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Review Article

REVIEW ARTICLE

Toward 2D materials for flexible electronics: opportunities and outlook

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ABSTRACT

Two-dimensional nanomaterials exhibit exceptional multifunctional properties including high-electron mobilities/saturation velocities, high surface to volume ratios, unique layered structures and mechanical compliance, positioning the class of materials to be influential in next-generation flexible electronics for applications in wearables and the Internet of things. In this perspective, three key areas of interest are identified that take advantage of the multifunctional nature of these materials including molecular sensing, van der Waals transfer and compliant radio frequency electronics. Significantly more progress needs to be made to realize commercialization of these materials, but the revolutionary accessible properties may reveal themselves in these three key areas of future flexible electronic systems.

Key Words: 2D materials; flexible electronics; sensors; nanomaterials; Van der Waals transfer; conformal radio frequency electronics.

INTRODUCTION

From graphene to transition metal dichalcogenides, black phosphorous and MXenes, the unequivocally astounding properties of two-dimensional (2D) materials have peaked the interest of physicists, materials scientists and engineers alike. This excitement within the past several decades has inspired revolutionary technological advancements in batteries, electronics, quantum sciences, sensors, composite materials and much more [1, 2]. Of the potential application areas where these 2D materials may play a substantial role is in flexible and conformal electronics. It is in the multifunctional properties of 2D materials that make them so attractive for this technology area, as 2D materials possess exceptional electronic/optoelectronic performance metrics, easily tunable properties through surface interactions, extremely high optical constants, all while demonstrating the ability to accommodate strain up to and exceeding 10% [3, 4]. The combinatorial need for bendable, flexible, stretchable electronic materials excludes many electronic materials that are traditionally very brittle and prone to issues with mechanical reliability. In contrast, organic electronic materials promise very low modulus and mechanical flexibility, but many times do not exhibit electronic properties that are necessary for functional devices. The key attributes of 2D materials may be the answer in this ever-expanding technology space requiring electronic/optical performance and mechanical flexibility [3].

In this short perspective, three specific areas incorporating both applications and unique processes enabled by 2D materials for future flexible electronics are discussed, namely in 1) highly sensitive molecular sensors, 2) van der Waals transfer strategies and (3) conformal radio frequency electronics (Fig. 1 [5, 6]). Sensors based on this intriguing class of materials demonstrate near-record-low sensitivities with facile integration into complex, flexible form factors with relative ease. Additionally, as a

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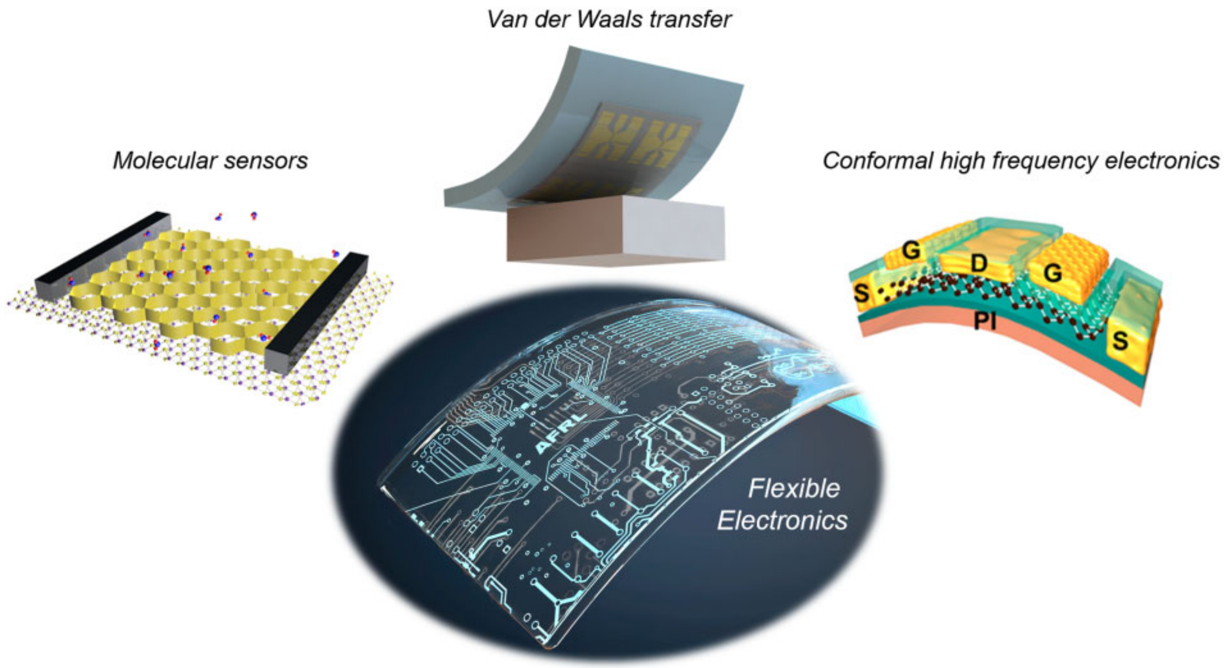

