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Flexible and Stretchable Memristive Arrays for in-Memory Computing

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With the tremendous progress of Internet of Things (IoT) and artificial intelligence (AI) technologies, the demand for flexible and stretchable electronic systems is rapidly increasing. As the vital component of a system, existing computing units are usually rigid and brittle, which are incompatible with flexible and stretchable electronics. Emerging memristive devices with flexibility and stretchability as well as direct processing-in-memory ability are promising candidates to perform data computing in flexible and stretchable electronics. To execute the in-memory computing paradigm including digital and analogue computing, the array configuration of memristive devices is usually required. Herein, the recent progress on flexible and stretchable memristive arrays for in-memory computing is reviewed. The common materials used for flexible memristive arrays, including inorganic, organic and two-dimensional (2D) materials, will be highlighted, and effective strategies used for stretchable memristive arrays, including material innovation and structural design, will be discussed in detail. The current challenges and future perspectives of the in-memory computing utilizing flexible and stretchable memristive arrays are presented. These efforts aim to accelerate the development of flexible and stretchable memristive arrays for data computing in advanced intelligent systems, such as electronic skin, soft robotics, and wearable devices.

Keywords: memristor, in-memory computing, flexible and stretchable electronics, flexible memristive array, stretchable memristive array

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INTRODUCTION

The emergence of the Internet of Things (IoT) and artificial intelligence (AI) technology has promoted the rapid development of flexible and stretchable intelligent systems, such as electronic skin (e-skin) (Yang et al., 2019; Wang et al., 2020b), soft robotics (Wang et al., 2018b; Park et al., 2020b), and wearable devices (Liu et al., 2020). In the ideal scenarios, these systems should be able to undergo complex deformations in response to external stimuli, such as bending, twisting, and shearing (Wang et al., 2020c; Wang et al., 2021b; Raeis-Hosseini and Rho, 2021). Each component in the system is expected to be highly flexible and even stretchable to accommodate external mechanical strains. However, as a basic component of the intelligent system, existing computing units based on complementary metal-oxide-semiconductor (CMOS) technology are usually rigid and brittle, which are difficult to be integrated into flexible and stretchable systems (Gao et al., 2019).

Recently, emerging memristive devices provide a promising approach to achieve flexible and stretchable computing units, due to their simple structure (Ielmini, 2016; Li et al., 2020; Liu et al., 2012; Wang et al., 2020f), low energy consumption (Carlos et al., 2021; van de Burgt et al., 2018), high operating speed (Ren et al., 2020; Xu et al., 2019; Zhang et al., 2019b), and easy fabrication process

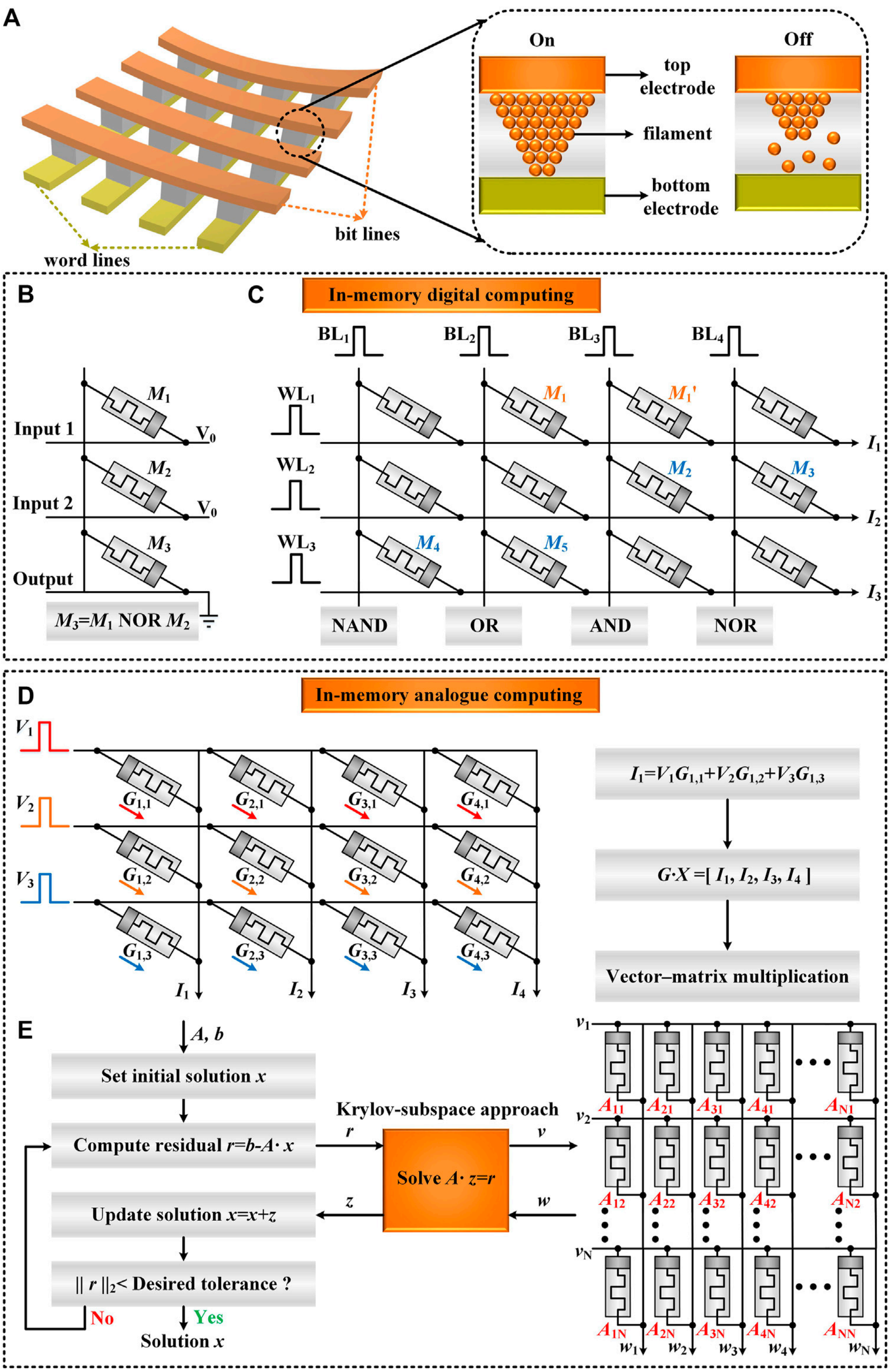

FIGURE 1 | In-memory computing using memristive array. **(A)** Schematic of the memristive array. The inset shows the memristive units with on and off states. b-c) Schematic of in-memory digital computing with memristive array, such as **(B)** the NOR operation, and **(C)** the parallel full adder. **(D)** The implementation of in-memory analogue computing with memristive array. **(E)** The schematic of solving linear formula with memristive crossbar arrays.

that is compatible with flexible and stretchable electronics (Wang et al., 2021a) (**Figure 1A**). Memristive devices are two-terminal electrical resistance switches whose resistance state can be tuned by the history of applied stimuli. The tuned resistance state can be configured to store data information for non-volatile memory, which has been intensively investigated for many years (Carlos et al., 2021; Yuan et al., 2021). In recent years, memristive devices are increasingly explored to execute data computing due to the direct processing-in-memory ability (Meng et al., 2021; Zhang et al., 2021; Zhou et al., 2021). The in-memory computing characteristics of memristive devices could break the bottleneck of traditional von Neumann computing architecture, resulting in high computational efficiency (Li et al., 2019; Xia and Yang, 2019).

A wide variety of semiconductor and dielectric materials such as inorganic (Yoon et al., 2018; Jo et al., 2021; Wang et al., 2021d), organic (van de Burgt et al., 2018; Park et al., 2020a), two-dimensional materials (Zhang et al., 2019a; Meng et al., 2021) and their derivatives (Gu and Lee, 2016) have been developed to obtain flexible and stretchable memristive devices and arrays. These devices and arrays are usually grown on flexible and stretchable substrates such as polyethylene terephthalate (PET) (Meng et al., 2021; Siddik et al., 2021), polyethylene naphthalate (PEN) (Zhang et al., 2019b), polyimide (PI) (Li et al., 2021) and polydimethylsiloxane (PDMS) (Hung et al., 2017; Rahman et al., 2018). Typical structural designs, such as wavy (Wang et al., 2020a) and island (Liu et al., 2017; Kim et al., 2021b) structures, have also been explored to endow memristive devices with mechanical flexibility and stretchability. However, most of the reported works on flexible and stretchable memristive devices focus on individual device units. For real applications, the array configuration of memristive units is typically required to execute specific computing functionality including digital and analogue in-memory computing (Jang et al., 2017; Cheng et al., 2019; Wang et al., 2020e; Wang et al., 2021a) Compared to intensively studied silicon-based counterparts, flexible and stretchable memristive crossbar arrays used to implement in-memory computing are still in their infancy, due to the relatively poor device performance and uncontrollable fabrication process (Shi et al., 2020; Gogoi et al., 2021). To further boost the development of flexible and stretchable memristive arrays, it is urgent to review the latest achievements in this area.

