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Binmin Wu,[1,2] Ziyu Zhang,[1] Chao Wang,[1] Enming Song,[3] Jizhai Cui,[1,4] Gaoshan Huang,[1,4] Peng Zhou,[2] Zengfeng Di,[5] and Yongfeng Mei[1,3,4,a)]

AFFILIATIONS

[1]Department of Materials Science, International Institute of Intelligent Nanorobots and Nanosystems, State Key Laboratory of ASIC and Systems, Fudan University, Shanghai 200433, People's Republic of China
[2]School of Microelectronics, Fudan University, Shanghai 200433, People's Republic of China
[3]Shanghai Frontiers Science Research Base of Intelligent Optoelectronics and Perception, Institute of Optoelectronics, Fudan University, Shanghai 200433, People's Republic of China
[4]Yiwu Research Institute of Fudan University, Yiwu, Zhejiang 322000, People's Republic of China
[5]State Key Laboratory of Functional Materials for Informatics, Shanghai Institute of Microsystem and Information Technology, Chinese Academy of Sciences, Shanghai 200050, People's Republic of China

[a)]Author to whom correspondence should be addressed: Email: yfm@fudan.edu.cn

ABSTRACT

Due to their unique structures and properties, emerging two-dimensional (2D) materials have been at the frontier of research in, e.g., materials science, physics, and engineering. Three-dimensional (3D) tubular geometry enables 2D materials unparalleled advantages for various applications, for example, wide-angle infrared photodetectors, extremely sensitive molecular sensors, and memory with high density. Furthermore, 3D tubular structures offer a promising integration platform into chips with a broad range of materials, especially 2D materials. In this Perspective, we highlight state-of-the-art methods to assemble/manufacture 2D materials into 3D tubular structures/devices via self-rolled-up or template methods. These tubular 3D devices inspire unique physical, chemical, and mechanical properties for optical microcavity, photodetector, on-chip electronics, and bubble-propelled microengines. On-chip manufacture of 3D tubular structures/devices provides great opportunity and challenge for 2D materials for More than Moore applications such as unconventional electronics, smart sensors, and miniaturized robots.

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I. INTRODUCTION

The promotion of nanomembrane materials makes manufactured structures more miniaturized and inexpensive, and corresponding device components have high performance and reliable.[1,2] The assembly of nanomembranes into 3D tubular structures through a micro/nanofabrication process offers advanced design for the L-C filter network,[3] infrared photodetectors,[4–6] optical microresonator,[7–9] energy storage device,[10–12] and micro/nano-engine,[13–15] which significantly accelerates the functional development of integrated circuit chips in applications for More than Moore. In general, the vast majority of conventional inorganic/organic nanomembrane materials fabricated by membrane deposition, crystal epitaxy, or thinning processes are suitable for 3D structural assembly, including but not limited to metallic conductors (e.g., Cr, Fe),[16,17] semiconductors (e.g., Si, Ge),[5,18] and insulators (e.g., ZrO_2, Si_3N_4).[6,19] For example, silicon bulk wafers, the cornerstone of advanced semiconductor chip fabrication, can be thinned to provide the flexural rigidity required for the fabrication of 3D assembled structures.[20] Researchers have developed a series of fabrication methods for the challenges raised by the fabrication of 3D micro/nano-devices such as membrane rolling, folding, buckling, and 3D printing.[21–25] The research on membrane materials and 3D assembly methods is still in the stage of rapid development, and the technological maturity is difficult to meet the needs of commercial product manufacturing.

Recently, as emerging nanomembrane materials, graphene-like layered 2D materials are constructed into 3D structures such as nanoscrolls,[26,27] origami,[28,29] and kirigami,[30,31] arousing interest in the exploration of tubular devices based on 2D materials. Graphene,[32] composed of a single layer of carbon atoms, exhibits excellent electrical, optical, thermal, and mechanical properties. The discovery of graphene's unique properties motivates researchers to investigate materials with similar structures and outstanding properties. 2D materials have received increasing attention, and they consist of stacks of independent ultrathin

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monolayers with atomically flat surfaces and no dangling bonds, which are naturally excellent nanomembrane materials with unique physical and chemical properties. Compared with traditional bulk materials, 2D materials have ultra-high carrier mobility, excellent bending flexibility, and optical transparency. Hence, 2D materials are revolutionary materials for both academia and industry in the 21st century. In recent years, the combination of nanomembrane rolled-up technology and 2D materials has attracted great attention, and various 3D tubular devices with different functions were fabricated and demonstrated, such as field-effect transistors (FETs),[33,34] photodetectors,[27] and molecular sensors.[17]

Here, we first introduce three promising methods for the assembly of tubular structures, which are compatible with the structural properties and growth processes of 2D materials. Then, we detail several state-of-the-art 3D tubular devices produced based on 2D materials. We review the physical properties of 2D materials and the effects of strain and stacking effects in 3D tubular structures. Finally, we provide an outlook on this rapidly evolving field and the challenges of transition from laboratory demonstrations of individual elements to large-scale production of integrated circuits for future applications.

II. ASSEMBLY METHODS FOR TUBULAR STRUCTURES OF 2D MATERIALS

The assembly of 2D materials into 3D structures has become a rapidly developing research hotspot.[25,35] Based on the inheritance of previous research results and the unique structure of 2D materials, a series of fabrication technologies for tubular 2D materials have been developed.[6,26,27,34,36–41] Three advanced assembly methods are discussed in Secs. II A–C, as shown in Figs. 1(a)–1(c). These three

