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Perspective

Two-dimensional materials for bio-realistic neuronal computing networks

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SUMMARY

Two-dimensional (2D) van der Waals materials have found broad utility in a diverse range of applications including electronics, optoelectronics, renewable energy, and quantum information technologies. Meanwhile, exponentially growing digital data coupled with the ubiquity of artificial intelligence algorithms have generated significant interest in edge neuromorphic computing as an alternative to centralized cloud computing. The drive to incorporate neuroscience principles into computing hardware is motivated by the low power consumption, parallel processing, and reconfigurability of the human brain. The diverse library of 2D materials with atomic-level thicknesses, exceptional electrostatic tunability, and integration versatility is particularly well-suited for realizing bio-realistic synaptic and neuronal functionality. Here, we summarize past and present work in this field and outline the frontier challenges that have not yet been overcome. We also delineate potential solutions and suggest that the neuroscience principles of criticality and synchrony have the potential to inspire breakthrough applications of 2D materials in neuronal computing networks.

PROGRESS AND POTENTIAL

Two-dimensional (2D) van der Waals (vdW) materials have shown unique electronic properties, form factors, and prototype devices that are well-positioned to accelerate the development of neuromorphic computing hardware. In particular, 2D materials and vdW heterojunctions can facilitate novel bio-realistic devices, algorithms, and integration schemes. Recent research in this field has produced diverse experimental demonstrations ranging from high-speed non-volatile memory switches in vertical geometries to gate-tunable, multi-terminal, multi-state synaptic devices in lateral geometries, thus positioning 2D materials to realize bio-realistic phenomena such as neuronal synchrony and criticality. Interdisciplinary cooperation among neuroscientists, materials scientists, and electrical and computer engineers coupled with precise control over the growth, assembly, and fabrication of 2D materials and vdW heterojunctions will be critical for achieving bio-realistic devices with the speed, scalability, and reliability required for high-performance computing.

INTRODUCTION

The realization of neural network algorithms in computing hardware holds promise for off-grid, low-energy processing of the immense data being generated in consumer electronics, healthcare, public safety, and environmental sciences.[1–4] Emerging neuromorphic hardware simplifies computational tasks, expedites analog calculations, and achieves parallel operation that exceeds conventional silicon-based complementary metal-oxide semiconductor (CMOS) technology.[5,6] Demonstrated neuromorphic hardware often relies on non-volatile memory (NVM), such as memristive and phase change devices, which have been realized using traditional bulk materials.[3,6] In parallel, two-dimensional (2D) van der Waals (vdW) materials have emerged as promising alternatives to bulk materials for ultrafast, compact, multi-state, and tunable NVMs.[4,7,8] While replacing incumbent NVMs with 2D materials still faces practical challenges, the unique properties of atomically thin devices provide additional opportunities for realizing bio-realistic neuronal computing networks with functionality that extends beyond existing neuromorphic paradigms. While current synaptic devices have demonstrated basic learning rules from theoretical neuroscience (e.g., Hebbian learning) using spiking neural networks (SNNs),[4–6] this approach is based on average spike rate models that lose temporal correlations between spikes.[9] In contrast, bio-realistic computing paradigms beyond spike rate models (e.g., synchrony, criticality, and recurrent feedback) require non-averaged device characteristics in heterogeneous circuits to establish correlated responses.[9–13]

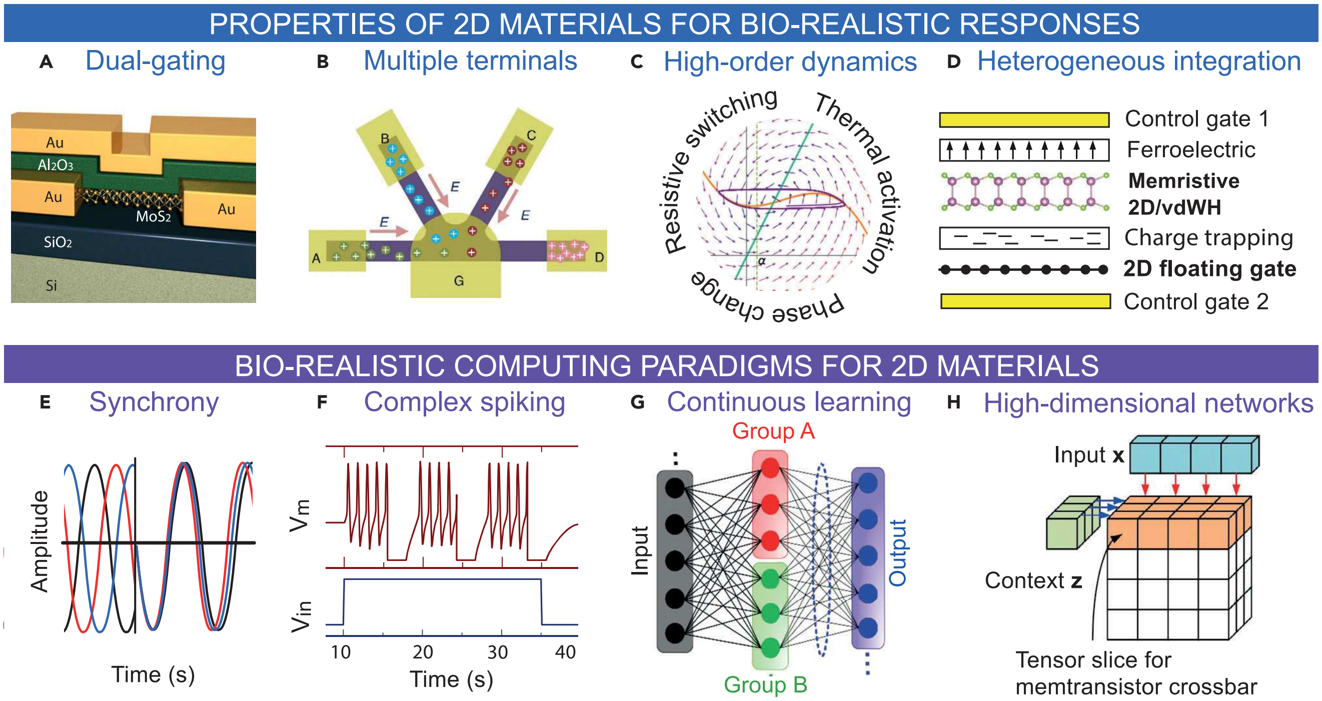

Figure 1. An outline of relevant properties of 2D materials for bio-realistic computing paradigms

(A) Schematic of a dual-gated monolayer MoS_2 memtransistor. Reproduced with permission.[15] Copyright 2020, Wiley-VCH.

(B) Schematic of a multi-terminal synaptic transistor obtained by reversible Li^+ intercalation in a few-layer MoS_2 flake. Reproduced with permission.[19] Copyright 2018, Springer Nature.

(C) Schematic showing the correlation among the internal state variables of resistive switching, thermally activated processes, and phase changes in higher-order 2D devices.

(D) Schematic showing heterogeneous integration of different memory elements, such as ferroelectric, floating gate, and resistive memories, for multiple synaptic responses with different characteristic time scales.

(E) Schematic showing the onset of transient synchrony in sinusoidal waves.

(F) Simulated complex bursting behavior from a spiking neuron circuit consisting of an anti-ambipolar vdW heterojunction transistor. V_{in} and V_m are the input voltage and membrane potential, respectively. Reproduced with permission.[16] Copyright 2020, Springer Nature.

(G) A schematic of a spiking neural network where two groups of neurons from the hidden layer are activated by different gate-tunable learning behaviors of MoS_2 memtransistors to achieve continuous learning. Reproduced with permission.[32] Copyright 2021, American Chemical Society.

(H) Schematic of a 3D dynamical neural network where the third dimension represents the context that is mapped to gate inputs of the memtransistor crossbars. Reproduced with permission.[33] Copyright 2022, Frontiers.