In this mini-review, we outline the recent progress in the actualization of in-memory computing based on flexible and stretchable memristive arrays. Firstly, the basic principle of in-memory computing based on the memristive array is briefly summarized. Afterward, the latest achievements in flexible memristive arrays including inorganic, organic and two-dimensional (2D) materials, and stretchable memristive arrays including material innovation and structural design are discussed. Furthermore, the critical challenges of flexible and stretchable memristive arrays for in-memory computing are identified, and future prospects are provided. We hope this mini-review would shed some light on the development of future flexible and stretchable intelligent systems.

COMPUTING PRINCIPLES OF IN-MEMORY COMPUTING

Benefitting from the unique processing-in-memory characteristics, memristive devices can be configured into specific arrays to implement Boolean logic and vector–matrix multiplication (VMM) operations, corresponding to in-memory digital computing and in-memory analogue computing, respectively. In the following section, the basic principles of in-memory digital and analogue computing using memristive arrays will be elaborated.

The memristive array can be used to implement basic digital logic operations, such as AND, OR, NOT, and NAND, by employing appropriate disposition (Hu et al., 2019; Jang et al., 2017; Sun et al., 2018). The resistance state of the memristive device participates in the computing process as a logic variable, and its on-state and off-state are defined as 0 and 1, respectively. **Figure 1B** shows a typical NOR implementation by using three memristive devices. Firstly, the output memristive device (M_3) is initialized to a low resistance state (LRS, logic 1), and then the voltage V_0 is applied to the input memristors M_1 and M_2. When devices M_1 and M_2 with the initial state of high resistance state (HRS, logic 0), the voltage across the device M_3 is not enough to reach its reset voltage, thus the device M_3 remains in the LRS (logic 1). For other input memristor states, applying voltage V will cause the output memristor voltage to reach its reset voltage, so the device M_3 changes to HRS (logic 0). Based on basic logic operations, the memristive array has been used to implement full adders (Huang et al., 2016; Hu et al., 2019; Zhang et al., 2021), showing great potential in energy-efficient electronics. Recently, Zhang et al. reported 2D conjugated PBDTT-BQTPA-based memristive devices to demonstrate all 16 Boolean logic operations and parallel 1-bit full adder circuit (Zhang et al., 2021). As shown in **Figure 1C**, the developed parallel 1-bit full adder circuit can be demonstrated with a 4 × 3 memristive crossbar array.

The core of in-memory analogue computing is using memristive array to implement the VMM operation based on the Ohm's law and Kirchhoff's current law (Ielmini and Wong, 2018). The computing principle is shown in **Figure 1D**. When a voltage vector is applied to the row line of the array, the current vector obtained on the column line is the product of the voltage vector and the conductance matrix of the memristive array. Thus, the VMM operation is successfully realized in a single operation cycle, in which each current value collected from one column represents one multiplication operation. It means multiple multiplication operations can be simultaneously achieved via multiple columns, indicating a highly parallel computing in memristive array. The in-memory analogue computing has been widely used for various applications such as linear system(Zidan et al., 2018), eigenvector solvers(Pedretti et al., 2021), and image processing (Li et al., 2018). For instance, the solution of eigenvectors (x) is actually an algebraic operation of VMM, which can be solved by power iteration (Pedretti et al., 2021). The parameters in a given matrix can be mapped to the

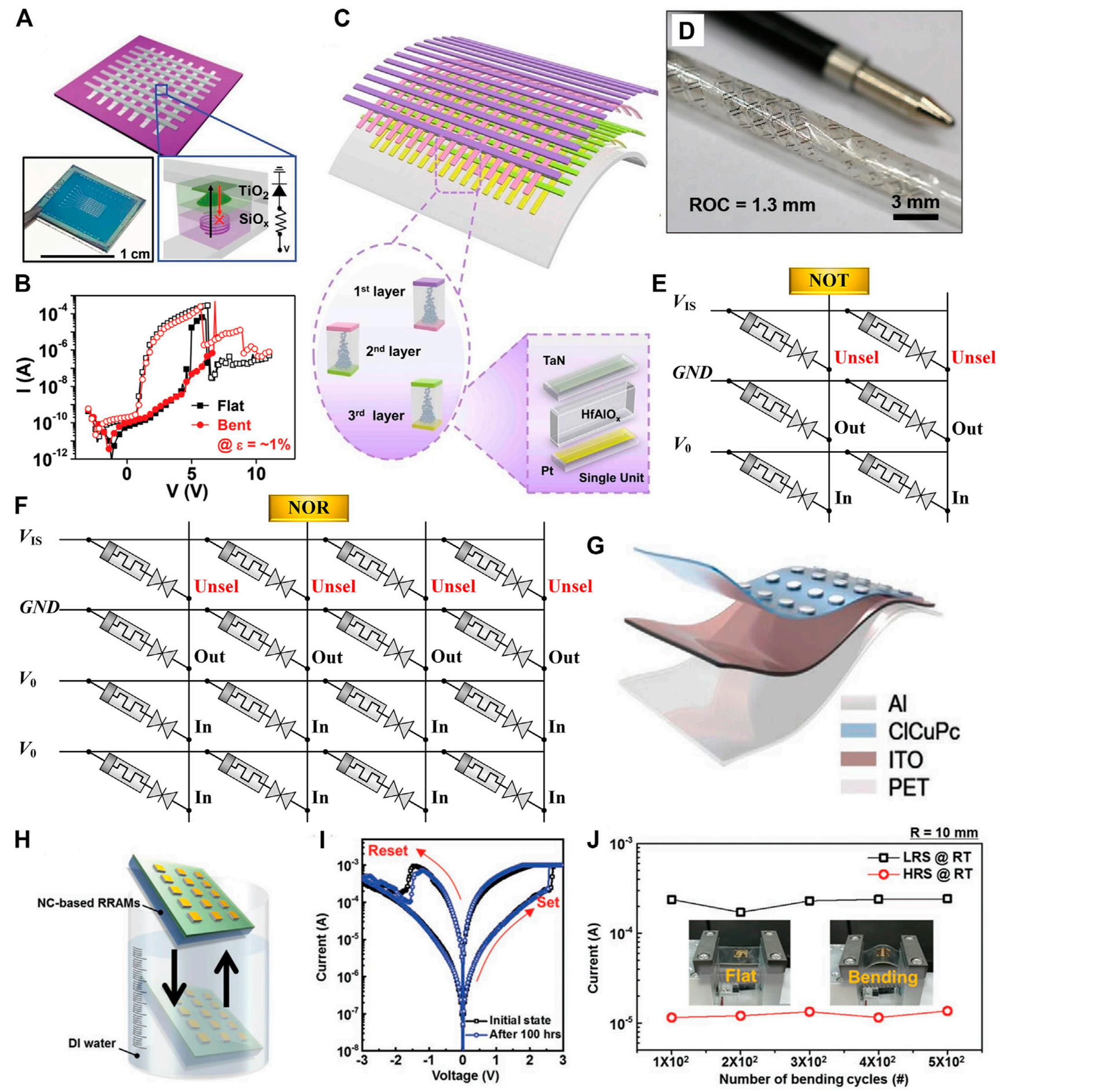

FIGURE 2 | Flexible inorganic and organic memristive arrays. **(A)** Schematic illustration of the 1D-1R memristive crossbar array (Yoon et al., 2018). **(B)** Current-voltage (*I–V*) characteristics of the 1D-1R memristive device under flat and bent states (Yoon et al., 2018). **(C)** The schematic of a 3D flexible memristive crossbar array with the Pt/$HfAlO_x$/TaN stack structure (Wang et al., 2021c). **(D)** Flexible memristive array rolled on a cylinder with a radius of curvature of 1.3 mm (Kook et al., 2020). e-f) The implementation of **(E)** NOT gate and **(F)** NOR within the 1S1M memristive array (Jang et al., 2018a). **(G)** The schematic of the flexible memristive array with an Al/ClCuPc/ITO/PET structure configuration (Zhou et al., 2021) **(H)**-**(I)** The schematic and *I–V* switching behaviors of nitrocellulose-based memristive devices before and after dipping in deionized water (Lee et al., 2020). **(J)** The flexible nitrocellulose-based memristive devices exhibited stable performance under the bending state (Lee et al., 2020). **(A, B)**. Reproduced with permission. Copyright 2018, Wiley-VCH. **(C)**. Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International License. **(D)**. Reproduced with permission. Copyright 2020, Wiley-VCH. **(E, F)**. Reproduced with permission. Copyright 2018, Wiley-VCH. **(G)**. Reproduced with permission. Copyright 2021, Wiley-VCH. **(H, J)**. Reproduced with permission. Copyright 2020, Wiley-VCH.

conductance of each device in the memristive network, the solution process can be performed by applying x_n to top electrode lines and obtaining the currents iteratively. Analogy with the principle of obtaining eigenvectors, the memristive crossbar array can also be used to solve linear formulas that are essential to computing. As shown in **Figure 1E**, the precise solution of a linear formula $A{\cdot}x = b$ is obtained by continuously updating the initial value x using the error correction term z (Le Gallo et al., 2018). The error correction term z is determined by the computing residual ($r = b{-}A{\cdot}x$) and the matrix A. By employing the Krylov-subspace approach, the term z can be achieved by solving equation $A{\cdot}z =$

r, where the matrix A is mapped to the conductance of memristive arrays. Consequently, the problem of the linear formula solution is converted to the analogue VMM computing in memristive arrays.