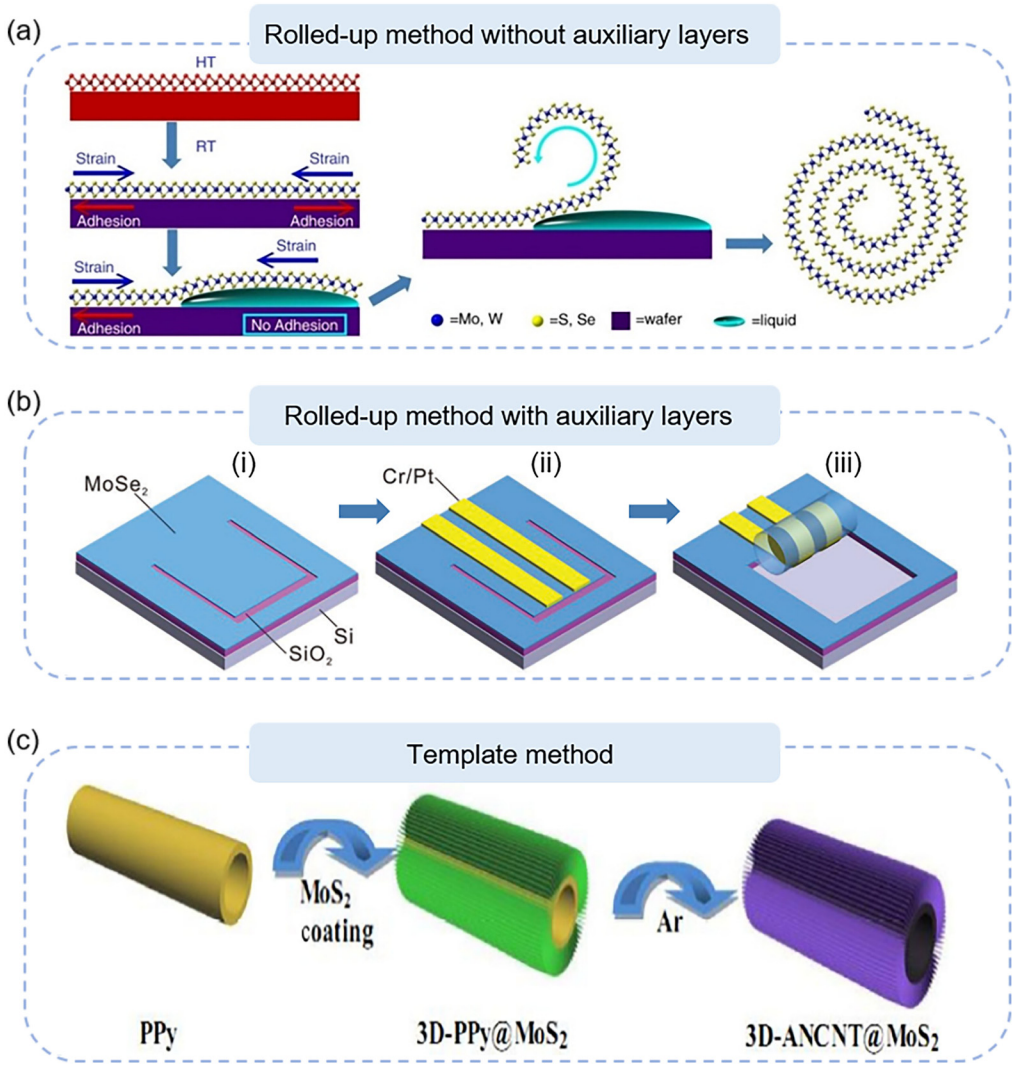

FIG. 1. State-of-the-art assembly methods for tubular 2D materials. (a) Rolled-up process of TMDs' nanoscrolls with a drop of ethanol. (b) Rolled-up $MoSe_2$ nanomembranes with auxiliary layers. (c) Scheme for the synthetic processes of the tubular MoS_2 composite by the template method. Panel (a) is adapted from Nat. Commun. **9**, 1301 (2017).[26] Copyright 2017 Springer Nature. Panel (b) is adapted from Small **15**, 1902528 (2019).[27] Copyright 2019 Wiley-VCH. Panel (c) is adapted from Nano Res. **11**, 3603 (2018).[11] Copyright 2018 Tsinghua University Press and Springer-Verlag GmbH Germany.

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methods have their own advantages and disadvantages, and they can highlight their respective advantages in different fields and face challenges. Tubular devices are still a long way from practical application.

A. Rolled-up method without auxiliary layers

Compared with traditional materials, the most notable features of 2D layered materials are their strong in-plane bonds and weak van der Waals (vdW) interlayer coupling.[42] The key to the rolled-up method without an auxiliary layer is to overcome the vdW force between the 2D material with internal strain and substrate or intermediate layers. General methods to form freestanding nanomembrane tubes are ion intercalation,[39] defect control,[43] surface treatment,[38] etc. Viculis *et al.*[39] prepared graphene tubes with a chemical method. The compound KC_8 is first inserted into the graphite in an inert gas atmosphere. After the chemical reaction between KC_8 and ethanol, the solvation of potassium ions, and the release of hydrogen, the graphite is dispersed into monolayer or few-layer graphene nanosheets. Finally, the graphene tubes are fabricated after the subsequent sonication process. Although this method can produce graphene tubes with high yield, the graphene tubes are suffered from the difficult separation process from the residual solvent. Meng *et al.*[38] selectively and partially removed the top sulfur atoms of monolayer MoS_2 by weak argon plasma and thereby created tension within the MoS_2 basal plane. After the plasma treatment, the MoS_2 nanomembrane was peeled off from the substrate and rolled-up. This low-cost, solvent-free rolled-up technology is also applicable to other 2D materials, but there are some shortcomings as it will inevitably damage the crystalline structure of 2D materials and introduce numerous interfacial defects.

In order to preserve the structural and electrical integrity of the 2D materials, a rolled-up method without chemical and physical damage is urgently needed. Typically, 2D materials are synthesized on substrates at high temperatures. While the samples are cooled down to room temperature, strain is introduced into the 2D materials due to different thermal expansion coefficients between the substrate and the as-synthesized materials. MoS_2 is released from the substrate when the solution intercalates between MoS_2 and the substrate. Free-standing MoS_2 self-rolls into tubular structures under internal strain. Whether the substrate is 3D bulk (such as SiO_2 and sapphire) or 2D layered (such as mica), the 2D materials are always adhered to the substrate by weak vdW force.[42] Cui *et al.*[26] introduced a method to fabricate transition-metal dichalcogenides (TMDs) tubes by intercalating ethanol between 2D materials and substrates, as shown in Fig. 1(a). This strategy can be used to create high-order superlattices consisting of monolayer, bilayer, and multilayer 2D materials. A high-order SnS_2/WSe_2 vdW superlattice was created by rolling up SnS_2/WSe_2 heterojunctions.[36] The structure and dimensions of an electronic band are tuned by the construction of these superlattices, and the bandgap is dramatically narrowed to greatly increase the charge transport in rolled-up vdW superlattices. The rolled-up 2D materials perfectly inherit the crystalline structures of the original 2D materials with improved electrical and optical properties.[26,36] Moreover, the strategy can also be applied to the fabrication of mixed-dimensional high-order superlattice devices involving other dimensional materials such as 0D quantum dots, 1D nanowires, and 3D nanomembranes, providing superior material platforms for next-generation fundamental research and engineering applications.