Here, we explore the attributes and properties of 2D materials that make them suitable for bio-realistic implementations of neuromorphic hardware.[1–3,7,14] For example, the atomically thin channels of 2D materials coupled with the atomically abrupt interfaces in vdW heterojunctions result in exceptionally strong electrostatic coupling, especially in a dual-gated geometry (Figure 1A).[15,16,17] This exquisite gate control enables tunable synaptic learning without relying on complicated circuitry to control the pulse shapes as is often required in conventional memristor-CMOS circuits. The planar architecture of 2D materials is also a natural platform for multi-terminal synaptic devices to mimic biological functionality (e.g., heterosynaptic plasticity) and implement reservoir computing algorithms (Figure 1B).[18,19] Because biological neurons are best modeled as nonlinear dynamical systems based on several independent variables (e.g., ion concentrations and membrane potential), bio-realistic spiking circuits require higher-order electronic devices that embody multiple state variables and internal oscillators. The diverse library of 2D materials with correlated electronic, thermal, magnetic, and photonic responses is ideal for realizing these higher-order devices for better emulation of criticality in the

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Hodgkin-Huxley neuron model (Figure 1C).[4,7,20–22] Moreover, bio-realistic responses require a delicate balance of excitatory and inhibitory processes operating on diverse time scales. Toward this end, the vdW bonding of 2D materials facilitates integration with diverse memory elements such as ferroelectric transistors and floating gate memories (FGMs) to achieve a combination of both short-term and long-term processes (Figure 1D).

Emerging bio-realistic computing paradigms for 2D materials include synchrony, complex spiking, continuous learning, and higher-dimensional dynamical neural networks. While neural networks based on spike rate models require homogeneous device responses, alternative approaches (e.g., reservoir computing) exploit heterogeneity within the device channel but do not utilize synchrony.[11] Although synchrony can arise from recurrent neuronal connections through feedback that is relatively insensitive to inputs, computationally relevant synchrony is elicited by stimuli where invariants in stimuli are decoded by synchrony-based processes (Figure 1E).[11,23] For example, in the Jeffress model of sound localization, the invariant in two sound waves reaching the two ears determines the location of the source of the sound.[24] Neuronal synchrony is also the basis of key cognitive and sensory tasks in the human brain including visual sensory pathways,[25] the Licklider auditory pitch perception model,[26] mechanisms of odor differentiation,[27] and anomaly detection in the cerebellum.[28–30] Since neuronal synchrony relies on coincidence detection that is achieved by the competition between excitatory and inhibitory processes, the heterogeneous integration potential of 2D materials holds promise for realizing solid-state electronic analogs.

Because current neuromorphic hardware has been optimized for dense circuits of fast NVMs for processing large data sets, the activation functions are based on simple integrate-and-fire neuron models or related machine learning algorithms (e.g., rectified linear units) that deviate significantly from their biological counterparts. In contrast, complex spiking behavior and variable short-term facilitation are of central importance in biological systems such as the climbing fiber-Purkinje cell circuit.[31] Similarly, 2D materials and vdW heterojunctions can achieve complex spiking behavior (Figure 1F)[16] with simplified circuitry compared with CMOS, although further advances are required for resonate-and-fire and differentiate-and-fire neurons. Another attribute of biological systems is continuous learning throughout the lifetime of an organism, which has been difficult to emulate in artificial neural networks (ANNs) that are hardwired to solve specific problems. Recently, SNNs based on the gate-tunable learning behavior of 2D memtransistors have shown promise for achieving continuous learning in solid-state electronics (Figure 1G).[32] Further improvements can be expected when the benefits of dual-gating and heterogeneous integration are fully exploited in atomically thin material platforms.

Learning in biological systems is highly adaptable to different inputs where the brain self-regulates based on the context or the situation. This context-based learning is currently lacking in existing NVM hardware that is optimized for matrix multiplication and pattern recognition, in part due to the architecture constraints imposed by the constituent two-terminal memristors. On the other hand, the gate electrodes in dual-gated 2D memtransistors allow the context input to be mapped to a separate dimension, thus achieving higher-dimensional hypernetworks for bio-realistic learning paradigms (Figure 1H).[33] Hypernetworks also improve predictive robustness for complex tasks by simultaneously processing both input and context information, thereby suggesting additional opportunities for multi-terminal 2D synaptic devices in bio-realistic neuronal computing systems.

In this perspective, we first summarize the most noteworthy historical developments in neuromorphic devices and circuits based on 2D materials while directing the reader to more detailed discussions of specific 2D devices, mechanisms, and comparative metrics in review articles published elsewhere.[3–5,7,8,34,35] We then delineate recent developments in 2D and vdW heterojunction devices in the context of bio-realistic implementations. These recent developments also provide context for the remaining challenges facing 2D materials such as scaling, device reliability, and incomplete understanding of the diverse mechanisms that underlie novel functionalities. Finally, we identify emerging neuromorphic computing paradigms where 2D materials are best positioned to have an impact including the bio-realistic concepts of criticality and synchrony.

PAST

For neuromorphic hardware, materials innovations have been primarily driven by the search for NVMs that can surpass the performance metrics of CMOS-based static and dynamic random-access memory. However, because synaptic connections outnumber neurons by multiple orders of magnitude in biological systems, bio-realistic NVMs would ideally possess the following attributes: high switching speeds, high-density nodes, high retention, high endurance, and multiple non-volatile states for high-resolution analog operations. Bio-realistic NVMs should also have an intrinsic propensity to implement weight update rules, such as the linear learning behavior in multi-level perceptrons and spike-timing-dependent plasticity (STDP) in SNNs. In this context, early research in NVM-based synaptic memory focused on 2D transition metal dichalcogenides (TMDCs), black phosphorus, hexagonal boron nitride (hBN), MXenes, post-transition metal chalcogenides, and vdW heterojunctions between these components.[4,15,18,36–41] Additional types of synaptic devices based on 2D floating gates, charge traps, ferroelectric transistors, photonic systems, and memristive memories have already been reviewed elsewhere.[4,7,34,35,42–44] In particular, Schuman et al. and Xia and Yang[2,5] provide a discussion on the generally accepted differences between bio-inspired (closer to machine learning) and bio-realistic (closer to theoretical neuroscience) computing paradigms.

Vertical 2D memristors have been demonstrated on few-layer and monolayer TMDCs, few-layer hBN, and composite films of solution-processed 2D materials (Figure 2A). The main advantages of atomically thin memristors are high thermal stability, mechanical flexibility, low-energy switching, and controllable synaptic response based on voltage pulses. These 2D memristive devices have shown a variety of switching mechanisms including phase change or resistive switching, filamentary or non-filamentary switching, and unipolar or bipolar switching.[38,39,41,45–49] As one example, the coexistence of bipolar and unipolar (or threshold) switching in hBN memristors is useful for realizing self-selective crossbar arrays without the need for an additional selector or transistor.[39,46] In addition, atomically thin vertical channels are expected to show small switching voltages (V_{switch}). However, except for one example of electrochemical switching in bilayer MoS_2 ($V_{switch} \approx 0.1$ V),[47] most vertical memristors show relatively large switching voltages ($V_{switch} > 1$ V for monolayer MoS_2) and fields (>6 MV/cm) that do not present significant advantages over conventional metal-oxide memristors.[5,41,48] Nevertheless, the writing process can still improve energy efficiency by limiting the current compliance (I_{comp}). In this manner, vertical oxidized hBN memristors (thickness ≈ 0.9 nm) can be switched ON by limiting I_{comp} to the picoampere scale, which translates into femtojoule synaptic operations. Ultrafast switching in TMDC memristors has also shown promise for zero-static power RF switches (50 GHz) due to the ultralow ON-state resistance of