FLEXIBLE MEMRISTIVE ARRAYS FOR IN-MEMORY COMPUTING

With the development of materials and fabrication process, a variety of materials including inorganics, organics and 2D materials have been developed to obtain flexible memristive arrays for in-memory computing. The following section will discuss the recent progress of flexible memristive arrays and identify the critical challenges in the actualization of data computing.

Flexible Inorganic Memristive Arrays

The most straightforward approach to obtain flexible memristive arrays is directly depositing inorganic materials onto flexible substrates via thin-film fabrication processes, such as evaporation and sputtering. Until now, a large number of inorganic materials, such as AlO_x (Lee et al., 2017; Jang et al., 2018b), SiO_x (Yoon et al., 2018), WO_x (Lin et al., 2018), and $HfAlO_x$ (Wang et al., 2020d; Wang et al., 2021c), have been fabricated as flexible resistive switching dielectrics by using this approach. For instance, J. Yoon et al. reported a 8 × 8 flexible SiO_x-based one diode–one resistor (1D-1R) memristive crossbar array on a plastic substrate by employing physical vapor deposition (PVD) technology (Yoon et al., 2018). The schematic of the SiO_x-based 1D-1R array is shown in **Figure 2A**. The fabricated 1D-1R devices exhibited robust electrical characteristics under the bending state with a radius of 8 mm, indicating an excellent mechanical flexibility (**Figure 2B**).

Most inorganic materials require high-temperature processes to guarantee the nucleation of particles and the quality of grown thin film. However, the extensively used flexible substrates only show low temperature tolerance (Wang et al., 2021c; Wang et al., 2021d), which limits the choice of inorganic materials. Hence, it is vital to develop low-temperature manufactural processes to prepare flexible memristive arrays for in-memory computing. Recently, Zhang's group reported a series of studies on the use of low-temperature atomic layer deposition (ALD) technology to fabricate flexible memristive devices (Wang et al., 2021c; Wang et al., 2019; Wang et al., 2020d). This approach not only provides an atomically dense resistive switching layer with a precise control of thickness, but also is compatible with flexible fabrication process. Based on this method, a three dimensional (3D) flexible memristive crossbar array with the platinum (Pt)/$HfAlO_x$/tantalum mononitride (TaN) stack structure was fabricated on a PET substrate at 130°C (**Figure 2C**) (Wang et al., 2021c). Typical forms of synaptic plasticity, such as long-term potentiation (LTP), long-term depression (LTD), and paired pulse facilitation (PPF), have been successfully demonstrated in the electronic synapses. In addition, the memristive devices in the flexible 3D array could also maintain reliable LTP and LTD behaviors under the bending state with the bending radius of 10 mm. These developed flexible memristive arrays offer a promising way to implement the wearable in-memory computing system.

Flexible Organic Memristive Arrays

Organic materials often show better flexibility than inorganic materials, which are potential candidates for fabricating flexible memristive devices and arrays. Common organic materials, such as albumin (Chen et al., 2015), collagen (Zeng et al., 2019), silk fibroin (Kook et al., 2020; Wang et al., 2016a), polyethyleneimine (PEI) (Yang et al., 2020) and poly(1,3,5-trivinyl-1,3,5-trimethylcyclotrisiloxane) (pV_3D_3) (Jang et al., 2019; Jang J. et al., 2018a), have demonstrated their promising application in flexible memristive devices and arrays. By employing wafer-scale ultraviolet photolithography technology, a silk fibroin-based memristive array was successfully fabricated on a parylene-C film with robust flexibility (Kook et al., 2020). The fabricated memristive crossbar array shows robust flexibility (bendable to a radius of 1.3 mm) and reliable electrical performance (**Figure 2D**). With the utilization of initiated chemical vapor deposition (iCVD) approach, Jang et al. demonstrated a flexible memristive crossbar array with a copper (Cu)/pV_3D_3/aluminum (Al) structure for in-memory logic computing (Jang et al., 2018a). Each memristive unit in the array is connected to one selector to construct the one-selector–one-memristor (1S1M) architecture to alleviate the leakage current of the array. Typical logic operations such as NOT and NOR, were successfully implemented in the flexible pV_3D_3-based memristive crossbar array (**Figures 2E,F**).

Although organic materials often show excellent flexibility, the chemical stability of most organic materials is usually poor. The inferior chemical stability hinders the practical application of flexible organic memristive devices and arrays, e.g. medical equipment typically need to withstand a high temperature of 121°C during steam sterilization (Kuribara et al., 2012). In addition, the implementation of large-scale memristive arrays leads to inevitable heat dissipation, which further affects the reliability and stability of organic materials. Among various organic materials, copper phthalocyanines have been intensively explored due to their reliable chemical and thermal stability (Choi et al., 2008; Lv et al., 2019; Wang et al., 2016b; Wang et al., 2017; Zhou et al., 2021). Recently, as shown in **Figures 2G,J**. Zhou et al. reported a highly chemically and thermally stable flexible memristive array by using monochloro copper phthalocyanine (ClCuPc) materials (Zhou et al., 2021). Benefitting from the intrinsic high thermal stability of CuPc and the further improvement of air stability caused by chlorination, the fabricated memristive device could exhibit reliable resistive switching behavior at 300°C. Typical synaptic behaviors, such as PPF, paired-pulse depression (PPD), and spike-rate-dependent plasticity (SRDP), have been implemented in ClCuPc-based memristive devices. Furthermore, a 7 × 7 memristive crossbar array was constructed based on the ClCuPc

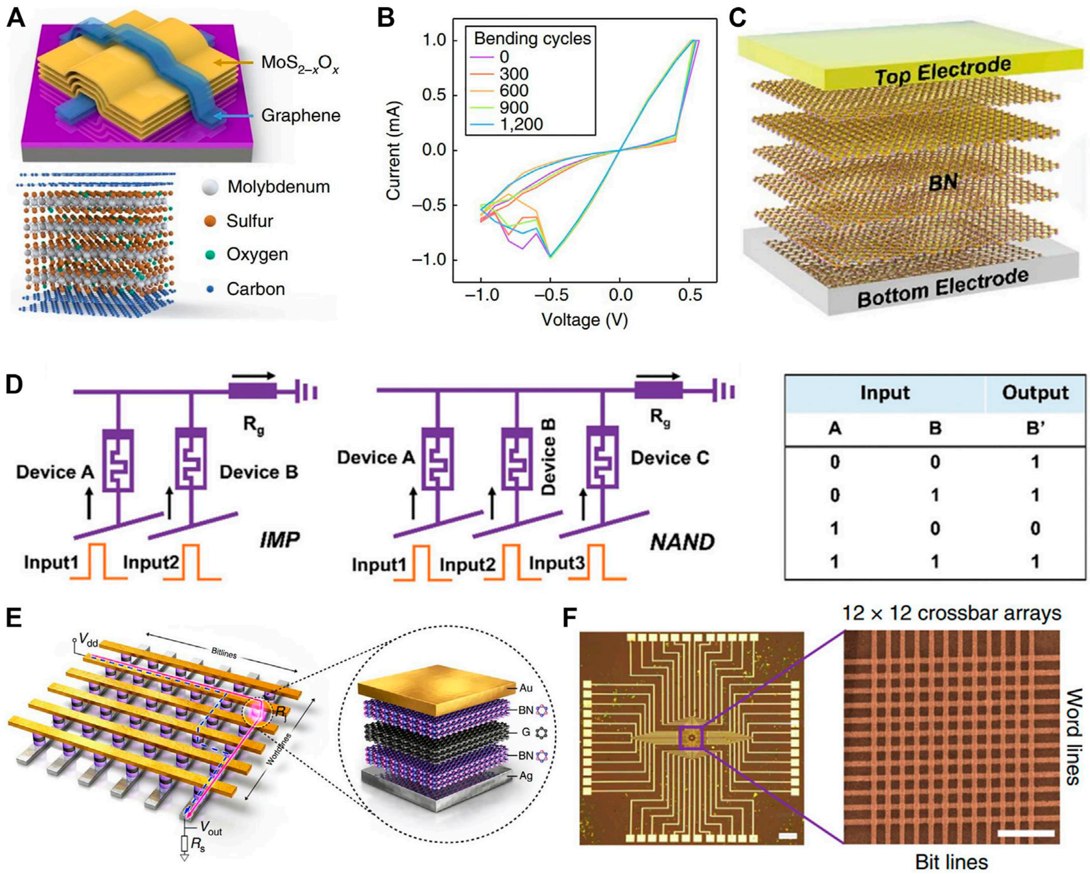

FIGURE 3 | Flexible two-dimensional material memristive arrays. **(A)** The schematic of graphene/$MoS_{2-x}O_x$/graphene-based device (Wang et al., 2018a). **(B)** The performance of the fabricated memristive device after 1,200 bending cycles at a radius of curvature of 1 cm (Wang et al., 2018a). **(C)** The schematic of BN-based memristive device (Meng et al., 2021). **(D)** The realization of in-memory digital computing with BN-based memristiors (Meng et al., 2021). **(E)** The schematic of a memristive crossbar array with a structure of Ag/h-BN/graphene/h-BN/Au (Sun et al., 2019). **(F)** The fabricated self-selective 12 × 12 memristive array (Sun et al., 2019). **(A, B)**. Reproduced with permission. Copyright 2018, Springer Nature. **(C, D)**. Reproduced with permission. Copyright 2021, Royal Society of Chemistry. **(E, F)**. Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International License. **(A, B)**. Reproduced with permission. Copyright 2018, Springer Nature. **(C, D)**. Reproduced with permission. Copyright 2021, Royal Society of Chemistry. **(E, F)**. Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International License.

memristive synapses, which was used to implement in-memory computing for game Tetris.

