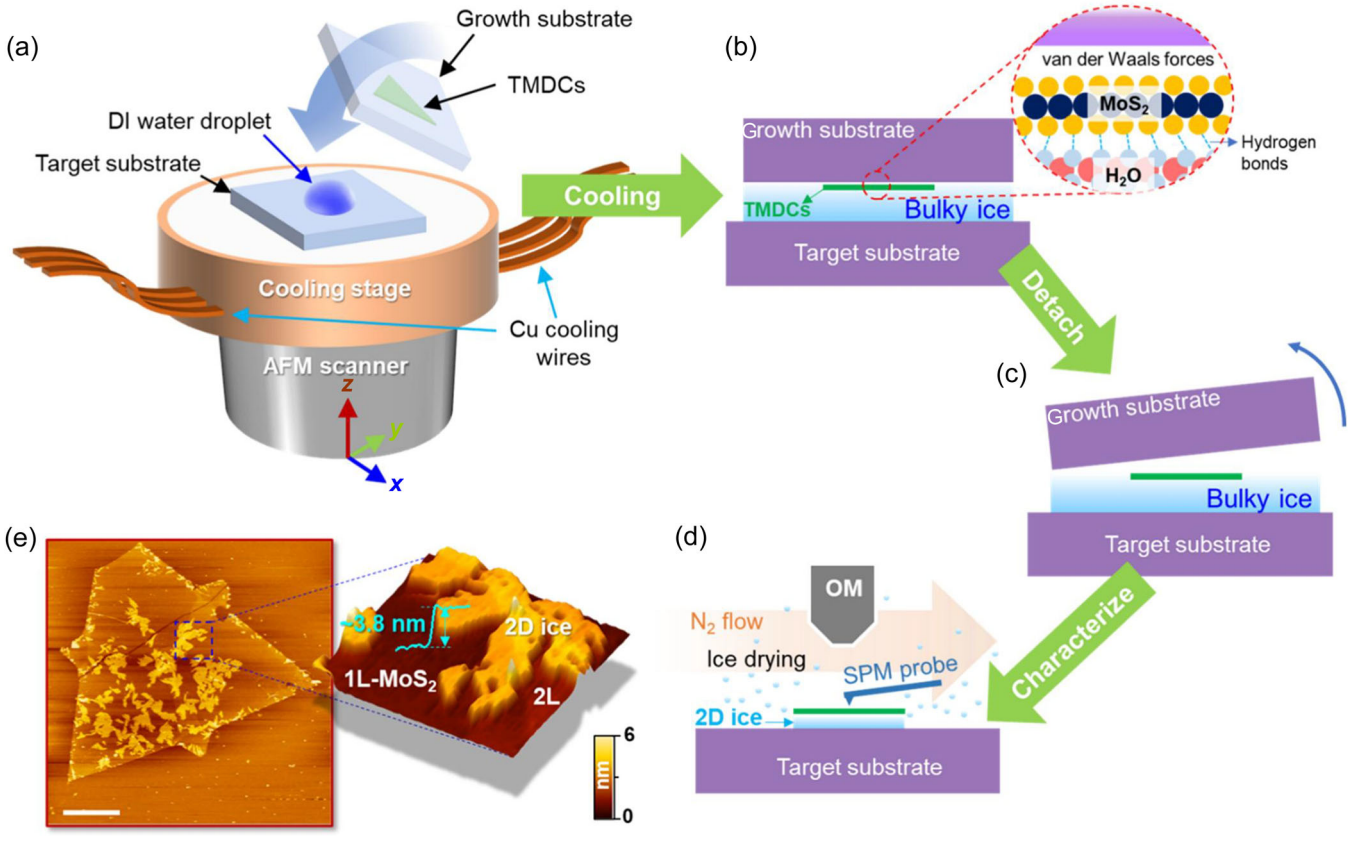

FIGURE 2 A new method to generate two-dimensional (2D) confined ice. (a) Schematic illustrating the experimental setup for the in situ AFM characterization of a 2D ice layer confined under the single layer (1L)-MoS_2. (b) The ice was formed between the growth substrate and the target substrate. (c) The growth substrate was detached and the MoS_2 remained on the ice surface. (d) The 1L-MoS_2/2D ice layer remained on the growth substrate for characterization. (e) Topographic images of the 2D ice layer under MoS_2 measured using the noncontact mode of AFM and the zoom-in topographic 3D image. Adapted with permission.[16] Copyright 2023, American Chemical Society. AFM, atomic force microscope; TMD, transition-metal dichalcogenide.