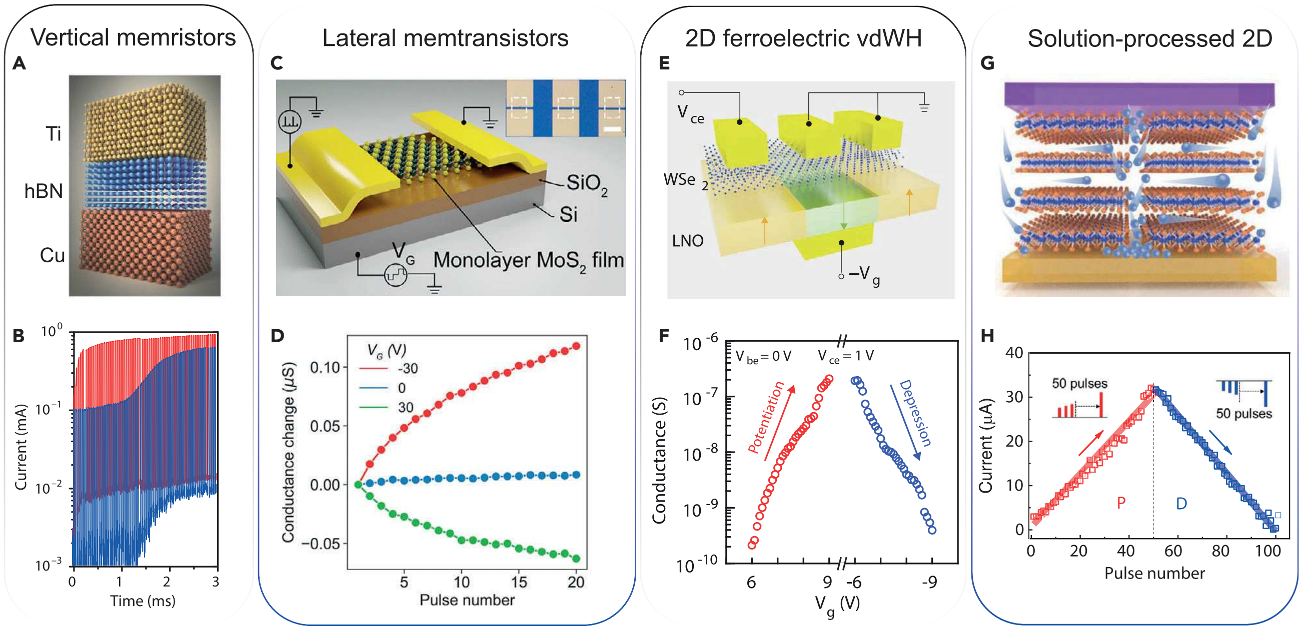

Figure 2. Selected examples of past research in neuromorphic devices based on 2D materials

(A) Schematic of a vertical hBN memristor. Reproduced with permission.[46] Copyright 2017, Wiley.

(B) Learning behavior of Au/Ti/h-BN/Au memristors where red and blue curves show LTP at voltage pulses of 0.9 V and 0.8 V, respectively. Reproduced with permission.[50] Copyright 2020, Springer Nature.

(C and D) (C) Schematic and (D) learning behavior of a monolayer MoS_2 memtransistor showing a transition from LTP to long-term depression (LTD) as the gate voltage (V_G) is changed from −30 to 30 V. Reproduced with permission.[32] Copyright 2021, American Chemical Society.

(E and F) (E) Schematic and (F) learning behavior of a bipolar junction transistor obtained by a vdW heterojunction between monolayer WSe_2 and ferroelectric $LiNbO_2$ film. Reproduced with permission.[66] Copyright 2021, AAAS.

(G and H) (G) Schematic and (H) learning behavior of a vertical solution-processed MoS_2 memristor obtained by electrochemical exfoliation. Reproduced with permission.[40] Copyright 2022, Springer Nature.

10 Ω.[48] By using the areal scaling of the intrinsic capacitance, a cutoff frequency of 10 THz has been extrapolated for sub-micron RF switches, making them suitable for 5G network technology.[3,48]

In the context of neuromorphic computing, hBN memristors are more promising than TMDC memristors due to their low ON-state current, operation as both bipolar and unipolar devices, and demonstration of bio-realistic responses such as pulsed pair facilitation, short-term and long-term potentiation (LTP), and STDP (Figure 2B).[3,49,50] Both TMDC and hBN memristors have been demonstrated at the wafer scale using chemical vapor deposition (CVD).[8,38,49,51] With respect to lateral size, the individual device footprint is determined by the width of the metal lines, which implies that similar areal scaling can be achieved as conventional metal-oxide memristors that have already been integrated with CMOS to realize the NeuRRAM chip with millions of synapses.[1,6] Therefore, 2D vertical memristors are likely to remain relevant for emerging hardware paradigms if bio-realistic responses can be achieved using interlayer diffusion and relaxation mechanisms of metal ions, defects, and other atomic species (Figures 2A and 2B).[3,46,48–50,52]

In addition to being the original device architecture envisioned for neuromorphic computing by Carver Mead,[53] lateral transistor-based digital and FGMs are also the leading commercial memory technologies. Lithography methods for chip integration on planar wafers are ideally suited for miniaturized lateral three-terminal devices. While lateral synaptic transistors allow fine control of conductance changes from gate voltage pulses due to the deterministic nature of charge trapping in dielectrics or redox reactions in the channel, vertical memristors suffer from relatively poor control over the synaptic response due to the stochastic nature of resistive switching. On other hand, vertical memristors have the advantage of simplicity for denser integration and faster switching speeds. Despite atomic-level channel thicknesses in vertical memristors, both lateral and vertical technologies face the same fundamental scaling limits determined by lithography resolution. In contrast, memtransistor resistive switching involves changes in Schottky barrier height driven by drain voltage pulses, while the gate terminal provides additional control over NVM and synaptic learning behavior that is not possible in synaptic transistors or memristors.[7] Lateral devices on polycrystalline monolayer MoS_2 were the first to reveal intrinsic memristive switching in 2D materials that could be controlled by grain boundary structures and gate voltage to yield gate-tunable memristors or memtransistors.[36] Recently, low-voltage 2D memtransistors have been realized using CVD-grown MoS_2 or exfoliated flakes with post-fabrication defect engineering.[54–57] Both 2D synaptic transistors and memtransistors can mimic the multi-terminal morphology of biological neurons that is central to heterosynaptic plasticity and non-Hebbian learning phenomena (Figures 2C and 2D).[7,15,18,32,33,56,58,59] Lateral memristive devices using intercalated ions in layered TMDCs provide another promising direction for multi-terminal synapses (Figure 1B), but this approach faces challenges stemming from poor memory retention and limited gate tunability in the metallic 1T′ phase.[19]

Recently, FGM devices, vdW heterojunctions, Mott metal-to-insulator transition (MIT) switches, and ferroelectric transistors have shown memristive switching in the lateral geometry with varying degrees of gate tunability.[7,60–62] FGM devices using graphene, hBN, and monolayer MoS_2 layers have achieved memristive responses (e.g., memflash), but the tunneling mechanism from the metal contacts precludes gate tunability of the synaptic response.[63] Meanwhile, weak electrostatic screening in monolayer MoS_2 has allowed the integration of the control gate and floating gate (FG) on opposite sides of the graphene/hBN/MoS_2/hBN/FG stack, where the drain/hBN/FG tunnel barrier is used for the writing process and the graphene control gate is employed to further tune the resistance states.[64] However, this scheme suffers from two problems: (1) the control gate only tunes the resistance states but not the synaptic learning rate and (2) an additional selector element for crossbar integration is required. Nevertheless, 2D materials have significantly improved the switching speed for FGMs. Whereas conventional FGM is limited to slow speeds (e.g., 75 μs for NAND flash) due to relatively large electron/hole tunneling barriers, the vdW heterojunction of MoS_2/hBN/graphene has achieved a writing speed of 20 ns by exploiting a large gate coupling ratio, atomically clean interfaces, smaller tunneling barriers, and triangle-barrier-modified Fowler-Nordheim tunneling.[65]

Phase change and ferroelectric memtransistors are particularly relevant for low energy and high speed since the switching operation only requires subtle displacement of atoms, often within the same unit cell. For example, Mott MIT-based memtransistors on polycrystalline VO_2 films have shown a switching voltage of 1 V and a speed of 35 ns in large-channel devices (100 × 1,000 μm^2), resulting in femtojoule synaptic operations.[61] Layered ferroelectric materials have expanded the functionalities of conventional ferroelectric synaptic transistors by coupling in-plane and out-of-plane polarizations that not only allow electrostatic gating but are also suitable for multi-terminal synaptic responses.[60] For example, few-layer α-In_2Se_3 memtransistors show low-voltage memristive switching and excellent photoresponses that can be exploited to decouple the read and write currents for low-energy inference.[60]