Apart from weak thermal stability, most organic materials are vulnerable to moisture, making organic-based memristive devices and arrays hard to maintain stable performance in a humid environment (Lee et al., 2020; Yuan et al., 2021). For instance, commonly reported organic memristive devices based on chitosan (Hosseini and Lee, 2015), pectin (Xu et al., 2019), perovskite materials (Hwang and Lee, 2017), and glucose (Park et al., 2018) usually exhibit poor water-resistance, and even water soluble. Hence, it is necessary to develop water-resistance organic flexible memristive devices and arrays for harsh environments (Lee et al., 2015). Lee et al. recently reported a flexible nitrocellulose-based memristive array with high water-resistance and mechanical flexibility (Lee et al., 2020). The fabricated memristive devices could exhibit reliable electrical switching behaviors after submersion in phosphate-buffered saline solution, artificial perspiration, and deionized water (**Figures 2H,I**). Moreover, the flexible memristive devices showed stable operations under the bending state with a radius of 10 mm (**Figure 2J**).

Flexible Two-Dimensional Material Memristive Arrays

In addition to inorganic and organic materials, 2D materials have recently received extensive attention due to their unique structural characteristics, good mechanical flexibility, and excellent electrical properties (Kim et al., 2021a; Meng et al., 2021). Plenty of 2D materials such as graphene (Sun et al., 2017), transition metal dichalcogenides (TMD) (Feng et al., 2019; Zhao et al., 2018) and boron nitride (BN) (Ge et al., 2021; Qian et al., 2017; Siddiqui et al., 2017) have been well studied for flexible memristive arrays. For example, Wang et al. reported a flexible memristive array with the graphene/$MoS_{2-x}O_x$/graphene structure, which has good mechanical

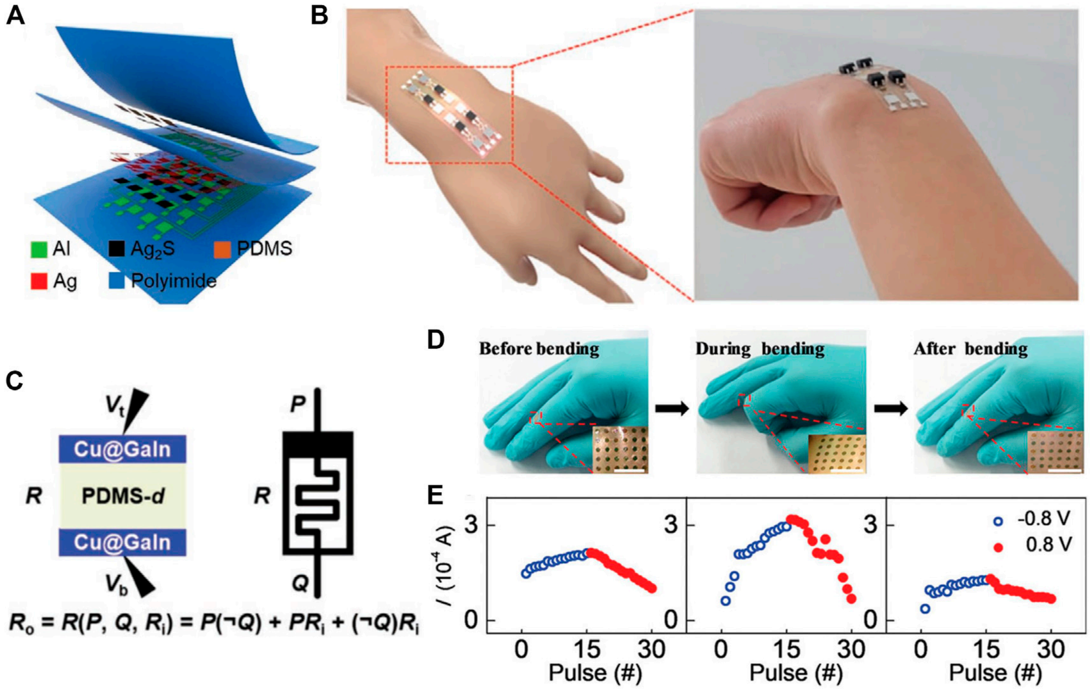

FIGURE 4 | Stretchable memristive array with materials innovation for in-memory computing. **(A)** The schematic of the PI/Ag/Ag_2S/Al/PI-structured stretchable memristive array (Jo et al., 2021). **(B)** The wearable healthcare-monitoring device integrated with Ag_2S-based stretchable memristive devices (Jo et al., 2021). **(C)** The implementation of the MIG logic with the memristive device (Lu et al., 2021). **(D)** The stretchable memristive crossbar array with a resistive switching layer of composite material (TPU: Ag NPs) under stretching strain (Yang et al., 2018). **(E)** Synaptic potentiation and depression characteristics under stretching strain (Yang et al., 2018). **(A, B)**. Reproduced with permission. Copyright 2021, Wiley-VCH. **(C)**. Reproduced with permission. Copyright 2021, Wiley-VCH. **(D, E)**. Reproduced with permission. Copyright 2018, Royal Society of Chemistry.

property and fabulous thermal stability (**Figure 3A**) (Wang et al., 2018a). The fabricated memristive device could exhibit reliable electrical performance after 1,200 bending cycles at a radius of curvature of 1 cm (**Figure 3B**). Recently, a flexible BN-based memristive array for in-memory computing was developed (**Figure 3C**) (Meng et al., 2021). The fabricated flexible BN-based memristive devices realized both in-memory digital and analogue computing, breaking the limitation caused by the difference in information form between digital data and analog data. The flexible memristive devices could implement FALSE,material implication (IMP) and NAND operations for in-memory digital computing (**Figure 3D**). In addition, the flexible memristive devices could also be used to realize the synaptic plasticity such as PPF, LTP/LTD and spike-timing-dependent plasticity (STDP) for in-memory analogue computing. Sneak current in the array is one of the primary issues limiting the high-density integration of memristive crossbar arrays. To solve this issue, Sun et al. developed a flexible self-selective memristive device based on a van der Waals heterostructure with the hexagonal boron nitride (h-BN)/graphene/h-BN structure (**Figure 3E**) (Sun et al., 2019). The middle graphene layer enables the volatile and non-volatile resistive behaviors in the same device, resulting in the self-selective effect. Based on the self-selective devices, a 12 × 12 memristive crossbar array was built, showing a promising application on future data storage in wearable artificial intelligence (**Figure 3F**).

STRETCHABLE MEMRISTIVE ARRAYS FOR IN-MEMORY COMPUTING

Apart from flexibility, the memristive array needs to be stretchable in scenarios such as e-skin and wearable devices. Developing stretchable electronics is highly contingent on the materials innovation and structural design.

Materials Innovation for Stretchable Memristive Arrays

Developing novel materials with ideal electrical and mechanical properties is an effective way to obtain stretchable memristive arrays. Recently, a stretchable $SrTiO_{3-x}$-based memristive crossbar array was successfully fabricated on a PDMS substrate via thin film deposition process (Rahman et al., 2018). In addition, a stretchable Ag_2S thin film-based memristive array has also been developed (**Figure 4A**) (Jo et al., 2021). The solution-processed Ag_2S film demonstrated excellent mechanical properties and could withstand a strain of