B. Rolled-up method with auxiliary layers

Before monolayer graphene was discovered, inspired by ancient origami techniques, researchers have been working on establishing a sophisticated process for assembling nanomembranes into 3D micro/nanostructures[1,25,35,44,45] such as tubes, wrinkles, and microchannels. The preparation of self-rolled nanomembrane tubes requires a large vertical strain gradient within the nanomembranes, which can be introduced by lattice mismatch, altering parameters during nanomembrane deposition or introducing a strain layer.[19,46–49] Furthermore, the diameters of the tubular structure devices can be adjusted according to the thickness, strain gradient, and material selection of the nanomembranes.[15,17] This method has achieved a large amount of self-rolled microtubules from the nanomembrane of various materials. For example, Wang *et al.*[4] fabricated a GaAs/AlGaAs quantum well-embedded self-rolled microtube for mid-wave infrared photodetectors. These tubular photodetectors exhibit enhanced photoresponsivity and wide detection angle without the assistance of external optical coupling structures. Nanomembranes with 3D structures combine the excellent properties of traditional materials with tubular geometries, overcome some shortcomings in planar configuration, and have been widely applied in the research of electronics,[5,50] electromagnetic waves,[8] robotics,[51] and energy storage devices.[10]

It is worth noting that the rolled-up technique is not only practicable for traditional nanomembranes but also is useful for both monolayer and multilayer 2D materials. Similarly, for 3D tubular structures assembly of 2D materials, the key of rolled-up method with an auxiliary layer is to control the gradient and the distribution of strain of the 2D material.[27,33] Because neither the growth nor the transfer process of 2D materials involves lattice mismatch,[42,52] which introduces controllable strain. The rolled-up method with auxiliary layers is currently the most popular method for the assembly of 2D materials. In addition, as mentioned in Sec. II A, the strain introduced into 2D materials by the high-temperature growth process only rolls up tubes with nanoscale diameters, making it difficult to roll up microtubes with precisely controlled diameters.[36] SiN_x layers grown by CVD under different plasma frequencies[34] and metallic chromium layers deposited by electron beam evaporation[17] can be the most commonly used strain layers. An auxiliary layer that completely covers or supports the entire 2D material protects the integrity of the 2D material,[53] which can greatly improve the yield and uniformity of tubular devices.[34] However, there are also disadvantages that the introduction of auxiliary layers inevitably destroys the crystal structure and physical and chemical properties of 2D materials.[54] Using sparse strips as auxiliary layers enables 2D materials to be free-standing on the substrate in a completely bare form,[17,27] which provides an excellent experimental platform for studying the intrinsic properties of 2D materials. As shown in Fig. 1(b), the 2D $MoSe_2$ materials and substrates are separated by the dry etching or wet etching process to prepare free-standing nanomembrane tubes.

Rolling 2D materials into microtubes by introducing strain layers is a well-established process that is inherited from traditional techniques for nanomembranes. Combining with the CMOS-compatible micro-nanofabrication technique, the constructed 2D material tubes have great potential for applications in FETs,[6] photodetectors, optical microcavities,[24] and actuators.[55] Tubular components based on 2D materials can greatly advance the development of future integrated chips.

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C. Template method

As a method to directly fabricate 3D tubular structures of 2D materials, the template method[11,56–58] usually starts with the preparation of columnar molds as the substrate for the growth of 2D materials. Monolayer or multilayer 2D materials are then grown on selected templates by common material growth methods such as hydrothermal method,[11] chemical vapor deposition (CVD),[59] and atomic layer deposition (ALD),[37] as shown in Fig. 1(c). Recent reports show that molds ranging from hundreds of micrometers[12] to 10 nm[37] in diameter are suitable for producing 2D material tubes, which can be used for FETs, energy storage devices, etc. The other significant advantage of this method is that the diameter and orientation of the tubes can be tuned by careful design of the template, for example, to obtain a curved tube with uniform diameter. Under the guidance of the template method, a multi-functional system with integrated micro-channels can be prepared,[12] which is expected to be used for drug delivery, nanofluidic channels, etc.

III. STATE-OF-THE-ART TUBULAR DEVICES BASED ON 2D MATERIALS

A. Tubular electronics

The development of semiconductor technologies has become strict requirements on the size and power consumption of devices. The traditional planar semiconductor devices are limited by Moore's law, which makes them difficult to meet the needs of current and future development of integrated circuits. The 3D tubular structures provide an opportunity for planar 2D materials devices to keep up with the development trend of Moore's law. Hou *et al.*[53] constructed tubular 3D resistive random access memory (RRAM) by rolling up a planar Cr/h-BN/Ti sandwich structure, as shown in Fig. 2(a). Unlike