Figure 1: Applications of 2D materials in flexible electronics including functionalized molecular MoS_2 sensors, h-BN as a van der Waals transfer material for GaN electronics and MoS_2 high-frequency transistors on plastic substrates. Figures reproduced with permission from Ref. [5]. Copyright (2020) American Chemical Society, Ref [6] Copyright (2016) American Chemical Society.

mechanical 'lift-off' layer, 2D materials offer a simple low impact transfer and integration method for high-performance devices. Finally, taking advantage of the clear electronic mobility advantage over silicon coupled with the flexibility rivaling many organic materials, conformal and flexible high-frequency devices using 2D materials as a semiconducting channel may revolutionize the communication space in wearables and Internet of things applications.

MOLECULAR SENSORS

The rapid development of electronics for wearables and Internet of things have pushed forward the need for sensors to detect chemical exposure, physiological conditions and other environmental factors. Sensors demonstrating an electrical conductance response to interactions with ambient molecules were first presented in the form of field-effect transistors five decades ago [7], where the devices were gated via doping by the adsorbed molecules. The original 3D embodiments of the active sensing layers lacked the sensitivity for detection below useful limits of biomarkers, toxic vapors, or pathogens in the vapor or liquid phase. Interest in electronic sensing by molecular gating was revived with the advent of 1D materials [8], especially silicon nanowires [9] and carbon nanotubes [10, 11]. While 1D materials demonstrate the extremely high sensitivity due to their high surface-to-volume ratios [12], scalable production still remains a challenge today. While the limits of electronic sensing were expanded via evolution from 3D to 1D materials, reports on the novel properties and extremely high surface area per gram of 2630 m^2/g (Fig. 2a [17, 18, 19, 20, 21, 15]) of 2D graphene captivated scientists across disciplines and motivated diverse efforts in sensor development [19]. In sensor platforms, graphene and other 2D materials also provide the mechanical strength and flexibility at the ultimate materials scaling limit [20], unique transport characteristics, tunable optical properties, controllable surface sites and the potential for facile device fabrication in contrast to their 1D carbon counterparts (Fig. 2b) [18]. While the majority of flexible 2D sensor works to date focus on graphene, other van der Waals materials including 2D transition metal dichalcogenides are currently of interest due to the large response accompanied by molecular adsorption events [21].

A number of reports stand out for demonstrations of the utility and performance of graphene in wearable sensing devices (Fig. 2c–e). A graphene-based sensor for detection of oral bacteria attaching directly to the tooth is an example of flexibility, biocompatibility and compact packaging [15]. In addition to oral health, such sensors interfacing with the human body could be used to detect oral bacteria linked to Alzheimer's disease [22] and monitor for other pathogens including viruses. Inkjet printing of graphene/boron nitride heterostructures on textiles [23] show functionality after multiple cycles in a washing machine, demonstrating potential for everyday use. In addition to molecular sensing, temporary graphene-based tattoo sensors monitoring of skin temperature [17], skin hydration, electrocardiograms (ECGs), electromyograms (EMGs) and electroencephalograms (EEGs) have been demonstrated. As our abilities to integrate other 2D materials into flexible platforms is expanded, it will be exciting to see enhancements in sensitivity and other sensor metrics as new 2D materials with all the useful properties of graphene plus benefits specific to the desired applications are integrated into flexible platforms.