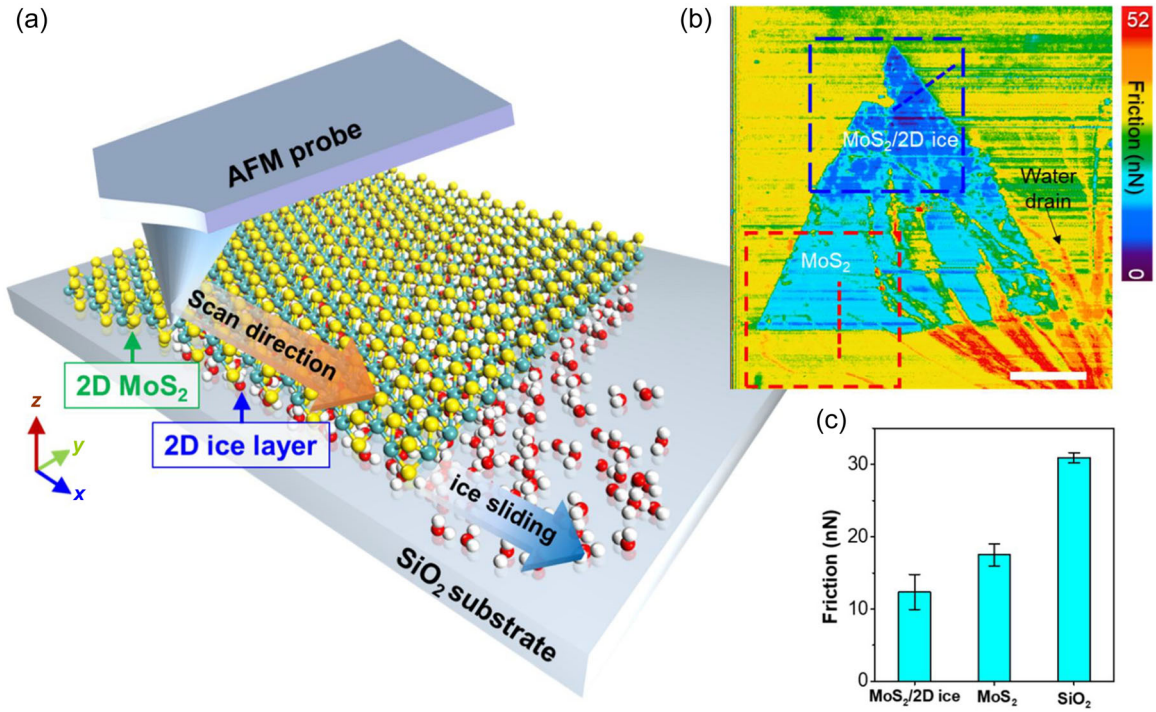

FIGURE 3 Reduced friction by the slippery two-dimensional (2D) ice. (a) Schematic illustrating the slippery 2D ice layer under MoS_2, pushed away by the AFM probe. (b) Friction force distribution on MoS_2 with the 2D ice layer being removed by the AFM probe. Scale bar = 5 μm. (c) Summarized friction force of the SiO_2 substrate and MoS_2 with/without the underlying slippery 2D ice layer. Adapted with permission.[16] Copyright 2023, American Chemical Society. AFM, atomic force microscope.

experimentally contrast the surface friction of MoS_2 with and without the underlying 2D ice (Figure 3b).[16] The friction of MoS_2 was reduced by approximately 30% due to the presence of 2D confined ice as compared to bare MoS_2 without the underlying 2D ice (Figure 3c).[16] This reduction was attributed to the weak binding at the interface between MoS2-2D ice and the 2D ice substrate. The weak binding was associated with the absence of hydrogen bonds on the surface of 2D ice, as predicted by the "interlocked" structure of hexagonal 2D ice. In addition, the atomically flat 2D ice acted as a spacing layer, effectively reducing the surface roughness and high friction induced by the underlying substrate on MoS_2.