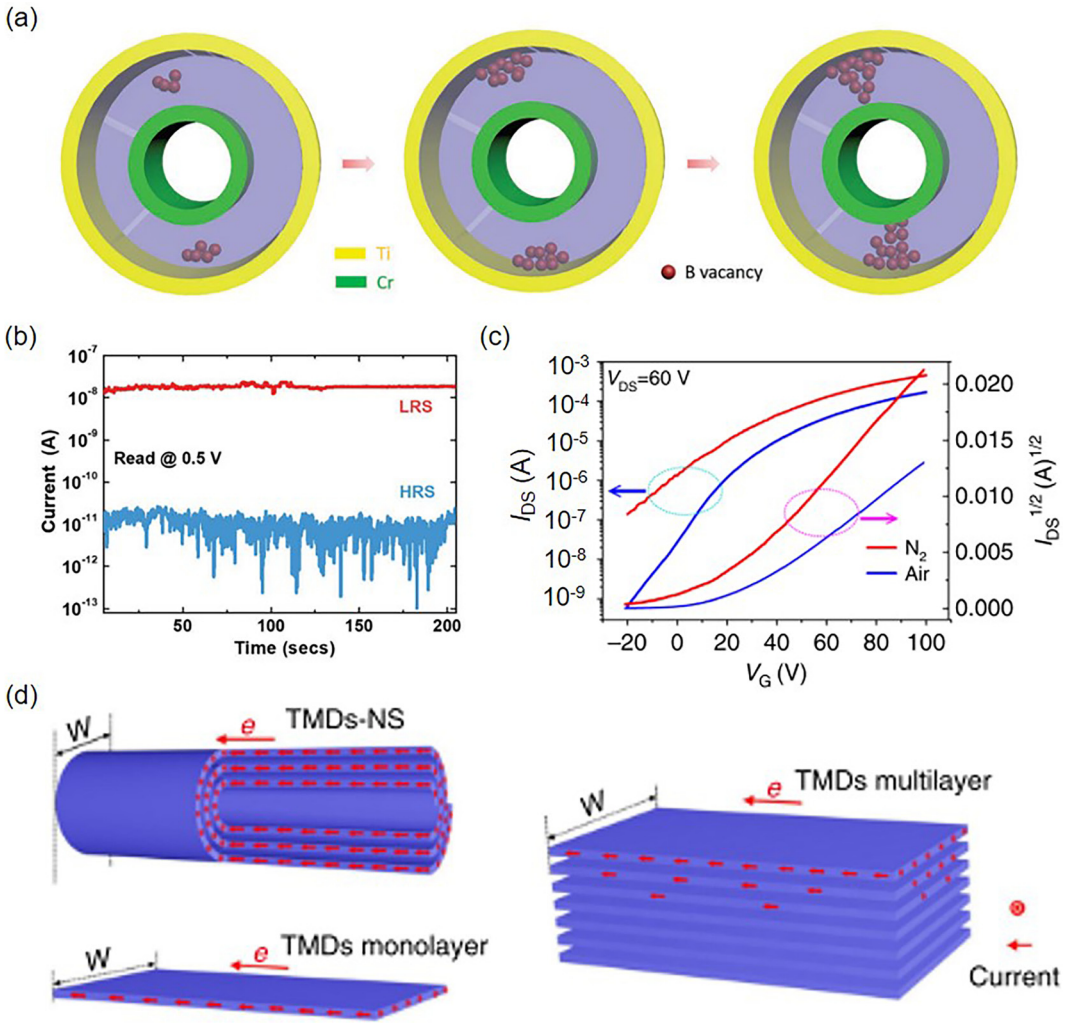

FIG. 2. 2D materials electronic devices with tubular geometry. (a) The formation process of the conductive filament in the tubular RRAM. (b) Retention property of nonvolatile tubular RRAM. (c) Transfer characteristics of a tubular MoS_2 FET tested in N_2 (red) and air (blue). (d) Schematic representations of current conduction in tubular, monolayer, and multilayer MoS_2 under bias. Panels (a)-(b) are adapted from Small **15**, 1803876 (2019).[53] Copyright 2019 Wiley-VCH. Panels (c)-(d) are adapted from Nat. Commun. **9**, 1301 (2017).[26] Copyright 2017 Springer Nature.

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the planar devices, the electric field distribution between the titanium and chromium electrodes in the coiled tubular devices is not uniform, resulting in relatively few B vacancy migrations and the formation of conductive filaments. Therefore, the rolled-up tubular RRAM exhibits higher set voltage and lower programming current, which has important implications for RRAM applications. As shown in Fig. 3(b), the tubular RRAM demonstrates a nonvolatile ON/OFF current ratio up to 10^3. In addition, the footprint of tubular RRAM is reduced by six times, and there is room for further improvement by increasing the number of turns. This tubular memory architecture lays the foundation for higher data storage density and low power consumption devices.

Ultrathin 2D materials with extremely high carrier mobility are considered to have the potential to replace silicon.[60,61] However, experimental values reported by various laboratories are far below the theoretical value yet. While inheriting the excellent properties of the original materials, the tubular structures also acquire unique electrical and optoelectrical properties that are different from existing material systems.[26,36] Unlike carbon nanotubes, the tubular structures are open and cannot be uniquely determined by the chiral vector.[62,63] The 2D material tubes rolled-up by nondestructive techniques are dense, impurity-free, and highly crystalline. Fig. 2(c) shows the transfer characteristic of a self-rolled MoS_2-based FET prepared by Cui *et al.*[26] with an on-off ratio over 10^5 and an electron mobility of 200–700 cm V^{-1} s^{-1}. Based on their Archimedes spiral structures, the entire 2D material nanomembranes in the tubes can participate in the transportation of carriers, as shown in Fig. 2(d). Compared with planar 2D material devices, the carrier mobility in rolled-up tubular devices is enhanced by more than an order of magnitude. In addition, the unique self-encapsulating geometry enables the 2D material tubes to exhibit higher optical and electrical stability.

B. Tubular photodetectors

Compared with conventional materials used for photodetection, such as silicon, germanium, and mercury cadmium telluride, 2D materials possess the characteristics of rich bandgap, high carrier mobility, room temperature operability, and low cost.[64–66] Typically, graphene is a semiconductor material with zero bandgap, which is expected to break the limitation of wavelength in conventional semiconductor optoelectronics. However, the ultra-thin thickness results in low light absorption efficiency and, thus, leads to lower quantum efficiency. Although conventional design of silicon waveguides,[67] quantum dots,[68] and microcavities[69] combined with two-dimensional materials are able to improve the absorption efficiency, at the same time, unfavorable factors, such as strong wavelength selectivity and complex fabrication processes, will be introduced inevitably.