While sensors based on 2D materials continue to push the limits of sensitivity and detection limits, advancing selectivity is equally important and perhaps more challenging. For effective sensing, the analyte of interest must be distinguished between other species which may have an even stronger affinity for reactions at the sensor surface, or counteract the effect of the target analyte. Intrinsic 2D material-based sensors generally enable distinction between donor/acceptor molecules [24], but identification of specific molecules requires surface

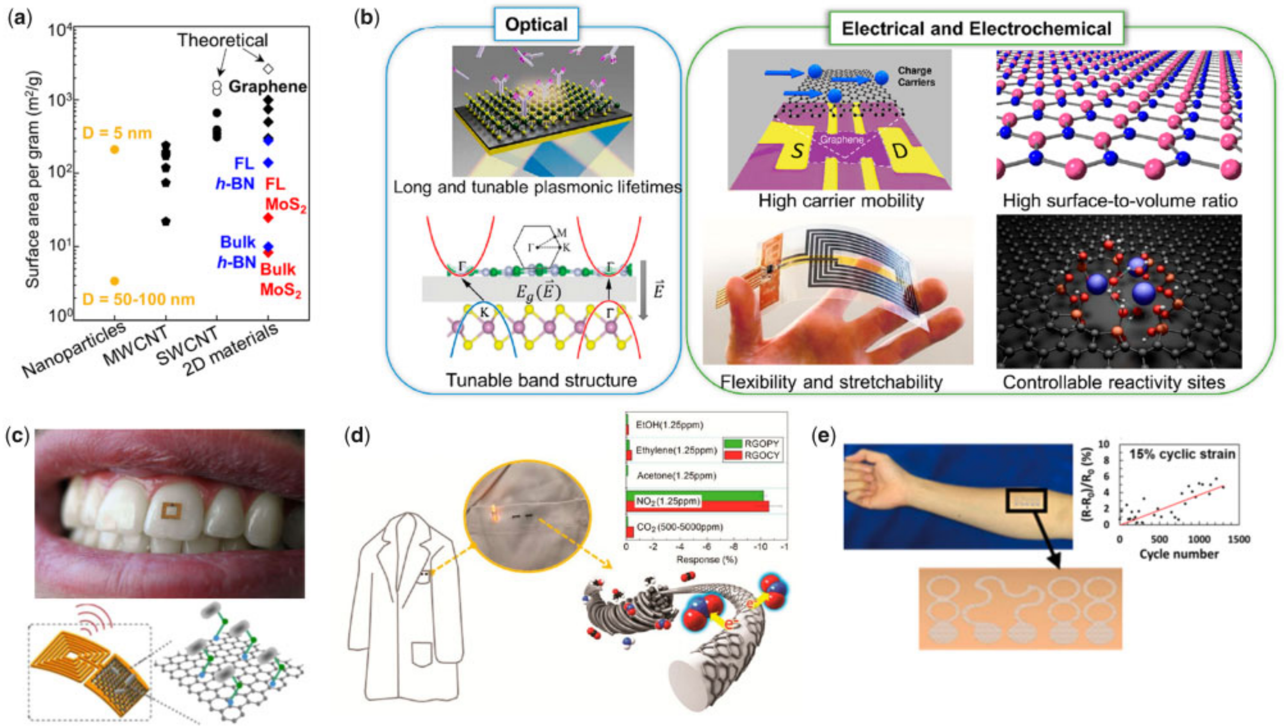

Figure 2: Advantages of 2D materials in sensor applications. (**a**) Surface area per gram comparison with other nanomaterials and (**b**) optical and electrical characteristics highly responsive to subtle changes in the ambient environment. Embodiments of 2D materials-based conformal sensors for (**c**) detection of pathogens [13–15], (**d**) concentrations of airborne substances [16] and (**e**) human performance [17] demonstrate the multifunctional nature of 2D materials in these applications. Figures reproduced from with permission from (a, b) Ref [18] Copyright 2019 American Chemical Society, (c) ref [15] Copyright 2018 Elsevier Ltd. and Ref [14] Copyright 2016 Elsevier B.V. (d) Ref [16] Copyright 2017 American Chemical Society (e) Ref [17] Copyright 2017 American Chemical Society.

functionalization. For example, selective sensing of ammonia in a gas phase mixture was achieved by coating graphene with Co(tpfpp)ClO_4 [25]. Targeting of dimethyl methylphosphonate was possible through graphene sensors functionalized with triphenylene, where the strong hydrogen bonding between the analyte and N-substituted triphenylene increases the binding affinity of the sensor and strengthens the chemresistor response [26]. Additionally, functionalization of graphene with biomolecules enables targeted detection of specific cancer cells [27] and other biological agents. While functionalization schemes are not as well established for 2D TMDs, the unique electronic interplay between substrate, metal and chalcogen layers enables some flexibility for attachments of antibodies, peptides, or other molecules for specific binding necessary for selective detection at the desired sensitivity limits [28, 29].