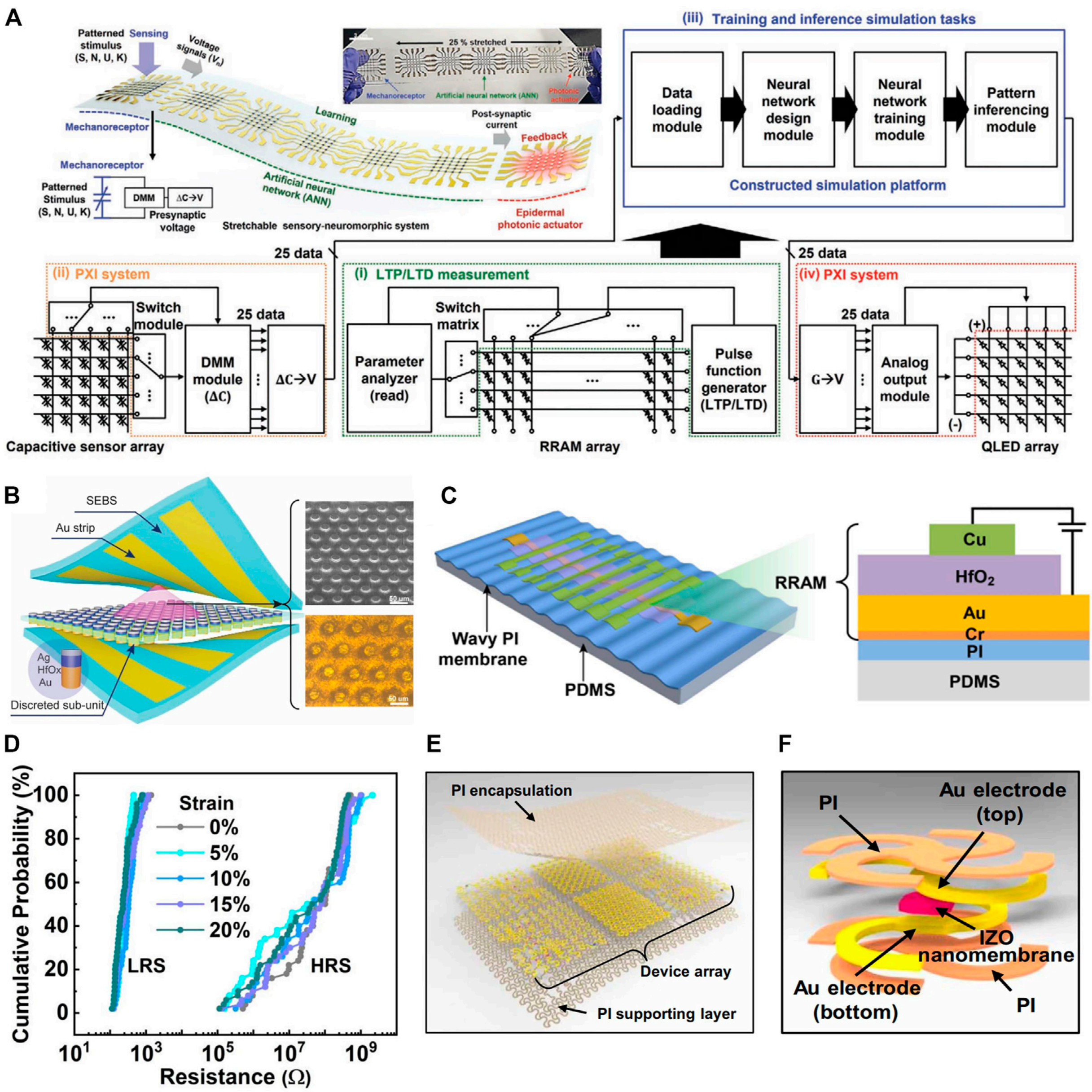

FIGURE 5 | Stretchable memristive array with structural design for in-memory computing. **(A)** Schematic of the in-memory computing system integrated with memristive crossbar array via the island-structure strategy (Kim et al., 2021b). **(B)** Schematic of the memristive crossbar array with a discrete structure (Wang et al., 2021b). **(C)** Schematic of the HfO_2-based memristive array constructed on the PDMS substrate with a wavy structure (Wang et al., 2020a). **(D)** The resistive switching behavior of the HfO_2-based memristor under the strain of 0–20% (Wang et al., 2020a). **(E)** Schematic of the wearable human-machine interfaces integrated with a IZO nanomembrane-based memristive array (Sim et al., 2019). **(F)** The diagram of the serpentine-structured memristor (Sim et al., 2019). **(A)**. Reproduced with permission. Copyright 2021, Wiley-VCH. **(B)**. Reproduced with permission. Copyright 2021, Wiley-VCH. **(C, D)**. Reproduced with permission. Copyright 2020, IEEE. **(E, F)**. Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International License.

14.9%. Based on the stretchable memristive devices, a wearable self-powered healthcare-monitoring system was successfully constructed (**Figure 4B**).

In contrast to inorganic materials, organic materials have inherent advantages in terms of lighter weight and better stretchability, thus they are more appropriate for realizing stretchable electronics (Fu et al., 2019; Yuan et al., 2021). Organic-based stretchable memristive devices such as PDMS/carbon nanotubes (CNTs)/maltoheptaose-block-polyisoprene (MH-b-PI)/Al (Hung et al., 2017) and Cu@GaIn/PDMS/Cu@GaIn (Lu et al., 2021) have been extensively studied. For instance, Lu et al. successfully demonstrated stretchable and twistable PDMS-based memristive devices for in-memory digital computing (Lu et al., 2021) (**Figure 4C**). On the basis of majority-inverter graph (MIG) logic, the fabricated stretchable memristive devices realized a one-bit full adder for digital computing.

Despite great progress has been made in organic materials, stretchable organic-based devices cannot offer reliable resistive switching properties. In the light of this, researchers attempt to exlpore novel materials that have both the good electrical characteristic of inorganic substances and the excellent stretchability of organic substances (Yang et al., 2018; Yi et al., 2019). Recently, by employing Ag nanoparticle-doped thermoplastic polyurethanes (TPU: Ag NPs) as the resistance switching material, Yang et al. demonstrated a stretchable memristive crossbar array constructed on a PDMS substrate for in-memory computing (Yang et al., 2018). The stretchable memory cell of the fabricated array implemented synaptic plasticity under stretching strain (**Figures 4D,E**).

Structural Design for Stretchable Memristive Arrays

Besides material innovation, designing a specific structure to withstand applied strain is another powerful approach for stretchable memristive arrays. Over the years, researchers have developed various effective structures, such as wave, island and serpentine-shaped, to enhance the stretchability of the system. Using an island-structure strategy, a stretchable in-memory computing system was developed by embedding rigid TiO_2-based memristive units into soft interconnects (Kim et al., 2021b) (**Figure 5A**). Benefitting from this structure, the fabricated in-memory computing system could exhibit reliable electrical performance under a strain of 25%. The TiO_2-based memristive array in the system was used to implement artificial neural network for learning and inferencing. A stretchable HfO_2-based memristive array with excellent damage endurance has also been demonstrated by using the island structure (Wang et al., 2021b). Moreover, each memristive unit in the stretchable array is also consisted of many discrete HfO_2-based sub-units (**Figure 5B**). This discrete structure enables the stretchable memristive device to have a large stretchability of 40% strain and good mechanical damage endurance.

Apart from the island-structure, the wavy-structure has also been developed to design stretchable memristive devices. Based on this strategy, stretchable HfO_2-based memristive devices were recently developed by directly laminating the $Cu/HfO_2/Au/PI$ film onto a pre-stretching PDMS substrate (Wang et al., 2020a). After releasing the pre-stretching substrate, wavy-structured memristive devices were formed (**Figure 5C**). The structure could significantly improve the stretchability of HfO_2-based memristive devices, exhibiting reliable switching characteristics up to 20% strain (**Figure 5D**).

Moreover, the mechanical flexibility of the system can also be effectively improved by configuring the system as a serpentine structure. Based on the serpentine-shaped configuration, a stretchable wearable device integrated with indium zinc oxide (IZO) nanomembrane-based memristive array was developed, where the Au electrode was made into a serpentine shape (**Figure 5E**) (Sim et al., 2019). Owing to the serpentine-shaped metal electrode (**Figure 5F**), the memory cell can be isolated from the strain under mechanical contortion, so that the system could exhibit stable electrical performance under 30% stretching strain. These structural designs can significantly enhance the mechanical flexibility of the rigid memristive array, providing a promising way to extend current stretchable memristive devices and arrays.

PERSPECTIVES

In the past few years, emerging manufacturing processes, material innovation and structural design have brought about the rapid progress of flexible and stretchable memristive crossbar arrays, providing opportunities for the realization of high-speed and energy-efficient information processing in e-skins, soft robotics, and wearable and implantable systems. Despite great progress has been made on flexible and stretchable memristive devices in recent years, highly reliable memristive arrays that could be used to implement in-memory computing are still challenging. We will briefly highlight several critical challenges in the following paragraphs.

Device reliability Compared to the traditional silicon-based devices, the main drawbacks of flexible and stretchable memristive arrays are poor performance and uncontrollable manufacturing processes. Most of the research focuses on the manufacture of memristive device prototypes, especially memristive devices based on organic and two-dimensional materials, while ignoring the optimization of device performance. The unreliability of memristive devices significantly deteriorates the computational accuracy of in-memory computing. It is imperative to explore new memristive devices with reliable resistive switching behaviors and good mechanical flexibility.

Array integration The realization of flexible and stretchable in-memory computing system relies on the configuration of the memristive crossbar array. Similar to the case of silicon-based memristive array, the notorious sneak current effect in the passive array limits the large-scale integration of flexible and stretchable memristive array. The common transistor selection devices used to solve the sneak current in the silicon-based memristive array do not meet the requirement of flexible and stretchable electronics. It is urgent to develop highly flexible and stretchable transistors or other new selection devices to enable large-scale array integration. In addition, the large-scale memristive crossbar array will inevitably produce parasitic resistance and capacitance, due to the conductivity of the materials and the parallel plate capacitor effect. These parasitic effects of the array directly affect the accuracy and operating speed of the in-memory computing. It is essential to optimize the architecture of the flexible and stretchable memristive arrays to alleviate the parasitic effects.