Tubular designs are also applied to the enhancement of photon absorption and the broadening of the response wavelength. Zhou *et al.*[27] deposited chromium (Cr, 20 nm)/platinum (Pt, 5 nm) as a

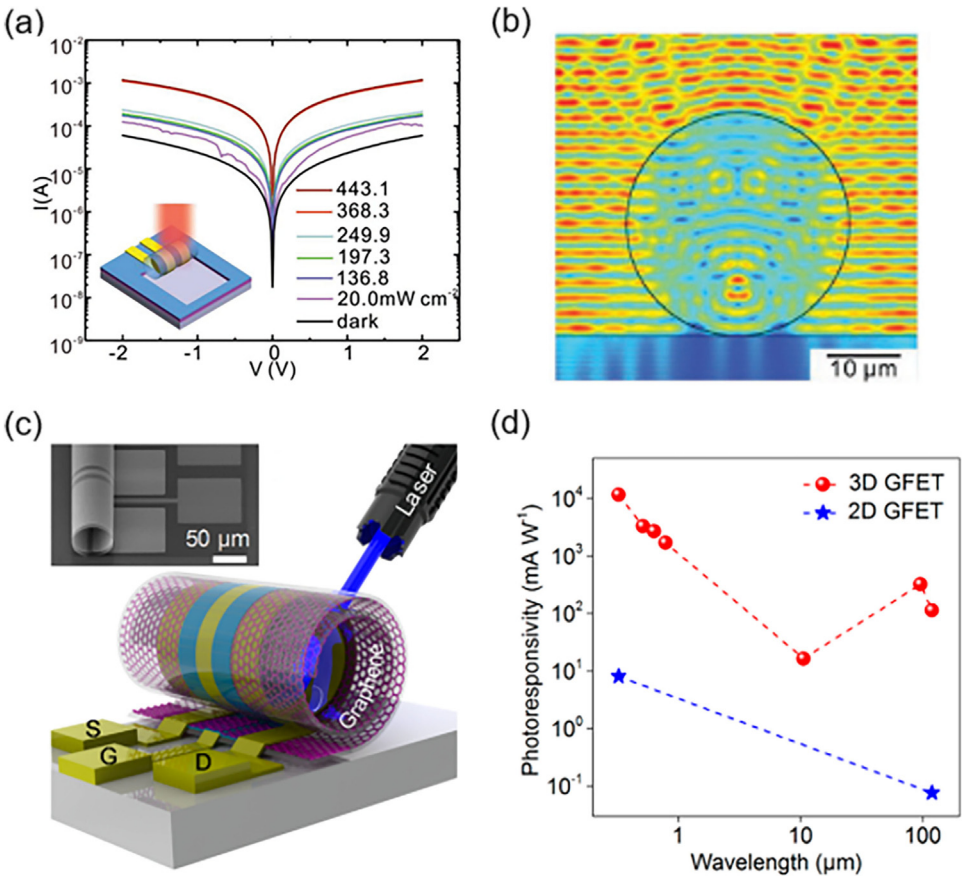

FIG. 3. 2D material photodetectors with tubular structures. (a) I-V characteristics of a tubular $MoSe_2$ device in the dark and under laser illumination. (b) Simulated electric field distribution at the illumination wavelength of 808 nm in a 3D tubular $MoSe_2$ device. (c) 3D tubular monolayer graphene phototransistors. (d) The measured photoresponsivity of the 3D GFETs at wavelengths of 325 nm to 96 μm. Panels (a)-(b) are adapted with permission from Small **15**, 1902528 (2019).[27] Copyright 2019 Wiley-VCH. Panels (c)-(d) are adapted from Nano Lett. **19**, 1494 (2019).[6] Copyright 2019 American Chemical Society.

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strain layer on $MoSe_2$ to fabricate tubular photodetectors, as shown in Fig. 3(a). Under the illumination condition, the vertical incident light is reflected multiple times by the wall of a tubular structure. Moreover, the electromagnetic field inside the microtubules is locally enhanced, as shown in Fig. 3(b), and the light is partly trapped in the microtubes, increasing the light absorption of the photodetectors. In this way, the poor photoresponsivity of a planar 2D materials photodetector deriving from its extremely thin thickness can be greatly compensated by the tubular structures. In addition, tubular structures also bring higher surface state density and modify the bandgap modification, resulting in increased photosensitivity of the tubular $MoSe_2$ photodetectors while reducing the dark current. Similarly, Deng *et al.*[6] fabricated a monolayer graphene FET with a 3D tubular structure, as shown in Fig. 3(c). With the assistance of the self-rolled structure, a 3D graphene FET exhibits the capability of room-temperature photodetection among the ultraviolet, visible, mid-infrared, and terahertz regions with bandwidths over 1 MHz, as shown in Fig. 3(d). 2D material optoelectronic devices with tubular geometry are perfectly capable to achieve the goal of high photoresponsivity, wide spectral range, and outstanding operating speed.

C. Optical microcavities

As discussed in Sec. 3.2, electromagnetic waves can be confined by successive internal reflections in microtubules, leading to an intensified local electric field in the tubes.[4,5] Enhanced photon–phonon interaction in the tubular structures results in strong surface-enhanced Raman scattering, providing a powerful nondestructive and ultrasensitive spectroscopic characterization technique.

Wang *et al.*[24] fabricated graphene-encapsulated oxide microtubules by a nanomembrane rolled-up technique, as shown in Fig. 4(a). Compared with graphene on planar germanium, the Raman signal intensity of the tubular graphene is enhanced drastically, which is caused by the strong confinement of an electromagnetic field around the 3D tubular structure, as shown in Fig. 4(b). In addition, the residual uniaxial strain in graphene introduced by the self-rolled process results in an in-plane anisotropic feature in the graphene lattice. Fig. 4(c) shows the polarization-dependent properties of the observed Raman scattering intensity due to the anisotropic absorption of graphene, which matches the simulations well. Compared with oxide-supported graphene microtubes, double-sided bare graphene microtubes have higher performance in surface-enhanced Raman scattering.