Another promising area of exploration is the *heterogeneous* integration of 2D materials with conventional ferroelectrics (Figure 1D) to achieve *homogenous* integration mimicking biological systems (i.e., co-location of computation and memory). Integration of NVM-based synaptic devices with CMOS peripheral circuits relies on analog-to-digital conversion (ADC). Both synaptic operation and ADC operation have been achieved in a bipolar junction transistor (BJT) based on the heterojunction between ferroelectric $LiNbO_3$ and monolayer WSe_2. Proximity effects from the ferroelectric domains in $LiNbO_3$ have been exploited from adjacent domains in WSe_2 with heavy electron and hole doping (controlled by a bottom gate) to produce lateral p-n-p or n-p-n BJTs where each domain in WSe_2 is contacted with metal pads on top. Consequently, the same device can be used as an operational amplifier or as a synaptic transistor by pulsing the gate voltage on the $LiNbO_3$ domain connected to the base, resulting in a high dynamic range of synaptic weights ($>10^3$) in a co-located computing-memory architecture (Figures 2E and 2F).[66] Additional gate tunability of the synaptic operation has not yet been achieved, but further integration with hBN/graphene stacks is a promising route.

Liquid phase exfoliation and solution-processing of 2D materials provide additional routes for scalable manufacturing with most efforts to date focused on printed flexible electronics, optoelectronics, and sensors.[67] Solution-processed 2D memristive and synaptic devices have rapidly advanced since the first demonstration in 2015.[40,68–70] In the majority of these cases, electrochemically active metals (e.g., Cu and Ag) have been used as the switching filament, which obscures the role of the 2D materials in the composite films, although these devices have realized low-voltage operation (<0.2 V) in thick films (>100 nm).[68,69] Recently, the combination of electrochemical intercalation of tetraheptyl ammonium bromide and subsequent megasonic exfoliation has produced a large fraction of high-quality monolayer MoS_2 flakes,[71] which are likely to be well-suited for solution-processed 2D semiconductor neuromorphic devices. For example, electrochemical exfoliation of MoS_2 has realized wafer-scale vertical memristors using noble metals where the resulting devices show an intrinsic switching mechanism that can be attributed to sulfur vacancy migration along the edges of the nanoflakes (Figures 2G and 2H).[40] In addition, large-area solution-processed memristors provide opportunities for *in situ* visualization of the switching mechanism.[70] Solution-processed 2D materials have further shown thermally activated volatile switching that is likely triggered by the large and localized electric fields at the edges of nanoflakes. These nonlinear device characteristics have been generalized to different 2D semiconductors, thus presenting a new platform for all-printed spiking neurons for both memory and activation functions.[70]

PRESENT

Recent work has attempted to realize prototype devices for bio-realistic computation paradigms (Figures 1E–1H) using 2D materials and vdW heterojunctions. The bio-realistic synchrony-based computing paradigms discussed here (Figure 1E) are distinct from the synchronous weight update learning rules in ANNs or CMOS-based computers controlled by a clock. In biological systems, synchrony plays a fundamental role in different classes of specialized computing systems ranging from detecting the invariant in sensing inputs (e.g., auditory system) to rapid identification of true anomalies in the cerebellum before signals are disseminated for motor control. A top-down approach for hardware realization is to first develop an *algorithm* of the complete system (e.g., synchrony in retinal ganglion cells)[72] and then map the full model onto existing CMOS hardware with and without the accelerators from emergent NVM devices for edge computing tasks. In contrast, a bottom-up approach is to first develop a *physical model* of individual components in a biological system (e.g., coincidence detection neurons) and then use the properties of emergent materials to simplify or accelerate the unit circuit that can then be integrated with CMOS periphery circuits for full system realization.

The bottom-up approach can benefit from advances in materials, devices, and circuits to achieve the relevant bio-realistic response. For example, the Jeffress model of sound localization is a spatiotemporal computing model that converts the phase difference or interaural time difference (ITD) between the sound waves received through the two ears into a spatially localized activity within a neural network (Figure 3A).[24,37] The three main components are (1) delay neurons that achieve ITD by varying the length of axons from the left and right cochlear nuclei, (2) coincidence detection neurons that fire only when input spikes are received within a finite interval, and (3) a spatial computation map of ladder-like structure with each coincidence detector situated between two delay neurons on the left and right side (Figure 3A). In this manner, the azimuthal angle of the sound source with respect to head orientation is converted into spatially addressable information with precision down to a single degree. A delay neuron can be realized from a tunable RC circuit using a field-effect transistor (FET). The planar architecture of monolayer MoS_2 presents a promising platform for coincidence detection neurons where a series of split gates (with varying distances between them) are fabricated on a MoS_2 FET in a ladder-like structure (Figure 3B).[37] Each pair of split gates can be used as a NAND gate (and coincident detector) since the current path in the MoS_2 channel is blocked only when both gates receive signals from the left and right axons within an interval.[37] While this function in principle could be realized using any FET, the dual gating of MoS_2 offers a unique advantage (Figure 1A). In particular, neuroplasticity can be achieved in this auditory system by using a dual-gated MoS_2 FET, where the global bottom gate can further tune the inhibition ratio of the channel current. Since coincidence detection is a versatile function in synchrony-based computing models beyond auditory, visual, and olfactory systems, the demonstration of this functionality in 2D devices has broad potential applications.

Another commonly observed function in synchrony-based computation is the competition between excitatory and inhibitory processes. For example, in the cerebellum, the granule cells generally inhibit the Purkinje cells and cerebellar nuclei, but an anomalous and persistent input signal from mossy fibers can cause a strong excitatory response that ultimately elicits an output signal to produce motor control.[29,30] A simpler example of excitatory-inhibitory neuronal competition is found in the neurobiological architecture of vision systems in insects (e.g., lobula giant movement detectors in locusts).[73] These movement detectors enable insects to maneuver in large swarms without colliding and could inspire hardware vision systems in autonomous vehicles. The underlying mechanism involves a multiplicative operation within a neuron where the broader features of angular size and angular velocity of looming objects determine the non-monotonic firing rate. The time evolution of angular size and velocity of the incoming objects is translated into excitatory and inhibitory signals that are fed into a multiplicative transfer function responsible for signal generation for motor interneurons.[73] Consequently, a nonlinear function allows highly precise discrimination between the objects in direct line of sight and the objects passing nearby amid a rapidly changing background. Heterogeneous