System integration Apart from the core memristive crossbar array, a fully flexible and stretchable in-memory computing system requires complex peripheral circuits that control the input/output of the array and sequential operations of the system. Although there has been great progress on flexible chips using advanced flexible fabrication process, the functionality and reliability of current flexible circuits are still insufficient. Recently, a flexible hybrid integration strategy has been proposed, which makes an attempt to combine conventional silicon-based circuits and flexible/stretchable components into one system. Based on this strategy, highly reliable and powerful silicon-based peripheral circuits could be integrated into a flexible and stretchable system to control the memristive crossbar array for information processing.

SUMMARY

With the popularization of information technology, data computing in flexible and stretchable intelligent systems is increasingly important. The implementation of in-memory computing based on memristive arrays provides a high-speed and energy-efficient approach to execute data computing for flexible and stretchable systems. In this review, we summarized the recent progress on flexible and stretchable memristive arrays, focusing on resistive switching materials, structure configuration, and computational implementation. The current challenges and future prospects of flexible and stretchable memristive arrays for in-memory computing are further discussed. Finally, we hope that the flexible and stretchable memristive array can efficiently implement processing-in-memory in emerging flexible intelligent electronics.

AUTHOR CONTRIBUTIONS

MW designed this study. XL wrote the manuscript. MW and QL supervised this work. All authors contributed to discussions and comments on the manuscript.

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REFERENCES

Carlos, E., Branquinho, R., Martins, R., Kiazadeh, A., and Fortunato, E. (2021). Recent Progress in Solution-Based Metal Oxide Resistive Switching Devices. *Adv. Mater.* 33, 2004328. doi:10.1002/adma.202004328

Chen, Y.-C., Yu, H.-C., Huang, C.-Y., Chung, W.-L., Wu, S.-L., and Su, Y.-K. (2015). Nonvolatile Bio-Memristor Fabricated with Egg Albumen Film. *Sci. Rep.* 5, 10022. doi:10.1038/srep10022

Cheng, L., Li, Y., Yin, K. S., Hu, S. Y., Su, Y. T., Jin, M. M., et al. (2019). Functional Demonstration of a Memristive Arithmetic Logic Unit (MemALU) for In-Memory Computing. *Adv. Funct. Mater.* 29, 1905660. doi:10.1002/adfm.201905660

Choi, S., Hong, S.-H., Cho, S. H., Park, S., Park, S.-M., Kim, O., et al. (2008). High-performance Programmable Memory Devices Based on Hyperbranched Copper Phthalocyanine Polymer Thin Films. *Adv. Mater.* 20, 1766–1771. doi:10.1002/adma.200702147

Feng, X., Li, Y., Wang, L., Chen, S., Yu, Z. G., Tan, W. C., et al. (2019). A Fully Printed Flexible MoS_2 Memristive Artificial Synapse with Femtojoule Switching Energy. *Adv. Electron. Mater.* 5, 1900740. doi:10.1002/aelm.201900740

Fu, L., Zhang, Y., Ren, Z., Li, H., Sun, X., and Yan, S. (2019). Flexible and Fatigue-Resistant Ternary Electrical Memory Based on Alternative Copolysiloxane with Carbazole Donors and Imidazole-Modified Naphthalimide Acceptors. *Adv. Mater. Technol.* 4, 1900084. doi:10.1002/admt.201900084

Gao, S., Yi, X., Shang, J., Liu, G., and Li, R.-W. (2019). Organic and Hybrid Resistive Switching Materials and Devices. *Chem. Soc. Rev.* 48, 1531–1565. doi:10.1039/C8CS00614H

Ge, J., Huang, H., Ma, Z., Chen, W., Cao, X., Fang, H., et al. (2021). A Sub-500 mV Monolayer Hexagonal boron Nitride Based Memory Device. *Mater. Des.* 198, 109366. doi:10.1016/j.matdes.2020.109366

Gogoi, H. J., Bajpai, K., Mallajosyula, A. T., and Solanki, A. (2021). Advances in Flexible Memristors with Hybrid Perovskites. *J. Phys. Chem. Lett.* 12, 8798–8825. doi:10.1021/acs.jpclett.1c02105

Gu, C., and Lee, J.-S. (2016). Flexible Hybrid Organic-Inorganic Perovskite Memory. *ACS Nano* 10, 5413–5418. doi:10.1021/acsnano.6b01643

Hosseini, N. R., and Lee, J.-S. (2015). Biocompatible and Flexible Chitosan-Based Resistive Switching Memory with Magnesium Electrodes. *Adv. Funct. Mater.* 25, 5586–5592. doi:10.1002/adfm.201502592

Hu, S.-Y., Li, Y., Cheng, L., Wang, Z.-R., Chang, T.-C., Sze, S. M., et al. (2019). Reconfigurable Boolean Logic in Memristive Crossbar: the Principle and Implementation. *IEEE Electron. Device Lett.* 40, 200–203. doi:10.1109/LED.2018.2886364

Huang, P., Kang, J., Zhao, Y., Chen, S., Han, R., Zhou, Z., et al. (2016). Reconfigurable Nonvolatile Logic Operations in Resistance Switching Crossbar Array for Large-Scale Circuits. *Adv. Mater.* 28, 9758–9764. doi:10.1002/adma.201602418

Hung, C.-C., Chiu, Y.-C., Wu, H.-C., Lu, C., Bouilhac, C., Otsuka, I., et al. (2017). Conception of Stretchable Resistive Memory Devices Based on Nanostructure-Controlled Carbohydrate-Block-Polyisoprene Block Copolymers. *Adv. Funct. Mater.* 27, 1606161. doi:10.1002/adfm.201606161

Hwang, B., and Lee, J.-S. (2017). Hybrid Organic-Inorganic Perovskite Memory with Long-Term Stability in Air. *Sci. Rep.* 7, 673. doi:10.1038/s41598-017-00778-5

Ielmini, D. (2016). Resistive Switching Memories Based on Metal Oxides: Mechanisms, Reliability and Scaling. *Semicond. Sci. Technol.* 31, 063002. doi:10.1088/0268-1242/31/6/063002

Ielmini, D., and Wong, H.-S. P. (2018). In-memory Computing with Resistive Switching Devices. *Nat. Electron.* 1, 333–343. doi:10.1038/s41928-018-0092-2

Jang, B. C., Kim, S., Yang, S. Y., Park, J., Cha, J.-H., Oh, J., et al. (2019). Polymer Analog Memristive Synapse with Atomic-Scale Conductive Filament for Flexible Neuromorphic Computing System. *Nano Lett.* 19, 839–849. doi:10.1021/acs.nanolett.8b04023

Jang, B. C., Nam, Y., Koo, B. J., Choi, J., Im, S. G., Park, S.-H. K., et al. (2018a). Memristive Logic-In-Memory Integrated Circuits for Energy-Efficient Flexible Electronics. *Adv. Funct. Mater.* 28, 1704725. doi:10.1002/adfm.201704725

Jang, B. C., Yang, S. Y., Seong, H., Kim, S. K., Choi, J., Im, S. G., et al. (2017). Zero-static-power Nonvolatile Logic-In-Memory Circuits for Flexible Electronics. *Nano Res.* 10, 2459–2470. doi:10.1007/s12274-017-1449-y

Jang, J., Choi, H. H., Paik, S. H., Kim, J. K., Chung, S., and Park, J. H. (2018b). Highly Improved Switching Properties in Flexible Aluminum Oxide Resistive Memories Based on a Multilayer Device Structure. *Adv. Electron. Mater.* 4, 1800355. doi:10.1002/aelm.201800355

Jo, S., Cho, S., Yang, U. J., Hwang, G. S., Baek, S., Kim, S. H., et al. (2021). Solution-Processed Stretchable Ag_2S Semiconductor Thin Films for Wearable Self-Powered Nonvolatile Memory. *Adv. Mater.* 33, 2100066. doi:10.1002/adma.202100066

Kim, D., Kim, I.-J., and Lee, J.-S. (2021a). Memory Devices for Flexible and Neuromorphic Device Applications. *Adv. Intell. Syst.* 3, 2000206. doi:10.1002/aisy.202000206

Kim, S. H., Baek, G. W., Yoon, J., Seo, S., Park, J., Hahm, D., et al. (2021b). A Bioinspired Stretchable Sensory-Neuromorphic System. *Adv. Mater.* 33, 2104690. doi:10.1002/adma.202104690

Kook, G., Jeong, S., Kim, M. K., Lee, S., Choi, N., and Lee, H. J. (2020). Fabrication of Highly Dense Silk Fibroin Biomemristor Array and its Resistive Switching Characteristics. *Adv. Mater. Technol.* 5, 1900991. doi:10.1002/admt.201900991

Kuribara, K., Wang, H., Uchiyama, N., Fukuda, K., Yokota, T., Zschieschang, U., et al. (2012). Organic Transistors with High thermal Stability for Medical Applications. *Nat. Commun.* 3, 723. doi:10.1038/ncomms1721

Le Gallo, M., Sebastian, A., Mathis, R., Manica, M., Giefers, H., Tuma, T., et al. (2018). Mixed-precision In-Memory Computing. *Nat. Electron.* 1, 246–253. doi:10.1038/s41928-018-0054-8

Lee, B.-H., Bae, H., Seong, H., Lee, D.-I., Park, H., Choi, Y. J., et al. (2015). Direct Observation of a Carbon Filament in Water-Resistant Organic Memory. *ACS Nano* 9, 7306–7313. doi:10.1021/acsnano.5b02199