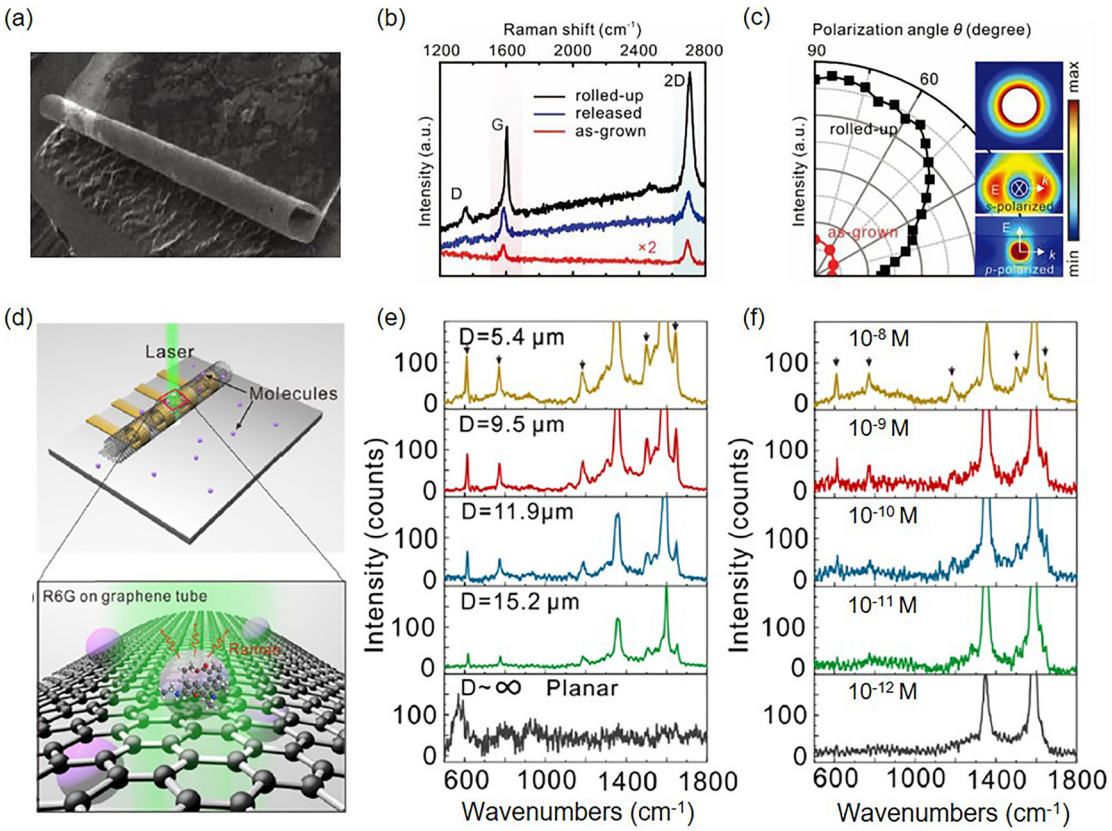

FIG. 4. Tubular 2D material optical microcavity. (a) SEM image of the rolled-up graphene oxide microtube on the Ge wafer. (b) Raman spectra of as-grown, released, and rolled-up graphene under an excitation laser wavelength of 514 nm. (c) Polarization-dependent characteristics of the graphene microtubes. (d) Schematic illustration of the Raman experiment for probing R6G molecules. (e) Raman spectra of R6G (8×10^{-9} M) on graphene microtubes with different diameters. (f) Raman spectra of R6G on graphene microtubes (~9.5 μm) with different R6G concentrations. Panels (a)-(c) are adapted from Small **15**, 1805477 (2019).[24] Copyright 2019 Wiley-VCH. Panels (d)-(f) are adapted from ACS Appl. Mater. Interfaces **19**, 1494 (2021).[17] Copyright 2021 American Chemical Society.

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As shown in Fig. 4(d), Ma *et al.*[17] proposed a monolayer graphene tubular device for ultrasensitive molecular sensing. The tubular geometry of the device further enhances the surface-to-volume ratio of the graphene devices, resulting in a significant increase in Raman intensities, and this greatly improves the sensitivity of molecular concentration detection. Fig. 4(e) shows the Raman spectra of R6G at a concentration of 8×10^{-9} M on graphene microtubes with different diameters. A smaller radius means that more layers of graphene with adsorbed R6G are probed, resulting in obvious enhancement of Raman scattering. Finally, an unprecedented low concentration of R6G molecules of 10^{-11} M was detected by a 9.5 μm diameter graphene tube, as shown in Fig. 4(f).

D. Bubble-propelled micromotors

In 2009, a bubble-driven microrocket is reported by utilizing rolled-up technology,[51] laying the groundwork for catalytic tubular micromachines, in which the bubble is an important factor to drive the movement of micromotors in liquid. However, the micromotors are easy to be absorbed by the bubbles in the liquid, as it is difficult to overcome the huge surface tension of the bubbles. Zhang *et al.*[55] proposed a tubular catalytic micromotor with single-layer graphene as the outermost layer [Fig. 5(a)]. Ti/Cr was sequentially grown on graphene as a strain layer. 2 nm Pt was then deposited on the top, acting as a catalyst for the decomposition of hydrogen peroxide to power the micromotor. The graphene/Ti/Cr/Pt nanomembrane was released and rolled-up under the control of built-in strain. In the tubular structure, graphene wrapped the outermost layer of the micromotor, which is characterized by Raman spectra as shown in Fig. 5(b). The problem of bubble absorbent is then solved due to the graphene hydrophobicity and the reduced interaction between the micromotors and air bubbles in the surrounding liquid. In Fig. 5(c), the micromotors without graphene coating adsorb numerous bubbles after passing through the bubbles, and the speed drops significantly. At the same time, the graphene-covered micromotors maintain their speed without adsorbing bubbles after passing through the bubble group. Sensing devices or actuators fabricated by combining graphene with other materials are continuously reported.[70–74] For example, Gracias *et al.*[75] introduced a solvent-driven self-folding method using graphene-polymer bilayers, laying the foundation for the creation of functionally critical 3D self-folding sensors and stimuli-responsive actuators. Currently, 2D materials have been found to be toxic to certain viruses and tissue cells and have also been reported to promote cell differentiation. As for biological applications such as surface or *in vivo* detection and microbial culture, it is necessary to further identify the biocompatibility and biodegradability of 2D materials to reveal the possible health risks of 2D materials.