VAN DER WAALS TRANSFER

A key application for 2D materials in flexible electronics is their ability to facilitate lift-off and transfer of pristine materials requiring high temperatures and/or epitaxial growth methods to a flexible substrate possessing high degrees of imperfection such as polymers. This is possible by taking advantage of the weak vdW bonding between layers enabling mechanical exfoliation of atomically thin and smooth layers. Traditional transfer of graphene and 2D materials using etching strategies can involve harmful chemicals and be very wasteful industrial processes, with some recent studies resorting to electrolytic delamination as a viable alternative [30, 31]. VdW transfer of thin crystalline 2D and 3D membranes for flexible electronics (for general details on vdW integration see ref. [32]) is an exciting approach enabling minimal waste, requires no chemical exposure and encourages reusability of the substrate material. The advantages of membranes over more conventional flexible films (amorphous, Si, organics) has led to various lift-off methods – laser, epitaxial, mechanical and vdW (see ref. [33] for a comparison). vdW lift-off offers a simple low impact process that produces strain-free layers with ultra-smooth surfaces. Beyond processing advantages, a wide range of materials can be epitaxially grown on the vdW surface [34–36].

The lack of dangling surface bonds and atomically thin nature of 2D materials allows for two special cases of epitaxy-vdW and remote epitaxy. In vdW epitaxy the layer registers via vdW forces to the 2D buffer relaxing substrate lattice requirements [37]. Using a graphene buffer (0001) GaN has been grown on amorphous SiO_2 [38] and (111) oriented GaAs on Si (100) [39]. 2D buffers have been shown to produce quality layers comparable to those on conventional non-native substrates [40–44]. Remote epitaxy takes advantage of wetting transparence through atomically thin graphene [33]. Here the growing layer is affected by the substrate below and can be used to grow a wide variety of ionic materials, Fig. 3a [34, 35, 47, 41]. Both cases provide the weak vdW interface necessary for mechanical exfoliation with tapes [5, 43], stamps [41] or Ni spalling [46] and wet etch methods [47].

One of the first examples of vdW transfer of 3D layers was by Chung et al. who grew GaN layers and LEDs on exfoliated graphene showing the possibility of the method [48]. Subsequently, more scalable methods using CVD graphene on metals have been successfully used for growth and transfer of 3D membranes [38, 49], however, metals present issues with process compatibility/contamination. A more suitable option is