Lee, J. H., Park, S. P., Park, K., and Kim, H. J. (2020). Flexible and Waterproof Resistive Random-Access Memory Based on Nitrocellulose for Skin-Attachable Wearable Devices. *Adv. Funct. Mater.* 30, 1907437. doi:10.1002/adfm.201907437

Lee, W., Jang, J., Song, Y., Cho, K., Yoo, D., Kim, Y., et al. (2017). Attachable and Flexible Aluminum Oxide Resistive Non-volatile Memory Arrays Fabricated on Tape as the Substrate. *Nanotechnology* 28, 135201. doi:10.1088/1361-6528/aa5f0d

Li, B., Hui, W., Ran, X., Xia, Y., Xia, F., Chao, L., et al. (2019). Metal Halide Perovskites for Resistive Switching Memory Devices and Artificial Synapses. *J. Mater. Chem. C* 7, 7476–7493. doi:10.1039/C9TC02233C

Li, C., Hu, M., Li, Y., Jiang, H., Ge, N., Montgomery, E., et al. (2018). Analogue Signal and Image Processing with Large Memristor Crossbars. *Nat. Electron.* 1, 52–59. doi:10.1038/s41928-017-0002-z

Li, C. S., Kuo, S. W., Wu, Y. T., Fu, F. Y., Ni, I. C., Chen, M. H., et al. (2020). Forming-Free, Nonvolatile, and Flexible Resistive Random-Access Memory Using Bismuth Iodide/van der Waals Materials Heterostructures. *Adv. Mater. Inter.* 7, 2001146. doi:10.1002/admi.202001146

Li, J., Hao, C., Guo, S., Li, Y., Ren, J., Zhou, L., et al. (2021). Flexible Ta/TiO_X/TaO_X/Ru Memristive Synaptic Devices on Polyimide Substrates. *Nanotechnology* 32, 335205. doi:10.1088/1361-6528/ac00e0

Lin, Y., Zeng, T., Xu, H., Wang, Z., Zhao, X., Liu, W., et al. (2018). Transferable and Flexible Artificial Memristive Synapse Based on WO_X Schottky Junction on Arbitrary Substrates. *Adv. Electron. Mater.* 4, 1800373. doi:10.1002/aelm.201800373

Liu, H., Lu, T., Li, Y., Ju, Z., Zhao, R., Li, J., et al. (2020). Flexible Quasi-van der Waals Ferroelectric Hafnium-Based Oxide for Integrated High-Performance Nonvolatile Memory. *Adv. Sci.* 7, 2001266. doi:10.1002/advs.202001266

Liu, Q., Sun, J., Lv, H., Long, S., Yin, K., Wan, N., et al. (2012). Real-time Observation on Dynamic Growth/dissolution of Conductive Filaments in Oxide-Electrolyte-Based ReRAM. *Adv. Mater.* 24, 1844–1849. doi:10.1002/adma.201104104

Liu, Y., Liu, Z., Zhu, B., Yu, J., He, K., Leow, W. R., et al. (2017). Stretchable Motion Memory Devices Based on Mechanical Hybrid Materials. *Adv. Mater.* 29, 1701780. doi:10.1002/adma.201701780

Lu, Y., Gao, S., Li, F., Zhou, Y., Xie, Z., Yang, H., et al. (2021). Stretchable and Twistable Resistive Switching Memory with Information Storage and Computing Functionalities. *Adv. Mater. Technol.* 6, 2000810. doi:10.1002/admt.202000810

Lv, Z., Hu, Q., Xu, Z. X., Wang, J., Chen, Z., Wang, Y., et al. (2019). Organic Memristor Utilizing Copper Phthalocyanine Nanowires with Infrared Response and Cation Regulating Properties. *Adv. Electron. Mater.* 5, 1800793. doi:10.1002/aelm.201800793

Meng, J.-L., Wang, T.-Y., He, Z.-Y., Chen, L., Zhu, H., Ji, L., et al. (2021). Flexible boron Nitride-Based Memristor for *In Situ* Digital and Analogue Neuromorphic Computing Applications. *Mater. Horiz.* 8, 538–546. doi:10.1039/D0MH01730B

Park, H. L., Kim, M. H., and Lee, S. H. (2020a). Introduction of Interfacial Load Polymeric Layer to Organic Flexible Memristor for Regulating Conductive Filament Growth. *Adv. Electron. Mater.* 6, 2000582. doi:10.1002/aelm.202000582

Park, H. L., Lee, Y., Kim, N., Seo, D. G., Go, G. T., and Lee, T. W. (2020b). Flexible Neuromorphic Electronics for Computing, Soft Robotics, and Neuroprosthetics. *Adv. Mater.* 32, 1903558. doi:10.1002/adma.201903558

Park, S. P., Tak, Y. J., Kim, H. J., Lee, J. H., Yoo, H., and Kim, H. J. (2018). Analysis of the Bipolar Resistive Switching Behavior of a Biocompatible Glucose Film for Resistive Random Access Memory. *Adv. Mater.* 30, 1800722. doi:10.1002/adma.201800722

Pedretti, G., Mannocci, P., Li, C., Sun, Z., Strachan, J. P., and Ielmini, D. (2021). Redundancy and Analog Slicing for Precise In-Memory Machine Learning-Part I: Programming Techniques. *IEEE Trans. Electron. Devices* 68, 4373–4378. doi:10.1109/TED.2021.3095433

Qian, K., Tay, R. Y., Lin, M.-F., Chen, J., Li, H., Lin, J., et al. (2017). Direct Observation of Indium Conductive Filaments in Transparent, Flexible, and Transferable Resistive Switching Memory. *ACS Nano* 11, 1712–1718. doi:10.1021/acsnano.6b07577

Raeis-Hosseini, N., and Rho, J. (2021). Solution-processed Flexible Biomemristor Based on Gold-Decorated Chitosan. *ACS Appl. Mater. Inter.* 13, 5445–5450. doi:10.1021/acsami.0c21300

Rahman, M. A., Ahmed, T., Walia, S., Sriram, S., and Bhaskaran, M. (2018). Oxygen-deficient Strontium Titanate Based Stretchable Resistive Memories. *Appl. Mater. Today* 13, 126–134. doi:10.1016/j.apmt.2018.08.011

Ren, S., Li, Z., Tang, L., Su, X., Zhang, H., Zhang, G., et al. (2020). Conduction Response in Highly Flexible Nonvolatile Memory Devices. *Adv. Electron. Mater.* 6, 2000151. doi:10.1002/aelm.202000151

Shi, Q., Wang, J., Aziz, I., and Lee, P. S. (2020). Stretchable and Wearable Resistive Switching Random-Access Memory. *Adv. Intell. Syst.* 2, 2000007. doi:10.1002/aisy.202000007

Shu, P., Cao, X., Du, Y., Zhou, J., Zhou, J., Xu, S., et al. (2020). Resistive Switching Performance of Fibrous Crosspoint Memories Based on an Organic-Inorganic Halide Perovskite. *J. Mater. Chem. C* 8, 12865–12875. doi:10.1039/D0TC02579H

Siddik, A., Haldar, P. K., Paul, T., Das, U., Barman, A., Roy, A., et al. (2021). Nonvolatile Resistive Switching and Synaptic Characteristics of lead-free All-Inorganic Perovskite-Based Flexible Memristive Devices for Neuromorphic Systems. *Nanoscale* 13, 8864–8874. doi:10.1039/D0NR08214G

Siddiqui, G. U., Rehman, M. M., Yang, Y.-J., and Choi, K. H. (2017). A Two-Dimensional Hexagonal boron Nitride/polymer Nanocomposite for Flexible Resistive Switching Devices. *J. Mater. Chem. C* 5, 862–871. doi:10.1039/C6TC04345C

Sim, K., Rao, Z., Zou, Z., Ershad, F., Lei, J., Thukral, A., et al. (2019). Metal Oxide Semiconductor Nanomembrane-Based Soft Unnoticeable Multifunctional Electronics for Wearable Human-Machine Interfaces. *Sci. Adv.* 5, eaav9653. doi:10.1126/sciadv.aav9653

Sun, L., Zhang, Y., Han, G., Hwang, G., Jiang, J., Joo, B., et al. (2019). Self-selective van der Waals heterostructures for large scale memory array. *Nat. Commun.* 10, 3161. doi:10.1038/s41467-019-11187-9

Sun, S., Zhuang, X., Wang, L., Liu, B., Zhang, B., and Chen, Y. (2017). BODIPY-based Conjugated Polymer Covalently Grafted Reduced Graphene Oxide for Flexible Nonvolatile Memory Devices. *Carbon* 116, 713–721. doi:10.1016/j.carbon.2017.02.034

Sun, Z., Ambrosi, E., Bricalli, A., and Ielmini, D. (2018). Logic Computing with Stateful Neural Networks of Resistive Switches. *Adv. Mater.* 30, 1802554. doi:10.1002/adma.201802554

van de Burgt, Y., Melianas, A., Keene, S. T., Malliaras, G., and Salleo, A. (2018). Organic Electronics for Neuromorphic Computing. *Nat. Electron.* 1, 386–397. doi:10.1038/s41928-018-0103-3

