Research Progress on Two-Dimensional Van der Waals Ferroelectric Materials

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Abstract. This article reviews the application of two-dimensional ferroelectric materials in next-generation high-performance storage and neuromorphic computing, summarizing how electronic doping can enhance the coercive field of sliding ferroelectric materials, thereby improving the stability and anti-interference capability of data storage. However, this article also points out the performance trade-offs it brings, such as the decrease in polarization intensity and the deterioration of crystal quality. Research has found that by using mechanical probe imprinting technology combined with a flexible substrate, it is possible to achieve bidirectional, reversible, and non-destructive polarization switching in ultrathin ferroelectric films, solving the longstanding problem that traditional methods could only switch in one direction. On this basis, this paper discusses the engineering challenges that constrain its commercialization process, such as maintaining a high signal-to-noise ratio in the output while improving storage stability, and converting the inefficient point-by-point probe operations into large-area parallel production techniques compatible with mainstream semiconductor processes.

Keywords: Two-dimensional van der Waals materials, Two-dimensional ferroelectric, Sliding ferroelectric, Ferroelectric field-effect transistor, Memory-computing integration.

1. Introduction

Advances in microelectronics technology have made the computing power-energy consumption bottleneck of the traditional von Neumann architecture more noticeable. Specifically, traditional bulk ferroelectric materials are strictly limited by depolarization fields and critical thickness at the nanoscale, making polarization prone to failure and thus difficult to directly meet the requirements of high-density integration. In contrast, two-dimensional van der Waals (vdW) ferroelectric materials have characteristics such as atomic-scale thickness, a surface free of dangling bonds, and excellent heterojunction compatibility, so they are considered the most promising new direction in this field. Vizner leveraged the characteristics of vdW bonding to overcome the lattice mismatch limitations of traditional epitaxial growth, enabling both stable polarization at ultra-thin scales and unconventional ferroelectric mechanisms such as interlayer sliding [1]. In recent years, this field has made significant breakthroughs in polarization control technologies, such as single-atom limit domain width regulation and mechanical imprinting [2, 3]. Two-dimensional vdW ferroelectric materials are significantly advancing the progress of ultra-low-power transistors, high-density storage, and neuromorphic computing hardware that integrates sensing, computing, and memory [4].

This article explores the application potential of two-dimensional vdW ferroelectric materials in next-generation storage and computing, analyzes their performance regulation mechanisms and microscopic physical characteristics, and aims to dissect the performance trade-offs and large-scale manufacturing bottlenecks faced by this technology in the commercialization process.

2. Physical Mechanisms and Regulatory Breakthroughs of Two-Dimensional Van der Waals Ferroelectrics

2.1. Unconventional Ferroelectric Mechanism

In addition to the ferroelectric mechanism typically based on intrinsic atomic displacements, two-dimensional vdW ferroelectric materials also exhibit unique interlayer *Ferroelectricity induced by*

interlayer *sliding* that is different from traditional bulk ferroelectrics [1]. This makes it possible to artificially construct ferroelectricity in non-polar parent materials despite the scarcity of intrinsic ferroelectric materials. For example, Vizner confirmed the feasibility of inducing interfacial ferroelectric polarization and achieving switching by introducing specific twist angle stacking or performing lateral sliding at the interface in a non-polar hexagonal boron nitride (hBN) system, pioneering a new form of regulation in slidetronics [1]. Thereafter, Chen and others took the graphene/boron nitride/graphene sandwich structure as an example to reveal the significant enhancement effect of interfacial charge redistribution on intralayer electric dipoles, and pointed out that the interlayer compression driven by extraordinary cross-layer electrostatic attraction simultaneously enhances interlayer polarization, thereby achieving a synergistic buildup of intralayer and interlayer polarization in nominally nonpolar vdW multilayer systems [5]. At the same time, Xu introduced a 'compressed modulation of interlayer sliding' mechanism in the bilayer vdW GeSe/SnS heterostructure, using vertical compression to trigger a novel structural phase transition, and found that the newly induced intermediate phase, together with the conventional ferroelectric/antiferroelectric phases, constructed an octuple-state sliding ladder ferroelectricity [6]. These studies all provide the feasibility of artificially constructing and precisely controlling polymorphic polarization in nonpolar vdW systems using interlayer coupling and mechanical fields. By leveraging the advantages of atomic-level thickness and the absence of dangling bonds in two-dimensional materials, it fundamentally addresses the depolarization field limitations of traditional bulk ferroelectric materials at the nanoscale, providing a feasible solution and laying a solid theoretical and experimental foundation for the development of ultra-low-power logic devices, high-density non-volatile memory.