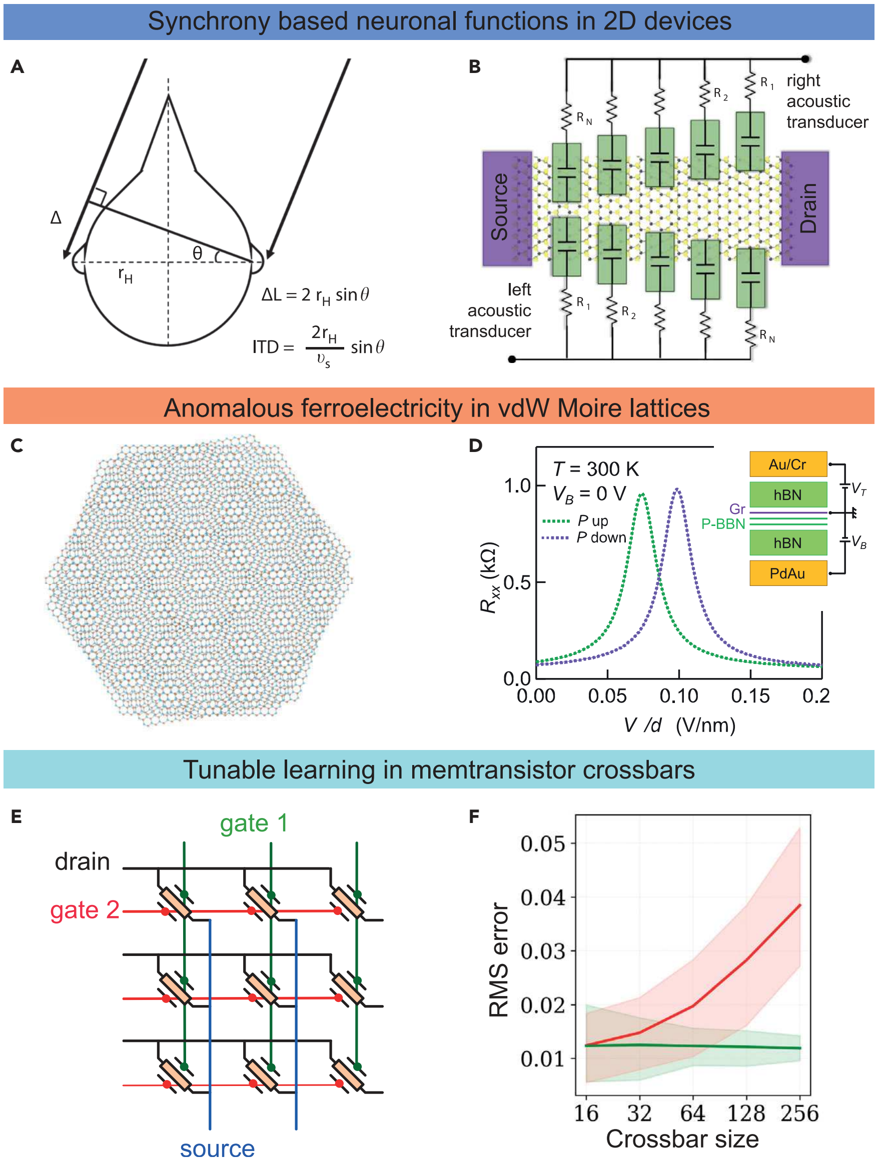

Figure 3. Recent examples of bio-realistic devices and circuits based on 2D materials and vdW heterojunctions

(A) Schematic showing the ITD between the sound waves reaching the two ears of a barn owl.
(B) Schematic of a MoS_2 transistor with five pairs of split-gate electrodes with varying channel overlaps (gate capacitance) to mimic the ITD response.
(A and B) Reproduced with permission.[37] Copyright 2020, Springer Nature.
(C) Schematic of a parallel stacked bilayer of hexagonal boron nitride (P-BBN) showing the formation of a moiré lattice.
(D) A plot of longitudinal resistance (R_{xx}) versus top-gate voltage (V_T) (d_T is the thickness of the top hBN) in a graphene/P-BBN heterostructure showing hysteresis due to anomalous ferroelectricity of the moiré lattice at room temperature. The inset shows a schematic of the dual-gated hBN-encapsulated graphene/P-BBN device. Reproduced with permission.[80] Copyright 2021, AAAS.
(E) Schematic showing a crossbar array of dual-gated memtransistors.
(F) Comparison of the root mean square (RMS) error of the training weights as a function of crossbar size for memristors and memtransistors.
(E and F) Reproduced with permission.[33] Copyright 2022, Frontiers.

integration of different memory and response functions could achieve this competitive behavior in 2D devices (Figure 1D). For example, a positive photocurrent in MoS_2 phototransistors has been used to generate an excitatory response as a function of incident light.[74] These MoS_2 phototransistors have been integrated with a non-volatile FGM stack of Al_2O_3/Pt/TiN/p^{++}-Si where an inhibitory response is obtained by using the floating gate separated by the Schottky barrier at the TiN/p^{++}-Si interface.[74] The desired nonlinear multiplicative response function is then achieved by tuning the gate voltage pulses.

Similar competing processes can also be achieved by integrating intrinsic short-term facilitation (STF) in 2D materials with intrinsic or extrinsic NVM. Semiconducting 2D magnetic/multiferroic materials such as NiI_2 and semiconducting 2D ferroelectric materials such as In_2Se_3 provide interesting alternatives where the positive photocurrent of the semiconducting channel could enable STF, while the intrinsic phase change achieves NVM operation.[60,75] Although vertical devices involving the layered antiferromagnet CrI_3 have shown memristive behavior, these responses are currently limited to low temperatures and non-zero magnetic fields.[76] In principle, both excitatory and inhibitory processes could be further controlled by one or dual-gate electrodes (Figure 1A). For example, the optical illumination of a MoS_2 memtransistor can be treated as an additional gating mechanism. MoS_2 photo-memtransistors not only show a large memristive switching ratio ($>10^5$) but also illumination-intensity-dependent synaptic learning rates.[77]

Another potential memory element for heterogeneous integration is the moiré lattice in twisted or parallel stacked 2D monolayers. Recently, 2D moiré lattices have shown a diverse range of many-body effects such as superconductivity, Wigner crystallization, and Mott insulator phenomena (Figure 3C).[78] In the context of neuromorphic devices, twisted vdW heterojunctions between bilayer graphene and hBN have shown anomalous ferroelectricity from the Coulomb blocking effect in moiré sub-band filling.[79] Although the initial results were obtained at low temperatures, this anomalous ferroelectricity has recently been achieved at room temperature by using a vdW heterojunction between graphene and a parallel stacked bilayer of hBN (Figure 3D).[80] Since these ferroelectric devices in a dual-gated geometry can isolate and intermix the effects of doping and vertical displacement fields as a function of the two gate voltage biases, they are ideally suited for functional synaptic and spiking neuron devices.

While simple spiking neurons can serve as effective activation functions in SNN algorithms, biological neurons show a wider range of complex spiking behaviors that play a key role in synchrony-based computation (Figure 1F).[31] For example, complex spiking in Purkinje cells with an initial Na^+ spike followed by prolonged depolarization is a heavily studied subject in the development of the cerebellum neural code.[31] Neuron spiking behaviors are typically modeled as nonlinear dynamical systems of coupled differential equations with multiple parameters, such as membrane potential and concentration of ions, as exemplified by the Hodgkin-Huxley model.[10] Even one pair of seemingly simple coupled equations can generate up to 20 different kinds of spiking and bursting behaviors by minor changes in parameters.[10] However, the physical realization of these nonlinear equations requires dozens of CMOS transistors and capacitors.[81] Scaling concerns thus motivate the development of devices with intrinsic excitatory and relaxation mechanisms that mimic these complex spiking features within more compact circuits. For example, diffusive memristors and memcapacitors operate on the competing mechanisms of Ag ion migration and surface tension of Ag islands within a SiO_x matrix. Consequently, they have been integrated with a pair of transistors to achieve integrate-and-fire spikes.[82]

However, this scheme has not yet realized complex spiking. In this context, the mixed-dimensional vdW heterojunction between monolayer MoS_2 and single-walled carbon nanotubes has simplified the circuit architecture of integrate-and-fire neurons compared with CMOS-based neurons.[16] These dual-gated self-aligned vdW heterojunctions produce non-monotonic anti-ambipolar (i.e., Gaussian) transfer characteristics. In this manner, the monotonic and non-monotonic temporal evolution of Na^+ and K^+ ions can be modeled using conventional transistors and anti-ambipolar transistors, respectively. Therefore, a circuit consisting of one anti-ambipolar transistor and only a handful of additional circuit elements can enable bio-realistic complex spiking behavior beyond constant spiking such as class-I spiking, spike latency, phasic spiking, phasic bursting, tonic bursting, and dampened tonic bursting.[10,16]