Further, the coefficient of friction (CoF) of typical MoS_2 when paired with different materials/surfaces is summarized in Table 1. In general, the sputtered MoS_2 thin films and CVD-grown MoS_2 on SiO_2 substrate have comparable CoF = 0.22.[62] Meanwhile, the performance of the MoS_2 dry powder is highly unstable depending on the size distribution of the powder and the working environment.[67,68] In particular, the MoS_2 powder exposed to ambient conditions was easily oxidized to molybdenum oxide, which has high friction[61]; therefore, studies on MoS_2 powder often show high CoF.[63] Combining MoS_2 with other materials like carbon fiber[64] or SiO_2 nanoparticles[65] can help to passivate the surface of MoS_2 and increase the antiwear as well, with the trade-off of deceased intrinsic lubricity performance of MoS_2. The negative effect of oxidation and humid absorption on MoS_2 can be prevented by adding an oil-based solution. However, the oil itself already has a low CoF ~0.14[66]; MoS_2 merely acts as an additive

TABLE 1 Tribological performance of MoS_2 paired with different materials/solutions.

MoS_2-based products	Coefficient of friction
MoS_2 thin film[62]	0.22
MoS_2 powder lubricant[63]	0.8
MoS_2–carbon fiber[64]	0.44
MoS_2–SiO_2 nanoparticles[65]	0.55
MoS_2–diesel oil[66]	0.13
Bare MoS_2 on SiO_2[16]	0.22
MoS_2/2D ice[16]	0.18

that further enhances the lubricity of the oil-based lubricants but does not significantly contribute to its performance. In this sense, the new finding of the MoS_2/2D ice interface is the only way that lowers the CoF of bare MoS_2 till date and is able to operate effectively at cryogenic temperatures,[16] while oil-based lubricants are not able to work at such low temperature condition. The highly slippery and removable 2D confined ice can act as a sacrificial layer to increase the antiwear performance of this composition. Therefore, the interfaces between 2D ice and 2D layered materials should be the primary focus in the development of alternative lubrication concepts.

It is worth noting that the new method for generating 2D ice can be used to transfer 2D layered materials from the growth substrate to

a clean target substrate (as shown in Figure 2a–d) without requiring the use of polymers or chemicals, unlike other general transfer methods.[69,70] The sample prepared using this ice-assisted transfer was free of contaminants and perfectly preserved the intrinsic properties of the as-grown samples.[47] Nevertheless, some small wrinkles on 2D layered materials after transfer can be relaxed/eliminated by the self-flattening effect during the 3D-to-2D ice transition.[47] The successful detachment and transfer of 2D layered materials from a growth substrate using ice-assisted transfer rely on the fact that water in contact with and absorbed to the surface of 2D layered materials at the liquid (mobile) phase can maximize the hydrogen bonds with the surface. During conversion into the ice phase (immobile) at subzero temperatures, the high-density bonding was preserved and overcame the vdW bonding between the 2D layered materials and the growth substrate. This enabled the 2D layered materials to remain on the ice surface when peeled off from the growth substrate. Moreover, the icing process induced volume expansion, which can create interfacial strain, ultimately contributing to the delamination of 2D layered materials from the growth substrate.[47]

Temperature plays an important role in this new approach of coupling 2D ice with 2D layered materials. Ice-assisted transfer requires a temperature below −20°C to maintain the high-density hydrogen bonding at the interface so that it does not break down during the detachment of the growth substrate (Figure 4a). Typically, samples need to be cooled to below −30°C to ensure a high-yield transfer process. Further, during the conversion of 3D ice into 2D ice under 2D materials, the surface temperature must be maintained below −5°C to avoid the melting of the 3D ice, which could cause the 2D interface to collapse and prevent the generation of 2D ice. Using this novel method, ultra-clean and flat 2D layered materials were transferred, which showed superior performance compared to samples prepared using the common transfer method (see Figure 4b). On the other hand, detachment at temperatures above −10°C allows the ice to remove contaminants from the surface of 2D layered materials without causing any damage to the sample (Figure 4a).[47] This implies that the new technique can also be adapted to enhance the common transfer method. When adding a water droplet, the present contaminants on the surface of 2D layered materials were wrapped up by the

