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VIEW & PERSPECTIVE

Novel intelligent devices: Two-dimensional materials based memristors

Two-dimensional (2D) materials with atomic thickness, non-volatile resistive switching feature and compatibility with the semiconducting technology are naturally a good media of memristors. 2D materials-based memristors with excellent performance, low-power consumption and high integration density can be integrated with other circuit components to implement the complicate logic computing, which will become a key driving force for the development of artificial intelligence.

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Artificial intelligence (AI) as a powerful tool capable of exceeding human capabilities in certain specific classes of problems has received significant attention owing to its increasing applications in various fields such as science, medicine, industry, and education. Over the last few years, AI experiments have used much more computation than previously (Fig. 1, this figure is referred to AI and Compute: https://openai.com/blog/ai-and-compute/) [1]. From Fig. 1, we can see since 2012, the amount of computation in AI has been increased exponentially with a 3.4-month doubling time. Improvements in computing have been a key component of AI progress. However, for the reasons of the discontinuation of Moore's law and low energy efficiency due to the architecture of storage and operation separation in the traditional von Neumann computing system [2], the computing is difficult to meet the growing need in big data era. Hence, it is urgent to explore revolutionary technology to develop artificial intelligence chips based on new devices and brand-new computing architecture system. To this end, the chips with functions of both information storage and processing become a desirable choice to overcome the bottleneck of "storage wall" and "power wall". Memristive device shows great potential to offer the next generation high-density nonvolatile memory (NVM) device and an energy efficient solution to implement computation-in-memory owing to its high speed, low-power consumption, nonvolatility, and capability of merging data storage and computing units [3]. In general, memristive device includes resistive random access memory (RRAM), phase change memory (PCM), ferroelectric devices and so on, and these emerging devices can be classified as resistive switching memory (RSM) devices due to their resistive switching behavior. In recent years, RSM devices have developed rapidly based on various materials and mechanisms, including metal oxides (e.g., TiO_x, MoO_x, WO_x, HfO_x, and TaO_x), metal materials (e.g., Ag or Cu), phase change materials ($Ge_2Sb_2Te_5$), magneto-resistive (MR) material, ferroelectric material, and so on. However, these devices based on conventional materials are encountering limitations such as low integration density, high operation current and insufficiently known mechanism. Therefore, new materials are urgently needed to further develop RSM devices with high integration, low power consumption, high programming speed to meet the requirements of artificial intelligence.

2D materials featuring extreme physical scale (e.g., atomic thickness), excellent mechanical properties, facile quantum control ability, remarkable optical and electrical performances and compatibility with the processing technologies of traditional complementary metal-oxide semiconductors (CMOS), have demonstrated to be promising materials in diversified applications of nanoelectronics and optoelectronics [4]. These burgeoning materials consisting of graphene, hexagonal boron nitride (h-BN), transition-metal dichalcogenides (TMDs), etc., are naturally good RSM media, and have been used as surface electrode or interlayer in the RSM devices. The development of new 2D materials boosts the progress of new memristor systems, which could exhibit properties that conventional memristors are not obtainable owing to a bunch of the particularities of 2D materials [5]. For example, to date, the operation current of conventional memristive devices remains at 10–100 μA level, while 2D-materials-based RSM could further lower the operation current to sub-μA or even nA [6, 7]. Furthermore, due to their atomic thickness, non-volatile resistive switching feature and compatibility with CMOS technology, 2D materials can scale down the device size to further enhance integration, pro-

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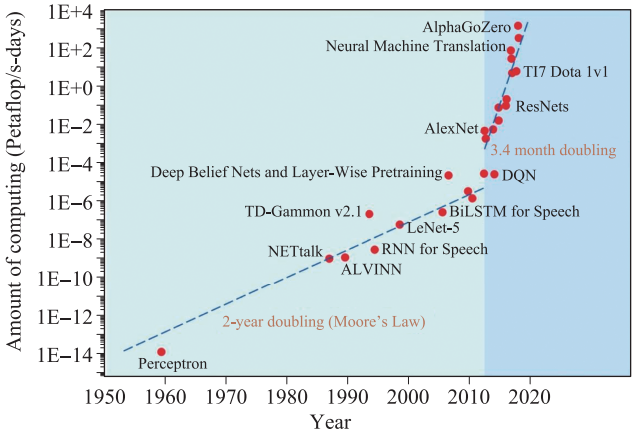

Fig. 1 The total amount of computing used in AI. This figure is referred to AI and Compute: https://openai.com/blog/ai-and-compute/.

viding a promising platform to explore the sub-nanometer scaling limit for RSM devices, which could be integrated with other circuit components to realize the implementation of complicate logic computing. In virtue of these potential merits, 2D-materials-based RSMs are regarded as new-generation intelligent memristors and have drawn giant interests in computing science particularly in the applications of energy-efficient memory.