Finally, recent work on lateral memtransistors has focused on reducing power consumption below the femtojoule level per event,[61] demonstrating scalable crossbar arrays,[15,56] and realizing bio-realistic functions such as heterosynaptic plasticity.[18,33] The synaptic response of monolayer MoS_2 memtransistors can be further tuned by controlling the stoichiometry and defect density in MoS_2 either during the growth or via post-growth defect engineering.[7,57] Since monolayer MoS_2 grown on atomically flat substrates (e.g., sapphire) has shown a reduced density of defects,[32] the resulting memtransistors possess an attenuated memristive switching ratio but also achieve stronger electrostatic control of memristive switching. Consequently, gate voltage tuning between LTP and depression behavior is possible without having to change the polarity of the pre-synaptic and post-synaptic neuron pulses.[32] Moreover, these MoS_2 memtransistors can achieve qualitatively different learning curves that allow efficient retraining of SNNs to achieve continuous learning (Figure 1G).[32] In another example, dual-gated MoS_2 memtransistor crossbar arrays solve the problem of sneak currents while also allowing gate-tunable learning behavior at each synaptic node.[15] Recent circuit-level simulations show that gate-tunable weight updates in memtransistors can significantly reduce the root mean square error in the weights of large-scale crossbar arrays compared with similar crossbar arrays based on conventional memristors (Figures 3E and 3F).[33]

PROBLEMS

Although progress has been made toward the realization of bio-realistic functions in individual 2D devices and small-scale circuits, numerous practical considerations have not yet been addressed for system-level implementations including detailed mechanistic understanding, performance metric optimization, scaling, integration, and device-to-device variability. The diversity of 2D materials and device characteristics implies that a range of physical mechanisms is possible, thus complicating the identification of the universal or dominant mechanism.[7,57] Typically, memristive switching in lateral memtransistors involves dynamic modulation of the effective Schottky barrier height (Φ_{SB}) as confirmed by *in situ* Kelvin probe force microscopy (KPFM) and electrostatic force microscopy.[18,77] KPFM measurements have revealed a change in the surface potential by at least 200 mV at both the source and drain electrodes during resistive switching in MoS_2 memtransistors.[77] Variable temperature transport measurements have also shown a change in Φ_{SB} by 40 mV between the two resistance states in MoS_2 memtransistors.[18] Possible physical mechanisms for Φ_{SB} modulation include redistribution of dopants via defect migration within a 2D lattice and/or varying spatial profiles of trapped charges in the dielectric near the contacts.[18,57] A recent self-consistent physics model attempted to describe WSe_2 memtransistors using a lattice heat flow mechanism, selenium vacancy

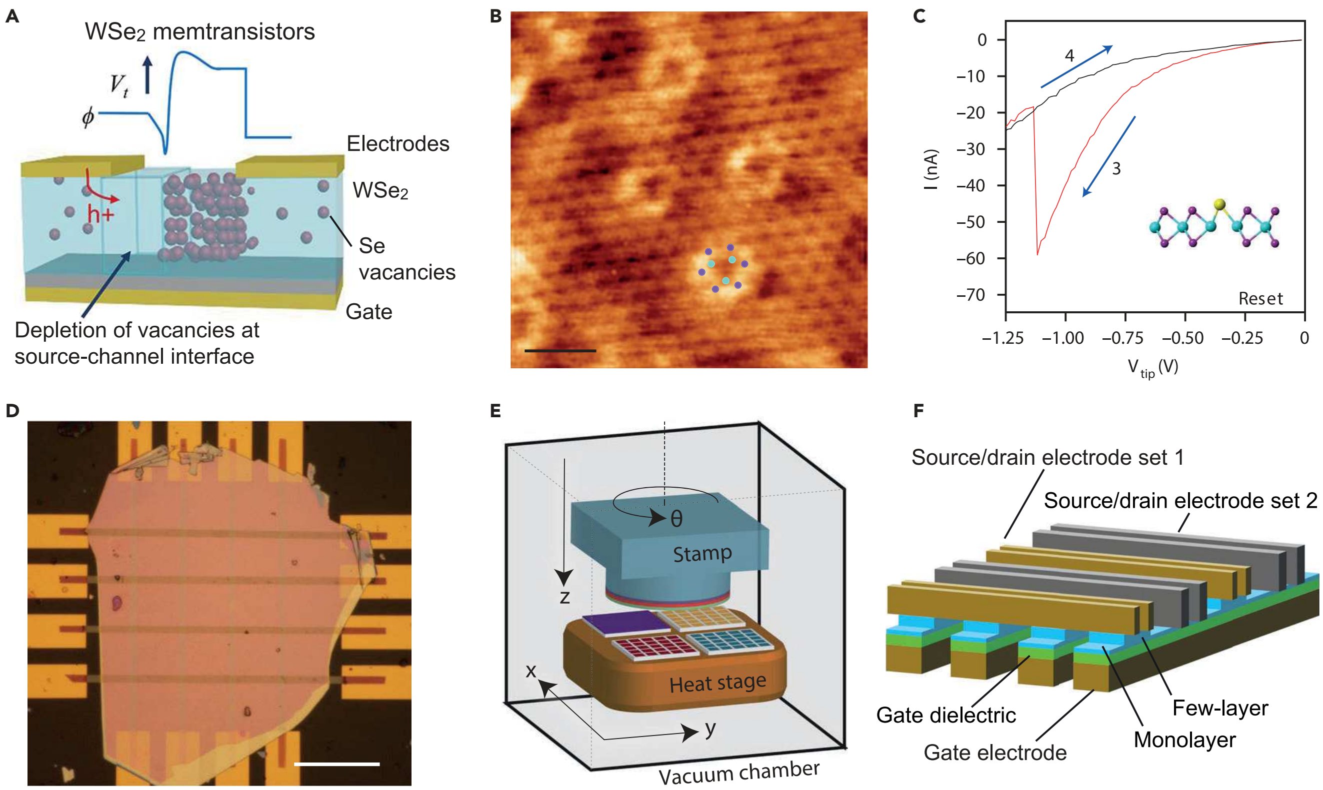

Figure 4. Remaining fundamental and practical challenges for 2D neuromorphic hardware

(A) Schematic showing the switching mechanism in a few-layer WSe_2 memtransistor involving migration of selenium vacancies near the contact-source interface. Reproduced with permission.[57] Copyright 2022, American Chemical Society.

(B) An STM image of a sulfur vacancy in monolayer MoS_2. The scale bar represents 1 nm.

(C) Current versus STM tip voltage showing resistive switching at two different defects. The inset shows a sulfur vacancy in monolayer MoS_2.

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(D) Optical microscopy image of a 4 × 4 crossbar on a few-layer MoS_2 flake. The scale bar represents 20 μm. Reproduced with permission.[38] Copyright 2018, Springer Nature.

(E) Schematic showing automated vacuum assembly of a vdW heterojunction involving 2D materials. Reproduced with permission.[88] Copyright 2022, Springer Nature.

(F) Schematic showing a network architecture of 2D memtransistors based on a periodic array of pre-patterned monolayer and few-layer channel regions.

migration near the contact, and drift-diffusion equations of an FET (Figure 4A). The model not only described the nuances of experimental current-voltage (I-V) characteristics such as self-crossing I-V curves in short-channel devices, but the simulated profiles of dopant density, electric field, and temperature also explained the large voltages required to recover memory states in memtransistors (i.e., reset process). In this model, the memtransistor is approximately modeled as two memristors at the contacts separated by a transistor with the majority of the applied voltage being dropped near the dominant contact resistance. This potential distribution makes it more difficult to reset the memory state in the other contact, which is essential for repeatable bipolar resistive switching.[18,57]

In situ visualization of defect dynamics has been critical in understanding the mechanism in memristors, suggesting that a similar approach can be valuable for lateral memtransistors.[83,84] *In situ* imaging of point defects, such as oxygen substitutional impurities or sulfur vacancies in MoS_2, has been performed through atomically resolved ultra-high vacuum scanning tunneling microscopy (STM) (Figure 4B).[52]