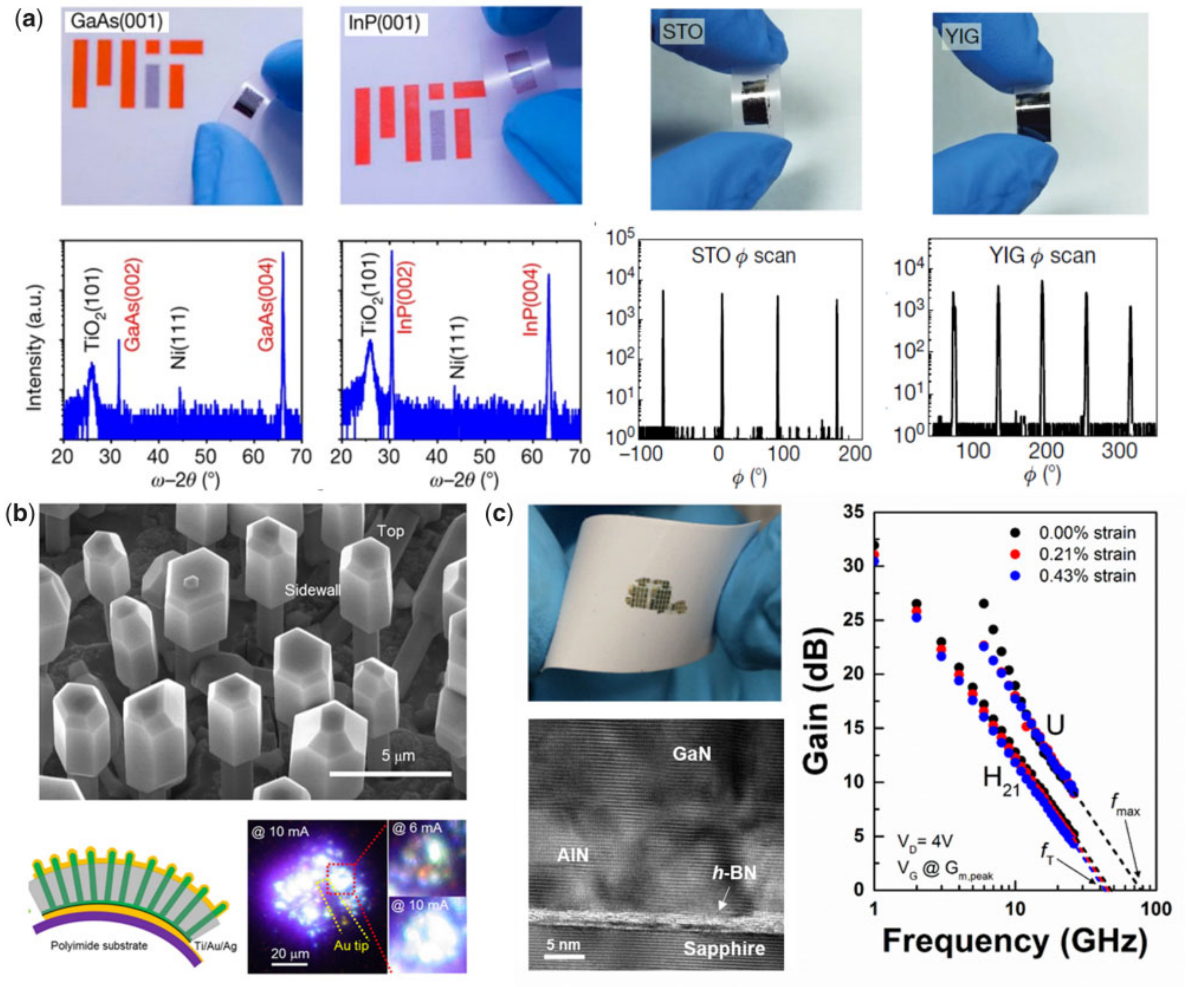

Figure 3: Van der Waals transfer of membranes and devices. (a) A variety of compound semiconductor and oxides grown by remote epitaxy and transferred to flexible substrates [34, 35]. (b) Flexible microrod InGaN/GaN MQW LEDs, demonstrating versatility of vdW transfer method to non-planar structures [45]. (c) Flexible high-performance AlGaN/GaN HEMTs operating at RF under stain up to 0.43% [41]. Figure reproduced from (a) Ref. [34], Copyright 2017 Springer Nature Ltd, Ref. [35], Copyright 2020 Springer Nature Ltd. (b) Ref. [45] under the terms of the CC-BY 3.0 License copyright 2014, AIP Publishing LLC (c) Ref. [41] Copyright 2017 Wyle-VCH.

epitaxial graphene on SiC or sapphire that can be produced uniformly over large areas. Kim *et al*. grew multiple quantum well (MQW) LEDs on epitaxial graphene then transferred to plastic substrates [46]. III-As, III-P and oxide membranes grown by remote epitaxy transferred to flexible substrates demonstrate the diversity of possible materials [34, 36]. Beyond planar films, 1D wires can be grown on 2D buffers for transfer. InGaN/GaN MQW microrods were grown on SiO_2/Si using a CVD graphene buffer then transferred to a polymer producing reliable flexible LED arrays (Fig. 3b) [45]. The layered hBN presents a viable option as a 2D buffer for III-nitrides, as it can be grown epitaxial over large areas by MOCVD [50, 51]. Using an hBN layer, Kobayshi *et al.* demonstrated improved LED intensity after vdW transfer due to reduced strain [44]. The hBN buffers have also enabled transfer of large-area films and devices; Ayari *et al.* transferred 2″ InGaN/GaN MQW structure using a metal tape [43]. Selective lift-off of small patterned areas can be achieved through selective growth of the hBN buffer [52]. Beyond LEDs, flexible high-electron-mobility transistor (HEMTs), solar cells and sensors have been produced [5, 35, 41, 53]. Flexible GaN HEMTs on hBN demonstrated the ability to transfer delicate high performance devices with F_t/F_{max} >47/74 GHz strained up to 0.43%, Fig. 3c [41]. The versatility of this method allows transfer of individual devices to full-wafers of materials from compound semiconductors to complex oxides proving membranes and complex devices for integration into flexible electronics.