IV. OUTLOOK

High-performance 3D tubular devices based on 2D materials can overcome the bottleneck of planar chip technology limited by Moore's

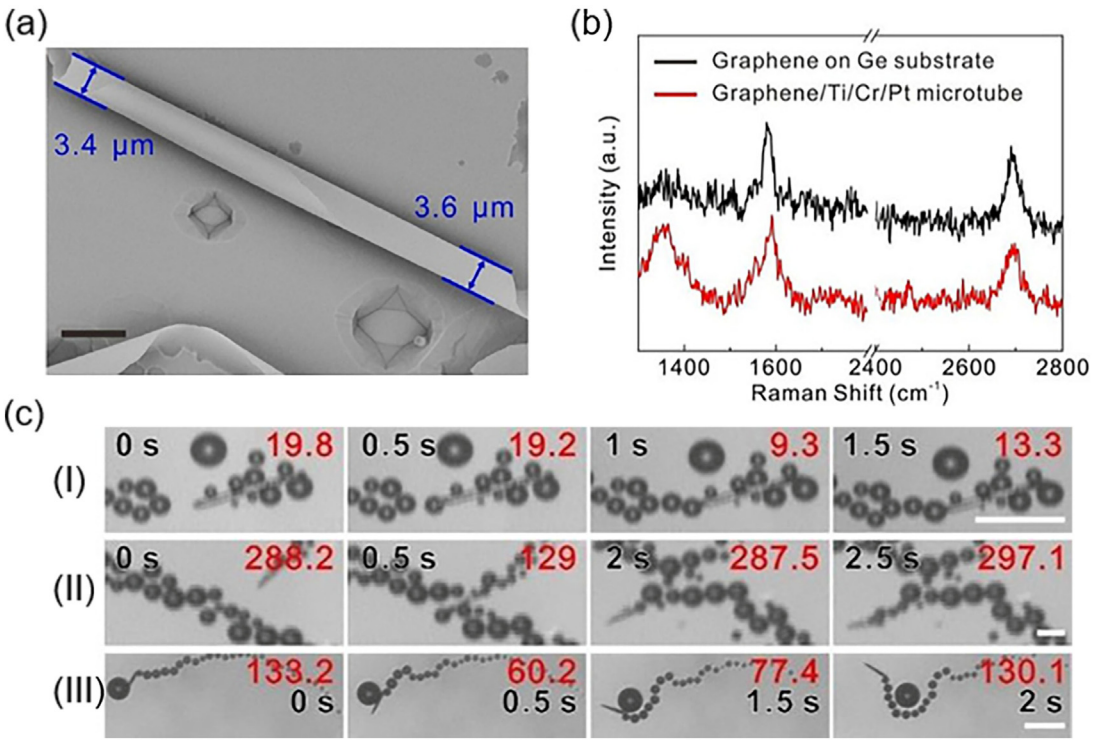

FIG. 5. Rolled-up monolayer graphene tubular microengines. (a) SEM image of a rolled-up tubular micromotor. (b) Raman spectra of graphene on the Ge substrate and the rolled-up graphene/Ti/Cr/Pt microtube. (c)Time lapse images of the micromotor without graphene covered moving with bubbles attached (I), the graphene-covered micromotor moving through a string of bubbles without bubbles attached to its surface (II), and the graphene-covered micromotor encountering a large bubble and bypassing it (III). Panels (a)-(c) are adapted from Chem. Asian J. **14**, 2479 (2019).[55] Copyright 2019 Wiley-VCH.

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Law,[76,77] providing a solution for future high-performance and multifunctional integrated chips. However, not only a tubular device based on 2D materials is a brand emerging field of research but also the study of 2D materials alone still faces a series of challenges.[78] Most of the research works about the integration of 2D materials with 3D tubular structures today are still in the proof-of-concept stage, and there are still huge challenges toward practical applications. As shown in Fig.6, we discuss the obstacles in fabrication and promising solutions for 2D material-based tubular devices at the material, device, and system levels.

As for the material level, although 2D materials can be easily produced by mechanical exfoliation, the mass production of large-area and misorientation-free 2D materials is still very challenging.[79–81] The quality and integrity of 2D materials greatly depend on the lattice-matched substrates and the strong interaction between the epitaxial layer and the substrate. Several works have demonstrated that high-quality 2D materials can be grown by CVD techniques on specially treated substrates[82–84] such as graphene and h-BN on copper (111) foils.[85,86] However, the high-temperature environment during the CVD process, pulsed laser deposition, and molecular beam epitaxy is not suitable for a silicon-process compatible substrate yet. Furthermore, wafer-level transfer technology of 2D materials also faces challenges. The transfer of 2D materials from the epitaxial substrate to the working substrate can cause wrinkles, cracks, and residual contaminants that are unfavorable to the devices. Aside from that, surface contamination during the material synthesis and device fabrication process, which affects the intrinsic properties and device performance of 2D materials, has long been an unsolved key problem.

At the device level, it mainly includes three parts: fabrication of planar devices, assembly of 3D tubular devices, and post-packaging. After more than ten years of development, great progress has been made in planar devices based on 2D materials.[60,67,76] There have already been many excellent articles discussing the upcoming challenges of planar 2D materials in the next generation, such as efficient doping methods,[87] Ohmic contacts,[54,88–90] and vdW integration,[42,91] so we will not discuss them here. Even with outstanding planar devices, advanced assembly and encapsulation processes are indispensable for device fabrication. The assembly of the 3D architectures is the most complex and vital step in the fabrication of tubular devices based on 2D materials, and there are numerous challenges that need to be solved. The characteristics and applications of the 2D materials' tubular devices are summarized in Table I. In addition to the three methods specifically discussed in this Perspective, other methods, such as chemical intercalation and plasma/microwave spark assistance, are also enumerated therein. Currently, the one of the most effective methods for assembling 3D structures is the nanomembrane rolled-up technique. There are a lot of variables that affect the assembly of 3D structures such as nanomembrane morphology, strain gradient distribution, deposition temperature, and various external disturbances.[2,92,93] Additionally, the introduction of controllable strain gradient and distributions into 2D nanomembranes and precisely tunable tube diameters still face great challenges due to the process complexity. For now, introducing a well-designed strain layer with a corresponding sacrificial layer is a wise choice to meet the demand. Nevertheless, the commonly used physical vapor deposition methods for nanomembranes (such as electron beam evaporation and magnetron sputtering) will cause certain damage to the surface of 2D materials,[54] which is fatal to semiconductor devices. Furthermore, once the nanomembrane is rolled up, the stability of the device will be impaired, whether the current passivation technology can solve this problem still needs more experimental verification in the future. For the time being, it is also uncertain if the rolled-up ultrathin 2D material can satisfy the need of a reasonably thick tube wall for the practical implementation of the optical cavity. Furthermore, during the 3D assembly process, slight