Memristive device characteristics have also been observed in current-voltage characteristics obtained by contacting an individual chalcogen vacancy with an STM tip, which provides evidence for their role in modulating local electronic properties for resistive switching (Figure 4C).[52] However, the large electric fields (>6 MV/cm) across atomically thick vertical TMDC memristors likely imply contributions from additional sources.[48] Therefore, the possibility of multiple mechanisms contributing to switching behavior cannot be ruled out for lateral or vertical devices. Electron microscopy techniques, coupled with device modeling at various length scales, can potentially provide further insight into the structural changes that occur during the switching operation.[83] For example, *in operando* hybrid microscopy approaches, such as scanning transmission electron microscopy and high-angle annular dark field imaging, have strengthened switching mechanism hypotheses by showing enhanced atomic-level spatial variation in imaging contrast, highlighting the structural and chemical changes during switching, and visualizing conductive filament formation/rupture and electrochemical nucleation near electrodes.[46,50,83] Recently, an innovative *in situ* visualization technique was developed through plasmonic coupling of an optically transparent, conductive atomic force microscopy cantilever with a gold nanoparticle electrode contacting an oxide film memristor. This approach enabled spatiotemporal visualization and accumulation of oxygen defects at the active material interface.[84] Diffraction-limited optical microscopy, near-infrared thermal imaging, and photoluminescence mapping have likewise been useful in revealing the switching mechanism in thermally activated memristive switching in lateral solution-processed 2D composite devices.[70]

The long-term expectation of scalable fabrication is additionally contingent on the growth of wafer-scale 2D materials. Wafer-scale growth of 2D materials for neuromorphic devices has been an active research field since the first realization of memristive switching in monolayer TMDCs.[36,49,50] CVD has been widely used to realize wafer-scale products where process variables tune precursor reactivity in the vapor phase. However, due to the all-surface nature of 2D materials, the growth kinetics of 2D materials can be highly sensitive to external variables such as precursor type, growth substrate type, chamber configuration, temperature, and humidity. This tight phase space can yield films of variable electronic quality and crystallinity, resulting in batch-to-batch variation and uncontrolled spatial distribution of heterogeneities within the same batch (e.g., defects, grain boundaries, and varying thicknesses). In addition, suitable growth conditions for some vdW materials have not been achieved or require extreme processing conditions. Fortunately, some TMDCs have robust thermal properties that can withstand moderate operating temperatures. For example, few-layer MoS_2 has been employed in robust vertical memristors with operating temperatures up to 340°C, which is well above the operating window of oxide memristors. These memristors have been integrated into 4 × 4 crossbars on individual flakes without using a selector (Figure 4D). However, wafer-scale growth of high-quality few-layer TMDCs with atomically uniform thickness has not yet been achieved with current growth methods primarily limited to monolayers.[38] An exception is the wafer-scale growth of few-layer hBN, although the demonstrated integration complexity of hBN vertical memristors is still limited to a 10 × 10 crossbar.[49]

Other engineering challenges for 2D neuromorphic devices include power consumption, switching speed, device footprint, integration, and device-to-device uniformity. Operating voltages range from hundreds of millivolts to tens of volts for lateral memtransistors and hundreds of millivolts to a few volts for vertical memristors.[7,47–50,56] Meanwhile, most lateral synaptic transistors and memtransistors show switching times from microseconds to tens of milliseconds. On other hand, vertical TMDC memristors and semi-vertical FGM devices based on atomically abrupt interfaces in MoS_2/hBN/graphene vdW heterojunctions have shown switching speeds of tens of nanoseconds that outperform commercial flash technology.[48,65] Thus, vdW heterojunctions present fertile ground for combining high switching speed with gate-tunable characteristics. The atomically thin bodies of 2D vertical memristors also provide a path to sub-nanometer thickness scaling,[38,52] but lateral scaling challenges remain. For crossbar integration, vertical 2D memristors with multilayer hBN have shown the coexistence of bipolar and unipolar switching.[46] Au/hBN/graphene/hBN/Ag vertical stacks have also been integrated into a self-selective crossbar array with graphene acting as an effective barrier for Ag ion diffusion, thus enabling non-volatile behavior for weight storage and volatile behavior for node selection.[39] However, this crossbar geometry currently has limited scalability due to mechanical exfoliation and transfer of the constituent 2D materials. In contrast, a recent CVD-grown MoS_2 crossbar array employed dual gating to enable selectivity without sneak current pathways, which alleviates some geometrical constraints.[15]

Device-to-device uniformity is another outstanding challenge for both lateral and vertical devices. Inconsistencies in the reporting of device metrics have hampered progress in this direction,[85] and thus a concerted effort is needed to benchmark device behavior and circuit performance metrics against conventional NVM and CMOS-based neuromorphic chips.[1,3,14,86] Unlike CMOS technology, neuromorphic research lacks a clear roadmap, which is especially true in the 2D materials community.[14] In some cases, neural network algorithms have been developed to be relatively immune against variabilities in memristive devices,[87] which has led to the impression that device-to-device heterogeneity is acceptable. However, not all heterogeneity is equal, so well-controlled heterogeneity should be the goal in cases where homogeneity is not easily achieved. Indeed, well-controlled heterogeneity could be useful in computing paradigms that exploit variability, noise, stochasticity, internal dynamics, and oscillations.[11,16,22]

Another opportunity for innovation is the controlled assembly of 2D materials with desired stoichiometry, band offsets, and lattice angles (Figure 4E).[88] Highly controlled assembly is especially interesting for vdW heterojunction memories and moiré anomalous ferroelectric lattices for emerging neuromorphic devices.[65,79,80] Further work to aid in 3D integration includes selective-area growth or selective-area transfer of 2D materials that can blend multiple crystallinities, thicknesses, and structural morphologies on one substrate (Figure 4F). Patterned seeding strategies are a potential path toward selective-area growth. Alternatively, selective-area transfer is promising for the integration of non-monolayer regions onto selected nodes on top of as-grown monolayer films, yielding crossbar geometries of alternating monolayer and few-layer devices. This scheme can adopt multiple functionalities by exploiting the interface tunability between monolayer and few-layer nodes without significant additional processing. Furthermore, the multi-terminal connectivity enabled by lateral or semi-vertical memtransistors provides plausible routes for the hyperconnectivity envisioned in bio-realistic networks and 3D-integrated chips. In the cases where polycrystalline domains and grain boundaries are important, lateral scaling will likely be limited to the grain size of polycrystalline MoS_2 monolayers.[17,18]

POSSIBILITIES

In light of the aforementioned scientific and engineering challenges, interdisciplinary collaborations will be vital to fully harness the potential of 2D materials in