CONFORMAL HIGH-FREQUENCY ELECTRONICS

It is unlikely that 2D materials will replace silicon in conventional logic applications with continual reduction in the cost of necessary elements and existing infrastructure. The true impact of 2D materials in nanoelectronic systems may lie within radio frequency electronics that take advantage of the high electron mobilities of materials such as graphene, TMDs and phosphorene [2, 54]. The high saturation velocity/mobility coupled with the mechanical stability, low manufacturing cost, low power consumption, position these materials as useful candidates in radio frequency electronics that are conformal, flexible and ubiquitous. As the radio frequency range of operation is dictated in part by the saturation velocity, the high mobility of graphene (10^6 cm^2/Vs) positions the material as most useful in operational frequencies ranging from 100 GHz to as high as 10 Thz, as depicted in Fig. 4a [55, 57]. With terahertz communications systems promising extraordinarily high data rates, solid-state devices optimized for operation at these frequencies would enable a new generation of communication systems. Graphene FET devices transferred to bendable glass with centimeter range bending radius revealed a record intrinsic cutoff frequency f_T of 100 GHz (Fig. 4b) [55]. Graphene transistor [58] devices are not just very useful for terahertz communication systems [59], but the high carrier mobility, gapless spectrum and frequency-independent absorption make graphene very promising for terahertz detection and modulator systems [60].

Semiconducting 2D materials including TMDs and phosphorene currently show promise at sub-terahertz frequencies around 1–20 GHz, where common applications include smart gadgets and Internet of things. Phosphorene exhibits a bandgap around 2 eV in the monolayer case and transistor devices show a high cutoff frequency, extremely high saturation velocity and resilience to applied strain (Fig. 4c and d). It has been clear that phosphorene may be the optimal 2D material for certain high-frequency applications [6], yet the material is still extremely difficult to synthesize and to maintain its chemical stability in ambient environments, two advances that will have to be addressed before any path toward commercialization becomes clear [61–63]. The TMD materials including molybdenum disulfide do not have as favorable electronic properties as phosphorene, but are progressing toward wafer-scale synthesis and are much more robust in ambient conditions [64–66]. Scalable bilayer MoS_2 films fabricated into RF transistors on flexible polyimide substrates show extremely high ‘on’ currents of 1.52 mA μm^{-1}, a high cut-off frequency and maximum frequency of oscillation at 4 GHz and 9 GHz (Fig. 4e and f) and even demonstrate mixing performance (Fig. 4e and g) [56]. Vertically oriented RF devices based on 2D materials also represent an exciting platform for high-frequency and power operation. In a vertical device geometry, the on/off characteristics rely on quantum tunneling across the thickness of the 2D material, rather than the thermionic emission processes found in lateral MOSFETs [67]. Hot electron transistor devices using single-layer graphene and TMDs [68, 69] have demonstrated high current densities, efficient electron injection and high-frequency operation approaching THz operation [70]. As the communications industry is seeking beyond-silicon devices that operate at higher frequencies and provide some mechanical reliability, 2D materials may find a place in this exciting technology space in both horizontal and vertically-oriented device architectures.

OUTLOOK

While 2D materials provide unique attributes for flexible electronic systems, advances in key areas such as repeatable synthesis, robust manufacturing and reliable mechanical characterization are still necessary. Synthesis methodologies such as chemical vapor deposition, metal-organic chemical vapor deposition, sputtering, pulsed laser deposition, exfoliation and more continue to push the limits of large area, high quality and uniformity [71]. With the access to over 1000 different 2D materials [72], it is important that the community continues to develop effective strategies for scalable synthesis while balancing and exploring the unique physics exhibited by newer members of the 2D family, including elemental materials, MXenes, heterostructures and twisted systems. Manufacturing of flexible electronics based on 2D materials compatible with silicon-based device fabrication will be among the first to achieve large-scale commercialization, with novel and unique manufacturing strategies requiring longer development times.