2.2. New Polarization Manipulation Technology

In current research on two-dimensional ferroelectric polarization switching and domain engineering, the control methods have gradually expanded from the traditional out-of-plane electric field to mechanical forces, novel electric fields, and strain engineering, and good results have been achieved in all directions. Specifically, first in terms of mechanical manipulation, Liu et al. proposed and realized a mechanical nano-probe imprinting technology based on the flexoelectric effect by utilizing the inherent in-plane anisotropic flexibility of ultrathin two-dimensional material ($CuInP_2S_6$) combined with an elastic substrate [3]. This technology has successfully overcome the limitation of mechanical polarization in traditional bulk materials, which could only flip in one direction, making bidirectional, reversible, and non-destructive polarization reversal possible, while also constructing high-density artificial ferroelectric nanodomains up to 31.4 Gbit/in² [3]. Secondly, in terms of novel electric field control, Guan and others successfully triggered and induced the antiferroelectric-ferroelectric phase transition of the material at room temperature by applying a special and specific electric field to the two-dimensional van der Waals layered GeSe [7]. The results obtained by this research institute that change the traditional polarization patterns are of great physical significance for exploring valley-selective excitation coupled with ferromagnetic characteristics and developing new types of nanoelectronic and ferrovalley devices. Finally, in terms of lattice and strain engineering control, Shi and others introduced specially designed in-plane strains and out-of-plane stacking methods in two-dimensional ferroelectric SnSe, successfully inducing and regulating the distortion behavior of domain walls. They experimentally confirmed the physical mechanism of this control at the atomic scale using 4D-STEM technology, thereby opening up a new physical perspective for the precise manipulation of artificial nanodomains [8]. Current techniques for regulating two-dimensional ferroelectric polarization and domain engineering have successfully evolved from conventional out-of-plane electric fields to multi-physical fields. These advancements in multi-dimensional manipulation technologies not only effectively address the constraints of traditional macroscopic methods but also offer essential engineering strategies and physical underpinnings for the creation of ultra-high-density artificial nanodomains, the investigation of valley-selective excitation mechanisms, and the innovation of novel valleytronic and nanoelectronic devices.

2.3. Material Modification and Exploration of Physical Limits

In order to achieve practical applications of two-dimensional ferroelectric materials, the academic community has conducted extensive exploration in both material modification engineering and the physical size limits. In terms of modification engineering, Liu and others achieved electron doping by introducing sulfur vacancies in multilayer 1T′-ReS_2, effectively increasing the energy conversion barrier of this sliding ferroelectric system, successfully enhancing the coercive field of the material, and providing a new pathway for the regulation of its polarization characteristics and non-volatility [9]. At the same time, in exploring the physical size limits of ferroelectrics, Han et al. successfully grew ultrathin two-dimensional bismuth tellurate (Bi_2TeO_5) using chemical vapor deposition (CVD) technology, demonstrating the presence of intrinsic in-plane room-temperature ferroelectricity in this material system [2]. Subsequently, the experiment also demonstrated that its spontaneous polarization primarily originates from the displacement of bismuth (Bi) atomic columns, and furthermore, successfully extended the continuously tunable scale of ferroelectric domain widths to the physical limit of a single atomic layer, thereby providing a clear and robust new approach for the development of ultra-high-density information storage and nanoelectronic devices.

3. Two-Dimensional Ferroelectric Integrated Device Architecture and Applications

3.1. Low-Power Logic Device

Due to the new requirements for materials brought by novel non-volatile memory and compute-in-memory architectures, Wu and others designed a wafer-level synthesis method compatible with back-end-of-line (BEOL) processes [10]. This method constructs a heterojunction FeFET with a high-quality coherent interface by combining the ultrathin vdW ferroelectric oxide Bi_2SeO_5 with the two-dimensional semiconductor Bi_2O_2Se channel, thereby successfully addressing the two challenges of critical thickness and large-area uniformity in traditional ferroelectric integration. The monolayer Bi_2SeO_5 devices it produces not only have excellent across-wafer uniformity (threshold voltage deviation <5%) but also feature a 42 mV/dec subthreshold swing that breaks the physical limit at room temperature [10]. At an ultra-low operating voltage of 0.8 V, the device exhibits a high on/off ratio of $>10^6$, an extremely fast writing speed of 20 ns, and an ultra-high cycling endurance of over 1.5×10^{12} times [10]. Furthermore, based on its polarization reversal and threshold voltage modulation characteristics, an experimental reconfigurable logic-in-memory circuit was constructed, achieving a voltage gain of approximately 15, with switching energy and static power consumption as low as 0.43 pJ and 30 pW, respectively [10]. Therefore, the experiment fully and rigorously verified the potential of ultra-thin two-dimensional ferroelectric materials in ultra-large-scale integration, while also providing an excellent reference for developing highly reliable, adaptive, low-power logic architectures.