Wang, H., Zhu, B., Wang, H., Ma, X., Hao, Y., and Chen, X. (2016a). Ultra-lightweight Resistive Switching Memory Devices Based on Silk Fibroin. *Small* 12, 3360–3365. doi:10.1002/smll.201600893

Wang, L., Wang, Z., Lin, J., Yang, J., Xie, L., Yi, M., et al. (2016b). Long-term Homeostatic Properties Complementary to Hebbian Rules in CuPc-Based Multifunctional Memristor. *Sci. Rep.* 6, 35273. doi:10.1038/srep35273

Wang, L., Yang, J., Zhu, Y., Yi, M., Xie, L., Ju, R., et al. (2017). Rectification-regulated Memristive Characteristics in Electron-type CuPc-Based Element for Electrical Synapse. *Adv. Electron. Mater.* 3, 1700063. doi:10.1002/aelm.201700063

Wang, M., Cai, S., Pan, C., Wang, C., Lian, X., Zhuo, Y., et al. (2018a). Robust Memristors Based on Layered Two-Dimensional Materials. *Nat. Electron.* 1, 130–136. doi:10.1038/s41928-018-0021-4

Wang, M., Guo, K., and Cheng, H. (2020a). Stretchable HfO_2-Based Resistive Switching Memory Using the Wavy Structured Design. *IEEE Electron. Device Lett.* 41, 1118–1121. doi:10.1109/LED.2020.2995201

Wang, M., Luo, Y., Wang, T., Wan, C., Pan, L., Pan, S., et al. (2021a). Artificial Skin Perception. *Adv. Mater.* 33, 2003014. doi:10.1002/adma.202003014

Wang, M., Wang, W., Leow, W. R., Wan, C., Chen, G., Zeng, Y., et al. (2018b). Enhancing the Matrix Addressing of Flexible Sensory Arrays by a Highly

Nonlinear Threshold Switch. *Adv. Mater.* 30, 1802516. doi:10.1002/adma.201802516

Wang, M., Yan, Z., Wang, T., Cai, P., Gao, S., Zeng, Y., et al. (2020b). Gesture Recognition Using a Bioinspired Learning Architecture that Integrates Visual Data with Somatosensory Data from Stretchable Sensors. *Nat. Electron.* 3, 563–570. doi:10.1038/s41928-020-0422-z

Wang, T.-Y., Meng, J.-L., He, Z.-Y., Chen, L., Zhu, H., Sun, Q.-Q., et al. (2019). Atomic Layer Deposited $Hf_{0.5}Zr_{0.5}O_2$-Based Flexible Memristor with Short/Long-Term Synaptic Plasticity. *Nanoscale Res. Lett.* 14, 102. doi:10.1186/s11671-019-2933-y

Wang, T.-Y., Meng, J.-L., Li, Q.-X., Chen, L., Zhu, H., Sun, Q.-Q., et al. (2021d). Forming-free Flexible Memristor with Multilevel Storage for Neuromorphic Computing by Full PVD Technique. *J. Mater. Sci. Technology* 60, 21–26. doi:10.1016/j.jmst.2020.04.059

Wang, T.-Y., Meng, J.-L., Rao, M.-Y., He, Z.-Y., Chen, L., Zhu, H., et al. (2020d). Three-dimensional Nanoscale Flexible Memristor Networks with Ultralow Power for Information Transmission and Processing Application. *Nano Lett.* 20, 4111–4120. doi:10.1021/acs.nanolett.9b05271

Wang, T., Cui, Z., Liu, Y., Lu, D., Wang, M., Wan, C., et al. (2021). Mechanically Durable Memristor Arrays Based on a Discrete Structure Design. *Adv. Mater.*, 2106212. doi:10.1002/adma.202106212

Wang, T., Wang, M., Yang, L., Li, Z., Loh, X. J., and Chen, X. (2020c). Cyber-Physiochemical Interfaces. *Adv. Mater.* 32, 1905522. doi:10.1002/adma.201905522

Wang, T. Y., Meng, J. L., Chen, L., Zhu, H., Sun, Q. Q., Ding, S. J., et al. (2021c). Flexible 3D Memristor Array for Binary Storage and Multi-states Neuromorphic Computing Applications. *InfoMat* 3, 212–221. doi:10.1002/inf2.12158

Wang, Z., Wu, H., Burr, G. W., Hwang, C. S., Wang, K. L., Xia, Q., et al. (2020e). Resistive Switching Materials for Information Processing. *Nat. Rev. Mater.* 5, 173–195. doi:10.1038/s41578-019-0159-3

Wang, Z., Zeng, T., Ren, Y., Lin, Y., Xu, H., Zhao, X., et al. (2020f). Toward a Generalized Bienenstock-Cooper-Munro Rule for Spatiotemporal Learning via Triplet-STDP in Memristive Devices. *Nat. Commun.* 11, 1510. doi:10.1038/s41467-020-15158-3

Xia, Q., and Yang, J. J. (2019). Memristive Crossbar Arrays for Brain-Inspired Computing. *Nat. Mater.* 18, 309–323. doi:10.1038/s41563-019-0291-x

Xu, J., Zhao, X., Wang, Z., Xu, H., Hu, J., Ma, J., et al. (2019). Biodegradable Natural Pectin-Based Flexible Multilevel Resistive Switching Memory for Transient Electronics. *Small* 15, 1803970. doi:10.1002/smll.201803970

Yang, D., Yang, H., Guo, X., Zhang, H., Jiao, C., Xiao, W., et al. (2020). Robust Polyethylenimine Electrolyte for High Performance and Thermally Stable Atomic Switch Memristors. *Adv. Funct. Mater.* 30, 2004514. doi:10.1002/adfm.202004514

Yang, J. C., Mun, J., Kwon, S. Y., Park, S., Bao, Z., and Park, S. (2019). Electronic Skin: Recent Progress and Future Prospects for Skin-Attachable Devices for Health Monitoring, Robotics, and Prosthetics. *Adv. Mater.* 31, 1904765. doi:10.1002/adma.201904765

Yang, M., Zhao, X., Tang, Q., Cui, N., Wang, Z., Tong, Y., et al. (2018). Stretchable and Conformable Synapse Memristors for Wearable and Implantable Electronics. *Nanoscale* 10, 18135–18144. doi:10.1039/C8NR05336G

Yi, X., Yu, Z., Niu, X., Shang, J., Mao, G., Yin, T., et al. (2019). Intrinsically Stretchable Resistive Switching Memory Enabled by Combining a Liquid Metal-Based Soft Electrode and a Metal-Organic Framework Insulator. *Adv. Electron. Mater.* 5, 1800655. doi:10.1002/aelm.201800655

Yoon, J., Ji, Y., Lee, S.-K., Hyon, J., and Tour, J. M. (2018). Low-Temperature-Processed SiO_X One Diode-One Resistor Crossbar Array and its Flexible Memory Application. *Adv. Electron. Mater.* 4, 1700665. doi:10.1002/aelm.201700665

Yuan, L., Liu, S., Chen, W., Fan, F., and Liu, G. (2021). Organic Memory and Memristors: from Mechanisms, Materials to Devices. *Adv. Electron. Mater.* 7, 2100432. doi:10.1002/aelm.202100432

Zeng, Y., Sun, B., Yu, H.-Y., Wang, X., Peng, H., Chen, Y., et al. (2019). A Sustainable Biomemristive Memory Device Based on Natural Collagen. *Mater. Today Chem.* 13, 18–24. doi:10.1016/j.mtchem.2019.04.008

Zhang, B., Chen, W., Zeng, J., Fan, F., Gu, J., Chen, X., et al. (2021). 90% Yield Production of Polymer Nano-Memristor for In-Memory Computing. *Nat. Commun.* 12. doi:10.1038/s41467-021-22243-8

Zhang, L., Gong, T., Wang, H., Guo, Z., and Zhang, H. (2019a). Memristive Devices Based on Emerging Two-Dimensional Materials beyond Graphene. *Nanoscale* 11, 12413–12435. doi:10.1039/C9NR02886B

Zhang, R., Huang, H., Xia, Q., Ye, C., Wei, X., Wang, J., et al. (2019b). Role of Oxygen Vacancies at the TiO_2/HfO_2 Interface in Flexible Oxide-Based Resistive Switching Memory. *Adv. Electron. Mater.* 5, 1800833. doi:10.1002/aelm.201800833

Zhao, X., Fan, Z., Xu, H., Wang, Z., Xu, J., Ma, J., et al. (2018). Reversible Alternation between Bipolar and Unipolar Resistive Switching in Ag/MoS_2/Au Structure for Multilevel Flexible Memory. *J. Mater. Chem. C* 6, 7195–7200. doi:10.1039/C8TC01844H

Zhou, J., Li, W., Chen, Y., Lin, Y. H., Yi, M., Li, J., et al. (2021). A Monochloro Copper Phthalocyanine Memristor with High-Temperature Resilience for Electronic Synapse Applications. *Adv. Mater.* 33, 2006201. doi:10.1002/adma.202006201

Zidan, M. A., Jeong, Y., Lee, J., Chen, B., Huang, S., Kushner, M. J., et al. (2018). A General Memristor-Based Partial Differential Equation Solver. *Nat. Electron.* 1, 411–420. doi:10.1038/s41928-018-0100-6

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