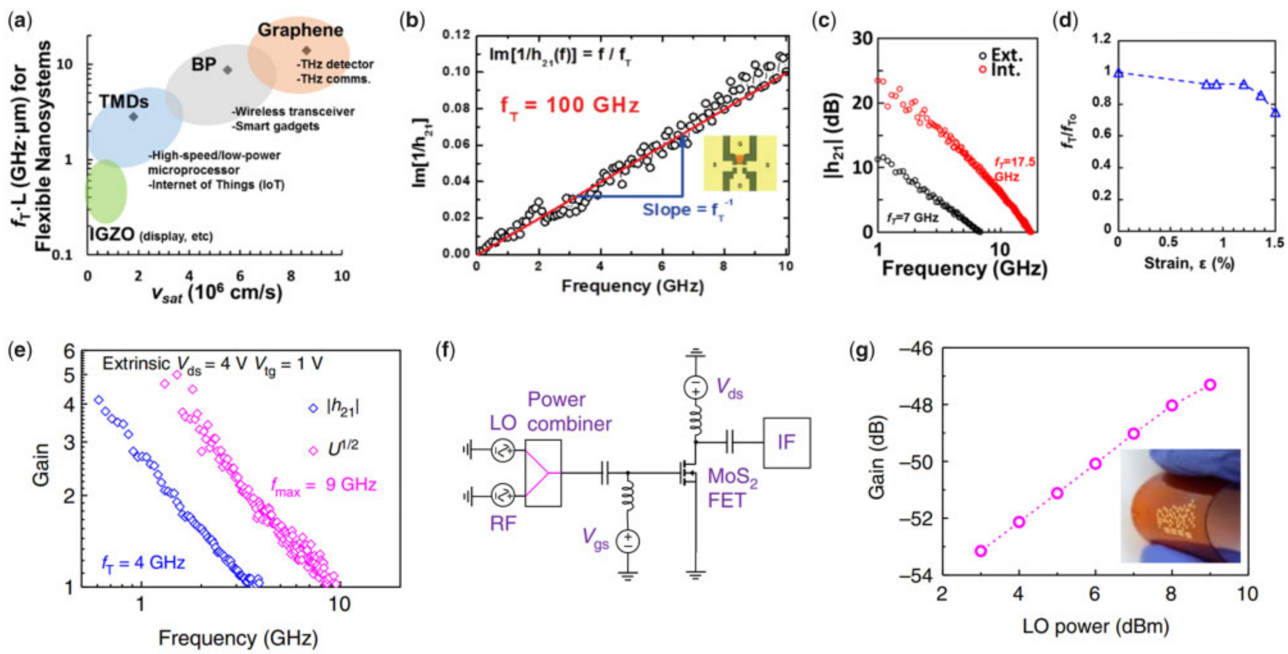

Figure 4: The 2D materials in flexible high-frequency applications. (a) Graphical depiction of frequency device metrics of various 2D materials [55], (b) radio frequency device performance of a graphene transistor device [55] (c) gain performance of phosphorene transistor [6] (d) RF performance retention of phosphorene transistor under strain, (e) RF performance of bilayer MoS_2 transistor (f) circuit schematic of Fet-based RF mixer and (g) conversion gain as a function of LO power of mixer on flexible substrates [56]. Figures reproduced from (a, b,) Ref. [55] Copyright (2015) IEEE, (c, d) Ref. [6] Copyright (2016) American Chemical Society, (e–g) Ref. [56] Copyright 2018 under the terms of CC-BY 4.0.

Utilizing 2D materials for high-performance device transfer to inexpensive flexible substrates may aid in this manufacturing challenge. Finally, as is the case with all flexible electronics, special attention toward understanding the role of mechanical deformation on both immediate and prolonged impacts on device performance is required. For instance, wearable electronics experience a broad range of dynamic strains and strain rates, where conformal electronics may be exposed to a constant strain at the same rate for years. Additionally, future fully-packaged flexible electronic systems with 2D materials will require an understanding of the strain tolerance of not just the active nanomaterial, but also the dielectrics, contact metals and surrounding materials that may govern the overall mechanical reliability limits. Clearly, there are many challenges in realizing 2D material-based flexible electronics, but the exciting combinatorial material properties will certainly be in the conversation for years to come as potential game-changers in a growing technological field.

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AUTHORS' CONTRIBUTIONS

N.R.G., C.M. and M.S. all drafted sections within the manuscript. N.R.G. compiled and finalized the manuscript.

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