3.2. High-Density Storage Device

Two-dimensional ferroelectric tunnel junctions (FTJs) are considered to have an important characteristic of efficient switching between high and low resistance states because they can modulate the tunneling barrier by polarization reversal. Specifically, the Janus GeS bilayer FTJ based on Ge-Ge contact exhibits an excellent storage switching ratio by utilizing the sliding ferroelectric mechanism [11]. Meanwhile, Zhou et al. proposed a thickness-driven interface state modulation strategy in $MoTe_2/BaTiO_3$ two-dimensional/three-dimensional (2D/3D) heterojunctions: by increasing the thickness of $MoTe_2$ from 18 unit cells (u.c.) to 20 unit cells, a work function shift of 0.44 eV is induced, resulting in a transformation of the interfacial contact characteristics [12]. Therefore, it is possible to read and switch the ferroelectric polarization state of the underlying $BaTiO_3$ without causing device failure, thereby enabling the driving transport mechanism to switch under the control of the polarization field from trap-assisted space-charge-limited conduction (SCLC)

and thermal emission to Fowler-Nordheim (F-N) tunneling. This provides a new approach for developing multi-level non-volatile hybrid-dimensional memory. This also indicates that the storage stability and control flexibility of a single ferroelectric system can be greatly enhanced through proper interfacial physical engineering.

In order to overcome the density limitations of traditional binary storage and achieve higher-dimensional logic operations, in recent years, the introduction of magnetic degrees of freedom into ferroelectric systems has received increasing attention and exploration. On the basis of enhancing the control dimensions, a two-dimensional multiferroic tunnel junction (MFTJ) combining ferroelectricity and magnetism was designed, thereby achieving higher-density multifunctional storage. Zhou and others constructed a $Mn_2P_2Se_6/Sc_2CO_2$ vdW heterojunction model and realized four non-volatile resistance states by utilizing a significant magnetoelectric cooperative coupling effect [13]. Specifically, in the P↑ polarization state of the AC stack, switching the electrode magnetic moments from parallel (M↑↑) to antiparallel (M↑↓) can achieve a giant tunneling magnetoresistance (TMR) of up to $8.48 \times 10^3\%$ [13]. The anti-parallel magnetic moment configuration (M↑↓) can greatly amplify the differences in electron transport. In this magnetic state, changing the ferroelectric polarization direction of Sc_2CO_2 can achieve a tunneling electroresistance (TER) effect of up to 621% [13]. Because the selected two-dimensional material is inherently extremely thin and has a very low polarization switching barrier (only tens of meV), the multiresistance characteristics controlled by crossed electric and magnetic fields provide a solid theoretical foundation for low-power, high-density non-volatile storage and spintronic applications. These studies, through precise interface physical engineering and the introduction of magnetic degrees of freedom, have significantly enhanced the storage stability and multi-dimensional tunability of two-dimensional ferroelectric tunnel junctions (FTJs). They not only demonstrate the feasibility of cross-control of multiple resistance states by electric and magnetic fields but also propose design concepts for ultra-high-density, low-power multi-level non-volatile memory and spintronic devices.

3.3. Emerging Brain-like Architecture Hardware

Due to vdW ferroelectric materials having atomic-level thinness and intrinsic non-volatility, they are considered an ideal platform for breaking the von Neumann architecture bottleneck and designing memory-computation integrated neuromorphic hardware. In terms of the development direction of artificial synaptic units, Li et al. constructed a two-dimensional sliding ferroelectric memory and synapses (SFeMs) based on rhombohedral-stacked (R-stacked) bilayer $MoS2$, utilizing interlayer shear-induced out-of-plane polarization reversal, making precise non-volatile regulation of channel conductance states possible [14]. The experimental results show that this device has a large storage window greater than 8V, a high conductance ratio exceeding 10^6, data retention characteristics of more than 10 years, and programming cycle endurance over 10^4 times [14]. In addition, thanks to the atomic-level thickness and mechanical toughness of van der Waals materials, the device maintains stable performance even after more than 10^3 bends on a flexible substrate [14]. Therefore, the developed sliding ferroelectric synapse can successfully and reliably simulate typical Hebbian plastic behaviors such as long-term potentiation/depression (LTP/LTD) and spike-timing-dependent plasticity (STDP) and has achieved an image recognition accuracy of up to 97.81% in convolutional neural network (CNN) simulations, providing a theoretical and practical foundation for realizing sliding ferroelectric devices as high-performance, flexible neuromorphic hardware units [14].