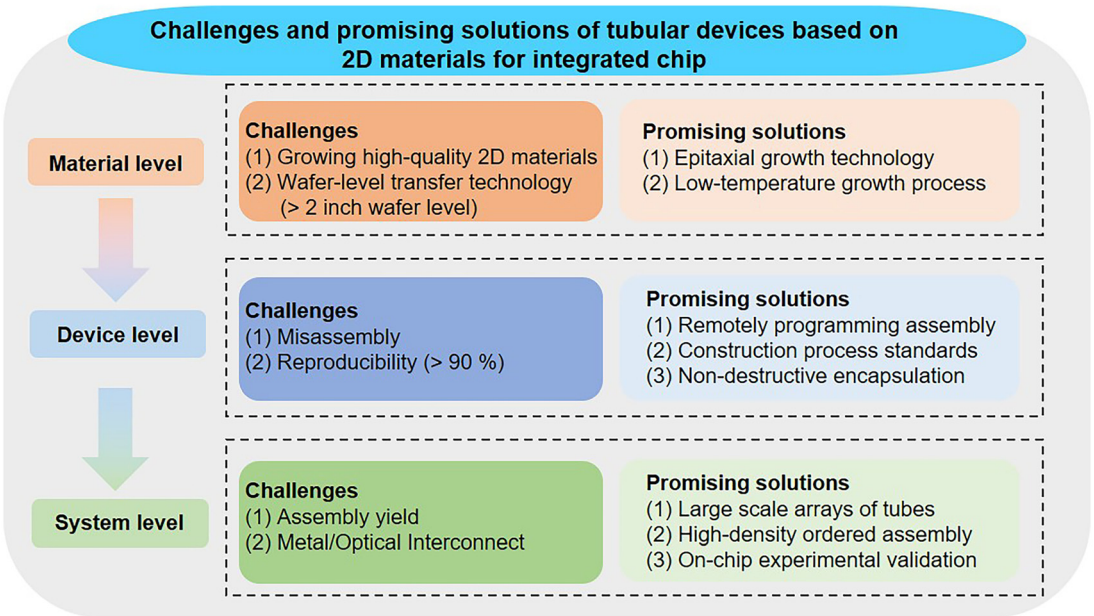

FIG. 6. Challenges and promising solutions of tubular devices based on 2D materials for an integrated chip.

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TABLE I. Characteristics and applications of tubular structures based on 2D materials.

Assembly method	Material	Tube diameter	Application	Reference
Rolled-up method with auxiliary layers	Graphene	60 μm	Phototransistor	6
		60 μm	Field-effect transistor	34
		5–14 μm	Molecular sensor	17
		~3.6 μm	Micromotor	55
	$MoSe_2$	~6.2 μm	Photodetector	27
	MoS_2	~62 μm	Photodetector	40
	h-BN	~10 μm	Memory	53
Rolled-up method without auxiliary layers	MoS_2	~20 nm	Field-effect transistor	26
	SnS_2/WSe_2	~20 nm	High-order superlattice	36
Template method	Graphene	40–150 μm	Micromotor	12
		80 μm	Graphene-copper wire	59
	MoS_2	~1 μm	Lithium storage	57
		216.6 nm	Ion batteries	11
		10–200 nm	Field-effect transistor	37
Chemical route	Graphene	~40 nm	Nonoscroll	39
Plasma assistance	MoS_2	5.673 nm	Nonoscroll	38
Microwave spark assistance	Graphene	15–150 nm	Field-effect transistor	41

external disturbances can cause nanomembranes to assemble in unexpected shapes such as bubbles generated by wet etching and flow fields in a solution.[94] All of these will cause the tearing of the nanomembrane, deviation of the curling path, and poor tightness of the tube wall, which ultimately lead to poor stability and repeatability of these devices. In addition, similar to the rolling process, the folding of 2D materials can also regulate corresponding physical properties for devices such as changes in the energy band structure[95–97] and electrical[98,99] and optical properties[100] of materials. Precise control of the angle and the number of layers[101] provides a feasible platform for exploring physical mechanisms such as magic-angle graphene[102,103] and moiré patterns.[104–106]

After assembly, the collapse of the 3D tubular structure occurs occasionally because of environmental disturbances. Moreover, 2D materials also lack stability in atmospheric environments. For example, oxygen and water vapor, which can oxidize the surface of black phosphorus, significantly reduce device performance and lifetime.[107–109] Therefore, it is necessary to use a passivation layer for encapsulation. The most commonly used 3D encapsulation method is to utilize ALD to deposit dense oxides to cover the surfaces of the devices.[8] Compared with the previous 3D devices (such as MEMS), the complexity of the tubular structure based on 2D materials in the manufacturing process is greatly reduced. For example, the fabrication of the rolling-up tubular structure usually only requires several processes of coating and etching. In addition, the fabrication of tubular devices is compatible with conventional silicon chip fabrication processes. Therefore, tubular devices are low-cost method to fabricate 3D devices.

At the system level, miniaturization and high integration of components contribute to the outstanding performance and high area efficiency of chips. Devices based on 2D materials allow components to be integrated on a variety of substrates, which will potentially break the previously unachievable limitations of single-chip integration. However, compared with planar devices, tubular devices are more likely to be affected by factors, such as complex fabrication processes and structural instability, resulting in low yields of high-quality devices. On the other hand, nanomembrane tubes are distributed on the chip in an array because of the area occupied for planar patterns before release. This results in low area efficiency for devices with large arrays such as infrared focal plane array detectors. Furthermore, although high-performance devices have been verified in the laboratories, their practical applications on chips have not yet been realized. More attention should be paid to the interconnection of the tubular device on the chip during the fabrication process.

In conclusion, unique 3D tubular devices based on 2D materials show potential in electronics, optics, and many other fields defining a rich material platform for fundamental research and exploring a path for the development of next-generation integrated chips with 2D materials. Despite the excellent achievements, there is still a series of challenges in material fabrication, device process, and system integration which require joint efforts in the coming decades.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

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Author Contributions

Binmin Wu: Writing – original draft (equal); Writing – review and editing (equal). **Ziyu Zhang:** Writing – review and editing (equal). **Chao Wang:** Writing – review and editing (equal). **Enming Song:** Writing – review and editing (equal). **Jizhai Cui:** Writing – review and editing (equal). **GaoShan Huang:** Writing – review and editing (equal). **Peng Zhou:** Writing – review and editing (equal). **Zengfeng Di:** Writing – review and editing (equal). **Yongfeng Mei:** Supervision (equal); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review and editing (equal).

DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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