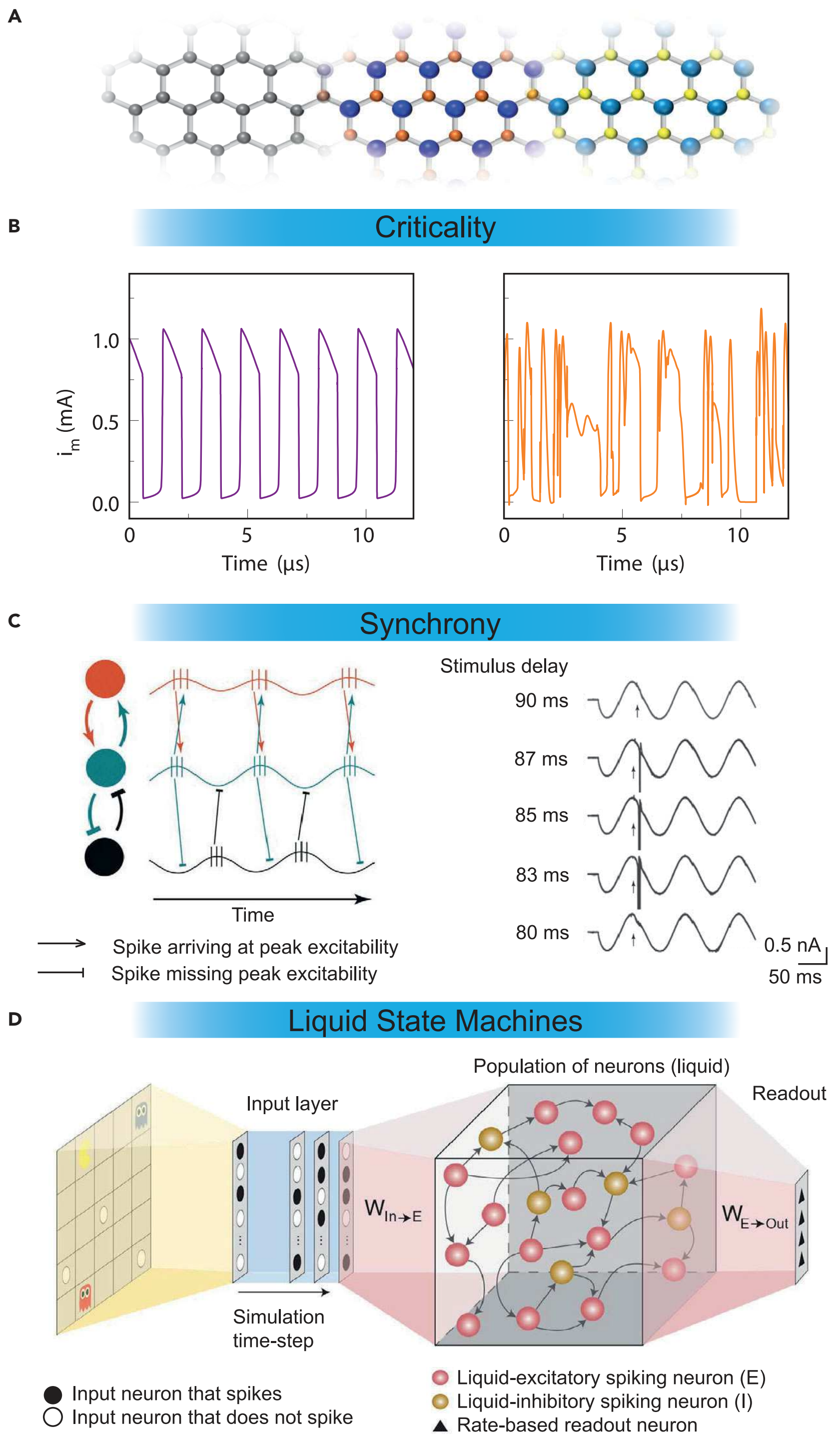

Figure 5. Opportunities for harnessing criticality and synchrony in 2D bio-realistic computing circuits

(A) Atomic-scale thickness, electronic tunability, and heterogenous integration of 2D materials are useful features to enable the bio-realistic phenomena of criticality and synchrony.

(B) The left panel illustrates regular periodic current levels in a memristor, and the right panel shows that coupling of a memristor with harmonic thermal fluctuations leads to chaotic behavior. Reproduced with permission.[90] Copyright 2017, Springer Nature.

Figure 5. *Continued*

(C) The left plot shows the neuronal groups (circles) and their action potentials (vertical lines). Red (sending) and green (receiving) neuronal groups undergo coherent excitability and transmit information (crest-crest envelope profiles), while the black and green groups do not (offset envelope shapes). The right plot shows the neuronal membrane potential upon stimulation at one afferent axon before, at, and after the neuronal excitability peak.[97] Copyright 2005, ScienceDirect.

(D) Schematic of reservoir computing, where the reservoir (liquid) is an intricate network of dynamically responsive neurons. Input signals can be mapped to higher dimensions based on neuron activation behaviors. Reproduced with permission.[92] Copyright 2019, Frontiers.

neuromorphic hardware (Figure 5A). Progress will likely require input from neuroscientists, materials scientists, and electrical and computer engineers to co-design bio-realistic paradigms. Here, we focus on two bio-realistic paradigms—criticality and synchrony—that are relatively underexplored in 2D neuromorphic devices and hold vast potential for bio-realistic hardware (Figures 5B and 5C). Criticality and synchrony are believed to contribute to the exceptional computing ability of the human brain.[11,89] The former describes the precarious state bordering order and disorder, termed the ''edge of chaos,'' at which biological neuronal networks are believed to maximize computing storage and capacity.[12] How a critical system is defined and quantified, and why the brain persists in and prefers criticality are not yet fully understood. Recently, chaotic behavior has been reported in a higher-order Mott memristor fabricated from NbO_2 films. In particular, when higher-order Mott memristors are coupled with an oscillator, harmonically decaying pulse amplitudes and frequencies are produced, which deviate from the regular oscillating behavior of a memristor alone (Figure 5B).[22,90] These tunable chaotic oscillations have been used to find global minima in a Hopfield network to solve complex problems (i.e., NP-hard computations with multiple local minima) with higher accuracy and efficiency than conventional neural networks.[90]

Recently, nanowire networks have proven to be another source of rich dynamical behavior that simulates the highly networked structure of the brain. Interconnected metal-insulator-metal device junctions in random nanowires have shown characteristics of critical systems including power-law spectral densities and high tunability and scaling as a function of order state variables with respect to nanowire diameter, length, and distribution.[91] The resulting nonlinear switching highlights the potential of nanowire networks for complex computing schemes, such as reservoir computing and liquid state machines (Figure 5D),[92] where the behavior of the intricate nanowire interconnectivity can enable higher-dimensional mapping of input signals.[93] In contrast, criticality has not yet been reported in 2D material devices, but their atomically thin, planar structure and weak interlayer bonding can provide unique advantages. For example, polycrystalline monolayer MoS_2 with a random network of grain boundaries has already shown non-local heterosynaptic responses, which could be explored for multi-terminal reservoir computing systems in addition to hypernetwork heterosynaptic learning.[18] Moreover, due to the chemical tunability of 2D materials through established CVD methods, the density, orientation, and distribution of defects, two-dimensional stacking faults, one-dimensional grain boundaries, and zero-dimensional point defects (e.g., substitutional impurities and vacancies) may be sufficiently well controlled to enable deterministic memristive responses. Recently, 2D transition metal halides and related quantum materials have shown novel magnetic ordering and related emergent behaviors, which suggest additional opportunities for criticality between ordered or disordered states as a function of temperature, voltage, layer thickness, and lattice angle in vdW heterojunctions.[94]

Similarly, neuronal synchrony could enable high-performance computing in critical systems.[95] Neuronal synchrony is characterized by the close temporal correlations of post-synaptic spiking responses of a homogenous network of neurons (Figure 5C).[96] While asynchronous spiking and event-driven computing are necessary for neuromorphic architectures to mimic the parallelized nature of the brain,[2] synchronous behavior among collective neurons can also enhance learning, memory capacity, and sensorimotor capability through sparse coding and detection of stimuli invariants.[96] In some cases, sparse coding and neuronal synchrony may even be more efficient, especially for a small group of homogenous neurons activating all at once compared with a larger proportion of neurons firing at disparate times. The cognitive parallel of invariant decoding is the Helmholtz principle of perception, which states that meaningful structures or stimuli invariants will be detected when their deviation from randomness is deemed significant.[11] In this manner, the ability for neurons to activate synchronously in response to stimuli invariants and then transmit signals synchronously to their post-synaptic neurons holds promise for computer vision, anomalous detection in media, vehicle automation, and reservoir computing systems. Hardware implementations of neuronal synchrony can be enabled by the higher-order, high-density, co-planar integration of 2D vdW heterostructures and memtransistors that present spiking neuron and heterosynaptic behavior, respectively. For example, stable homeostasis can likely be achieved by constructing a feedback loop with the co-integration of 2D artificial spiking neurons and synapses, where the post-synaptic output signal can be rerouted as an input signal into the gates of the artificial spiking neuron across multiple devices.

The fundamental versatility of 2D materials is a critical advantage to accelerating neuromorphic hardware development that has been challenging to emulate in 3D bulk material counterparts. For instance, dual-gating, heterogenous integration, and high-order dynamics that have been reported in individual 2D devices and crossbars provide unique synaptic and neuronal behavior. A detailed understanding of the physical mechanisms that underlie these unique responses will be facilitated by non-invasive characterization methods that can be performed under device operating conditions. Furthermore, innovative growth, fabrication, and transfer methods will accelerate efforts to scale down device geometries and achieve integration into sophisticated neuronal circuits and systems. Ultimately, by uniting the expertise of theoretical neuroscientists, materials scientists, and electrical and computer engineers, bio-realistic hardware co-design will enhance the functionality and performance of off-cloud data processing units and edge computing systems.

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DECLARATION OF INTERESTS

The authors declare no competing interests.

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