With significant progress in the performance of individual two-dimensional ferroelectric devices, the research focus is shifting from single memory units to system-level architectures. Two-dimensional ferroelectric materials, with intrinsic advantages such as dangling-bond-free surfaces, ultrafast polarization reversal, and ultra-low power consumption, lay the physical foundation for constructing in-sensor and in-memory neuromorphic computing architectures [4]. This type of architecture can efficiently simulate synaptic weights, neuron models, and neural network image processing functions by integrating sensing, storage, and computing units in situ. More importantly, unlike traditional architecture, this architecture fundamentally eliminates the transmission delays and

energy consumption caused by frequent data movement between perception and processing units, thereby fundamentally solving the von Neumann bottleneck. Therefore, it is not only beneficial for efficient visual information preprocessing but also provides a complete and reliable theoretical framework and experimental support for the subsequent design of high-performance retinomorphic computing-integrated neuromorphic chips.

4. Discussion

However, because pushing the discussed technology towards large-scale commercialization inevitably requires solving several major and difficult engineering problems, there are two most prominent problems that need to be addressed. First is the problem of optimizing the enhancement of the coercive field and the maintenance of polarization intensity simultaneously. In the study of two-dimensional sliding ferroelectric 1T′-ReS_2 by Liu et al., the energy conversion barrier of the system was effectively improved through doping and annealing [9]. Specifically, after H_2/Ar annealing treatment, the coercive field of the sample significantly increased from 530 kV/cm to 870 kV/cm, which greatly improved the non-volatility and anti-interference capability of the device [9]. However, this annealing process also slightly damages crystal purity and preferred orientation, which in turn causes a decrease in polarization intensity [9]. A decrease in polarization intensity naturally leads to a reduction in the signal-to-noise ratio during signal reading, thereby increasing the difficulty of peripheral circuit design. At the same time, randomly distributed defects/vacancies trigger random microscopic distributions of charge fluctuations, which directly cause intrinsic performance loss. The trade-off between high stability and high signal strength, which is difficult to achieve simultaneously, is the main obstacle hindering the mass production of high-performance storage using two-dimensional ferroelectric materials. To break this bottleneck, future research could focus on developing more precise atomic-level defect engineering, using single-atom substitution to replace traditional large-area annealing doping. At the same time, it is also possible to introduce strain engineering or construct Van der Waals heterojunctions with band alignment, in order to achieve a balance between coercive field and polarization strength through interfacial coupling effects without compromising the intrinsic lattice perfection.

Secondly, there is the problem of the scalability and parallelization of polarization control. Taking the ultrathin two-dimensional ferroelectric $CuInP_2S_6$ (CIPS) as an example, Liu and others used the tip imprinting technique to overcome the limitation of traditional flexoelectric control methods that can only switch in one direction, achieving high-density, bidirectional, reversible, and non-destructive arbitrary polarization manipulation at the nanoscale [3]. However, the point-by-point scanning mode of the currently used atomic force microscope (AFM) probes, although extremely precise in the laboratory, is highly inefficient and costly, and therefore cannot directly meet the requirements of industrial-scale manufacturing [3]. The biggest obstacle currently faced by research on polarization operations is how to transform nanoscale manipulations into large-area, high-throughput parallel technologies. To meet mass production demands, future technological developments are expected to introduce wafer-level nanoimprint lithography (NIL) technology compatible with existing CMOS processes, using hard templates with micro-nano periodic structures to apply large-area mechanical stress, enabling parallel batch writing of polarization arrays. Secondly, exploring global non-contact polarization switching mechanisms based on photostrictive effects or surface acoustic waves (SAW) is also a highly potential development direction to replace probe-by-probe operations and significantly improve production throughput.

5. Conclusion

Overall, two-dimensional van der Waals ferroelectric materials have made remarkable progress in recent years, from fundamental physical mechanisms to integrated device applications. In terms of materials and physical mechanisms, research has progressed from simple observations of polarization

phenomena to the control of domain widths at the single-atom limit, phase transition induction assisted by multiple physical fields, and the precise shaping of ferroelectric stability through interface engineering. In terms of device architecture, ferroelectric tunnel junctions (FTJs), ferroelectric field-effect transistors (FeFETs), and novel computing-in-memory neuromorphic hardware designed based on these materials make excellent use of their atomic-level thickness, lack of dangling bonds, and ultrafast switching characteristics. They have already demonstrated outstanding performance in low-power logic computing and high-density non-volatile storage, particularly achieving experimental validation in neuromorphic computing applications such as image recognition and retina-like chips.

However, moving two-dimensional ferroelectric storage technology from the laboratory to commercial use still requires facing unavoidable engineering challenges. Namely, the issues of balancing performance and stability, as well as the bottleneck problem in production efficiency. Therefore, although significant progress has been made in the research of two-dimensional van der Waals ferroelectric materials and some results are promising for industrial pilot production, at the same time, most studies still need to first address various issues, such as 'performance loss' and 'production speed', before the technology can truly be applied practically.

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