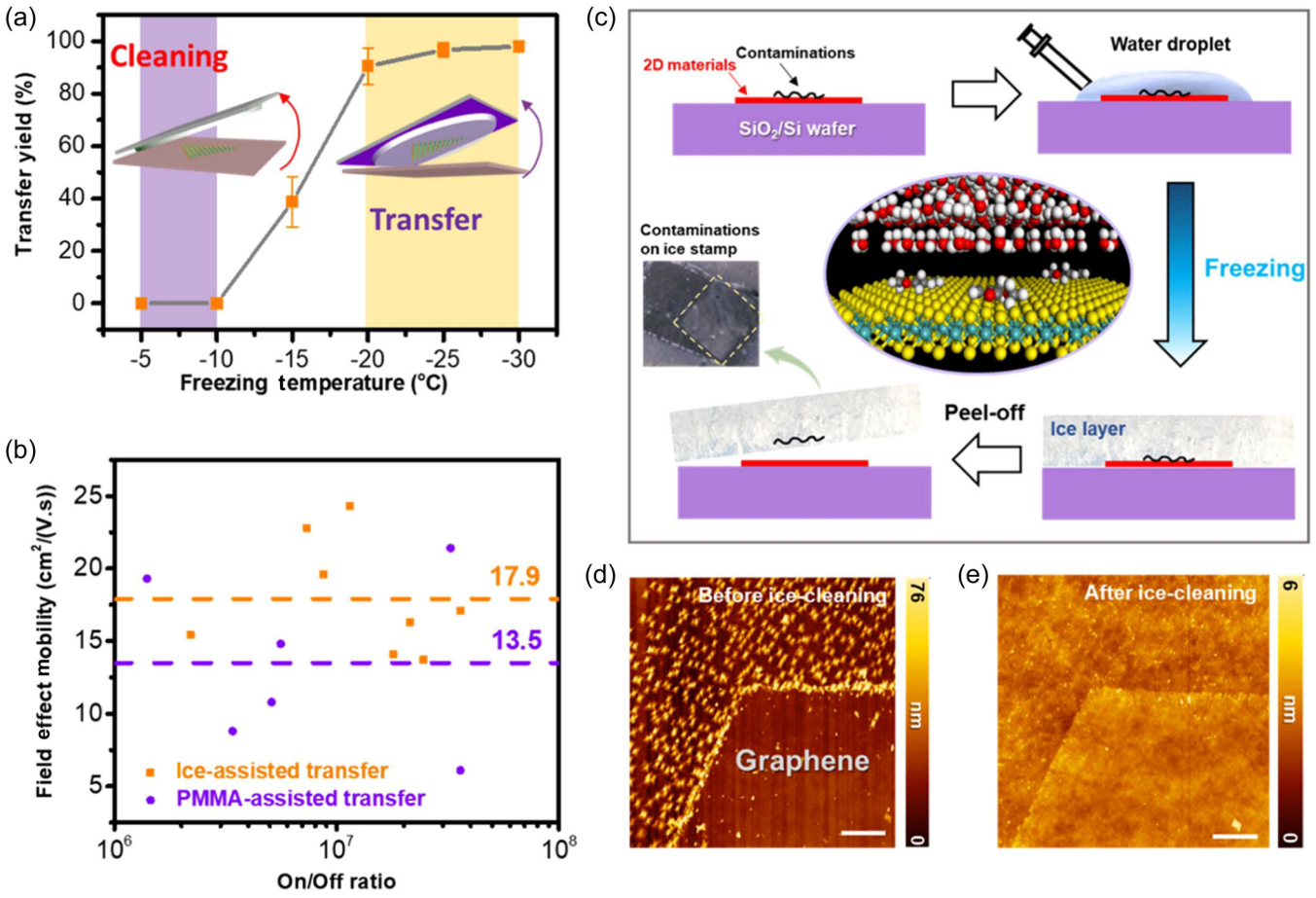

FIGURE 4 Ice-cleaning method. (a) The temperature dependence of transfer yield shows the available working temperature range of ice cleaning (from −5°C to −10°C) and the ice-assisted transfer method (below −20°C). (b) Comparison of field effect mobility of monolayer MoS_2 prepared by ice-assisted transfer and the PMM-assisted transfer method. (c) Schematic illustration of the ice-cleaning process. (d) AFM topographic images of graphene on a SiO_2 substrate prepared using the general polymer-assisted transfer method before and (e) after ice cleaning to remove the polymer residue. Adapted with permission.[47] Copyright 2023, John Wiley & Sons. AFM, atomic force microscope.

water, and then peeled off together with the ice film at cryogenic temperatures (Figure 4c).[47] The ice-cleaning process can also effectively remove the polymer residue on 2D layered materials transferred from general transfer methods or the lithographic process (Figure 4d,e).[47] This finding not only represents a high-efficiency approach to maintain the cleanliness of 2D materials during the transfer/fabrication process but is also appliable in cleaning any flat and sensitive surface, removing congestion in nanofluidic systems.