In a very recent review [8], the memristive phenomena of 2D materials involving graphene, TMDs and h-BN as well as the progress of memristors based on these materials are systematically introduced. A statistical analysis of performance index in these memristors is performed, aiming to compare the existing data visually and promote the index renewal in the memristors. Moreover, the related progress of memristive mechanisms has been remarked and an outlook of the new-generation memristor is presented.

RSMs or memristors typically have a simple form of two terminals devices consisting of two electrodes and a sandwiched resistive switching layer. As a basic passive circuit element, the device resistance can be modulated between a high resistance state (HRS) or a low resistance state (LRS) depending upon the history of applied electrical field. RSMs with unipolar/bipolar feature can be digital or analog process based on different responsive mechanisms. In the review [8] the working principle of RSMs has been classified into three sides: (i) valence change memory, (ii) electrochemical metallization and (iii) thermochemical mechanism. The formation/rupture of a conductive filament (CF) in the dielectric layer is the key factor in the set/reset switching process.

Graphene, as the first fabricated 2D materials with excellent electrical properties, has been studied as electrodes and interfacial materials for improving the performances of memristive devices. In contrast to graphene, graphene oxides are a good choice of switching medium for memristors due to their electrical insulation nature. In addition, TMDs have the advantage over graphene by tunable bandgap through composition variation, thickness control and doping engineering. In 2015, TMDs was first introduced into a memristor and subsequently many types of memristors have been developed based on TMDs. Although the performance of the TMDs-based memristor was improved rapidly, some key parameters (such as switching speed and retention time) are thought to be still unsatisfactory. Except for graphene and TMDs, h-BN as another typical 2D material and sometimes referred as "white graphene", with high flat surface, high thermal conductivity and superior chemical stability can be used to fabricate high-performance memory devices. An atomically thin femtojoule memristive device was fabricated based on h-BN and the operation current is only sub-pA level [7].

From statistics data of performance index shown in Ref. [8], one can see that the highest $I_{\mathrm{ON}}/I_{\mathrm{OFF}}$, cycle times, and retention time are 10^{10}, 10^{7} times and 10^{6} s, respectively. Among them, although the retention time remains unsatisfactory, the values of $I_{\mathrm{ON}}/I_{\mathrm{OFF}}$ and cycle times may meet the development requirements of memristor devices. Controlling the CF formation is very important in obtaining reliable memristive devices. The latest progress in the control strategies of the CF formation was analyzed, including engineering electrodes shapes, intercalating methods and using mesopores materials. (For more details, please refer to Ref. [8]).

In summary, compared with conventional RSM devices, 2D material-based resistors possess advantages of high resistive switching ratio (On/Off ratio), low-power dissipation (low operation current), and high integration density. These memristive devices show the ability to store and process information simultaneously, and have been demonstrated a great potential in intelligent nanoelectronic devices. Although emerging memristors have been explored in the current research, the practical applications of memristor are still facing several challenges: the device reliability needs to be improved; high quality and large-area 2D materials are still urgently needed; the working mechanism and theoretical support are incomprehensive; and most importantly, the performance of logical operation, matrix multiplication and other relative modules based on such new intelligent memristors still need to be further enhanced. The ultimate application and industrialization of these memristive devices based on 2D materials still need a long way to go. However, considering the fast growth of advanced 2D materials with large area and uniform thickness, the realization of high performances of these memristive devices have gotten a great progress [9]. Further, with the development of an appropriate platform including systematic theoretical support, technological innovation [10] and ingenious architectures in the future, we believe that 2D materials-based resistors will be gradually put into practical applications for next-generation highly

scalable memory devices and will thus become a key driving force for the development of artificial intelligence.

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