CHALLENGES AND FUTURE PERSPECTIVES

These new discoveries have also raised several challenges in investigating the structure and properties of confined 2D ice. The most notable one is that both 2D ice and 2D layered materials have ultrahigh surface instability. In particular, 2D ice is inherently unstable due to the lack of hydrogen bonding in the out-of-plane dimension, making it vulnerable to disturbance and rearrangement into different structures by applied disturbances or temperature fluctuations.[33] Another limitation arises from the characterization techniques. Many surface characterization techniques, such as scanning tunneling microscopy (STM) and AFM, are only applicable for the topmost surface layer and cannot directly reveal the stacking of multiple 2D ice layers or the atomic structure of inner layers.[32,34] Other techniques, such as X-ray scattering and neutron scattering, have limited spatial resolution and cannot precisely determine the in-plane structure of confined 2D ice.[71] Even with TEM, it is difficult to detect light atoms like hydrogen, and thus it cannot distinguish water molecules from other possible impurities.[52,72] Moreover, preparation of clean samples is also challenging. To study the intrinsic properties of the interface between 2D layered materials and confined 2D ice, clean samples with minimal contaminants are required. However, fabrication of atomically clean and well-defined surfaces in a controlled manner is challenging.

Despite the challenges that remain, the interfaces between 2D ice and 2D layered materials offer a promising avenue for the development of advanced technologies. This provides a new strategy for studying heterostructures, where the phase of inner layers can be modulated. For example, sandwiching 2D ice layers between two identical or different 2D materials can create tuneable interfaces with hybrid properties by controlling the phase of confined 2D ice via external conditions. Notably, the sliding friction between incommensurate and commensurate surfaces can differ by ~100 times.[73] Thus, combining phase control of 2D ice with different isotropic/anisotropic 2D layered materials can create an integrated nanomechanical system in which the medium between two atomic layers can be modulated. The motion of the intersystem can be either fast or immobile, depending on the interface between the 2D ice and the sandwich layers.

The friction reduction by 2D ice also represents a new breakthrough in lubricity technologies. The 2D ice can be developed as sustainable lubricants working at low temperatures and high-humidity environments, which is a critical solution for outdoor traffic and machineries in very cold winter seasons in many countries.[74] Instead of consuming energy or chemicals to remove it, the naturally existing ice cover can be utilized to lubricate movable parts. Additionally, due to its mobility in confined spaces, 2D ice can serve as a sacrificial layer that reduces wear during operation. The best part is that these sacrificial layers are pollution-free, and can be collected and recycled with ease.

CONCLUSION

In conclusion, the current understanding of the interface between 2D ice and 2D layered materials is just the tip of the iceberg. A wide range of mutual influences at interfaces, such as optical, ferromagnetic, electronic, and superconductive effects, remain unexplored. The unveiled ultrahigh lubricity at the interface of 2D ice/2D layered materials, combined with the novel phase transition approach for producing ultrathin 2D ice, holds great promise for the future development of solid-state lubricants, nanomechanics, and nanofluidic systems that can work at low temperatures and high-humidity conditions. Additionally, 2D ice can be used as a supporting layer for ultrahigh-quality and exceptionally clean transferred 2D flakes and continuous 2D films. Moreover, theoretical frameworks are continually advancing to complement experimental work and investigate the interface under critical conditions that are currently inaccessible by measurement technologies. The future applications for 2D ice and 2D layered materials are ever-expanding, and the real application begins with fundamental studies of their interface behaviors. Although significant technical and theoretical challenges still exist, sustained efforts are necessary to develop new experimental techniques, improve sample preparation methods, and develop advanced theoretical models to gain a better understanding of the interface behaviors between 2D layered materials and confined 2D ice.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflict of interest.

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