

Research Progress on Performance Improvement of Spin Valve

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Abstract. Spin valves, as core devices in spintronics, hold an irreplaceable position in cutting-edge fields such as information storage, magnetic sensing, and quantum computing. With increasing application demands, enhancing spin valve performance has become a common focus in both academic and industrial sectors. This paper systematically reviews recent research progress in spin valve performance enhancement, exploring key technological pathways for improving magnetoresistance ratio, enhancing temperature stability, and optimizing switching characteristics. Analyzing spin valve performance enhancement strategies from multiple dimensions: at the process level, precise control of interface quality and lattice matching significantly improves carrier transport properties; in hardware aspects, high spin polarization materials (such as Heusler alloys and Fe/MgO structures) combined with perpendicular magnetic anisotropy designs enhance device integration and thermal stability; in structural design, double-barrier structures and superlattice structures strengthen magnetoresistance response through multi-interface synergistic effects; in functional regulation, external field technologies including electric field effects, strain control, and optical field regulation provide new approaches for dynamic optimization of spin valve performance, with multi-field coupling control showing particularly significant advantages. Research indicates distinct differences in optimization strategies across application scenarios: laboratory research emphasizes combining material optimization with interface engineering, while industrial production focuses more on process simplification and cost control. Future spin valve performance enhancement will tend toward diversified development, with promising prospects for two-dimensional materials and topological materials, further deepening of multi-field coupling control technologies, and potential breakthroughs in quantum spin technology. For industrial applications, improving device reliability, consistency, and cost control remain key challenges. Cross-disciplinary integrated innovation will become the core driving force for next-generation high-performance spintronic devices, providing a solid foundation for spintronic applications in emerging fields.

Keywords: Spin valve; Magnetoresistance effect; Organic spin valve; Performance optimization; External field control.

1. Introduction

Spin valve technology began in 1988 with the discovery of the GMR effect by Fert and Grünberg (Nobel Prize in Physics 2007). In 1991, the concept of spin valve was proposed, and Dieny et al. first proposed the concept of spin valve and realized the GMR effect at room temperature ^[1] (giant magnetoresistance effect). In 1995, IBM (International Business Machines Corporation) applied spin valves to hard disk read heads, greatly increasing storage capacity. Since that 21st century, with the further study of TMR effect and the introduction of MgO and other high quality tunnel barrier materials, the magnetoresistance of spin valve has increased from a few percentage points of early GMR type to hundreds or even thousand percentage point of modern TMR type ^[2]. Various new spin valves are emerging, such as vertical spin valves, multilayer spin valves and organic spin valves ^[3]. In 2001, MgO barrier TMR effect was predicted theoretically (>1000%); in 2004, Xiong et al. realized organic spin valve based on Alq₃ for the first time, opening a new era of organic spin electronics ^[4]. Spin valve technology has experienced evolution from metal to organic material, from single structure to complex design. 2010 CoFeB/MgO/CoFeB structure room temperature TMR>200%; after 2015 flexible and multifunctional spin valve rise; in recent years multi-physical field control spin valve development.

Spin valve with high sensitivity, low power consumption, fast response and other advantages, applied in various fields: (1) information storage field: as the core component of hard disk read head, promote

the storage density from GB to TB era; STT-MRAM is expected to become the next generation of "universal memory" [5]; (2) sensing technology: used for position detection, angle measurement and biological detection, such as automotive ABS, electronic compass, etc. [6]; (3) quantum computing and neuromorphic computing: realize quantum bits and artificial synapses, build ultra-low power computing hardware [7]; (4) medical health, aerospace, Internet of Things and other emerging fields also show unique advantages.

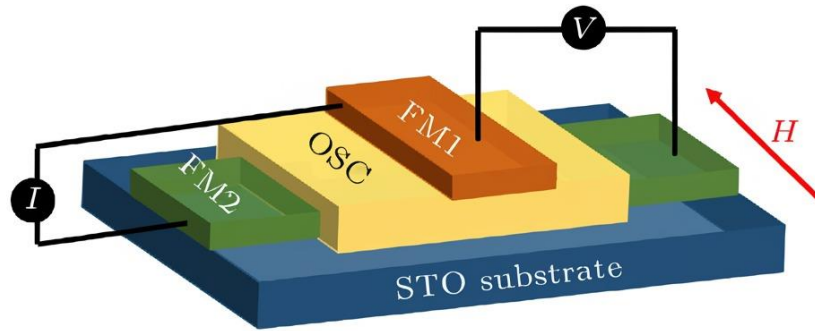


Figure 1. Typical structure diagram of spin valve

The structure of the spin valve is as shown in FIG. 1, as long as it includes a base layer (SiO_2/STO), a bottom ferromagnetic layer FM1 (pinned layer), an antiferromagnetic layer (fixed magnetization direction of pinned layer), a nonmagnetic spacer layer (Cu for GMR type, [2] $\text{MgO}/\text{Al}_2\text{O}_3$ for TMR type), a top ferromagnetic layer FM2 (free layer), and a protective layer [8]. The magnetization direction, circuit connections, and external field direction are indicated. TMR spin valves are based on the correlation between spin polarizability and tunneling probability of ferromagnetic electrodes [9,10]

Although remarkable progress has been made in the research of spin valves, the spin valve technology still faces challenges: the magnetoresistance ratio of commercial TMR spin valves at room temperature (200%-600%) is far from the theoretical prediction (>1000%); the magnetoresistance ratio of organic spin valves is low (<20%) and is mostly limited to low temperature operation [11]; interface quality control, mechanism understanding and process stability restrict performance improvement. Researchers have made breakthroughs in many directions: material optimization (ferromagnetic materials, spacer layers, organic materials), structure design (multilayer structure, interface engineering), process improvement (deposition, heat treatment) and external field regulation (electric field, strain, optical field) [3,12]. This paper will systematically sort out the research progress and advantages and disadvantages of these methods. Figure 2 shows the classification.



Figure 2. Classification of spin valve performance improvement methods

2. Main methods to improve the performance of spin valves

2.1. Material optimization

Material selection and optimization is the basis and key to improve the performance of spin valves. The performance of spin valves depends largely on the spin polarizability of ferromagnetic electrode materials, the interface quality of spacer/barrier materials and the spin-dependent transport properties of organic semiconductor materials [13]. This section will systematically introduce the research progress of material optimization from these three aspects.

2.1.1 Ferromagnetic electrode material optimization

Traditional ferromagnetic electrode materials mainly include Fe, Co, Ni and their alloys (such as NiFe, CoFe, etc.) [14], and their spin polarizations are usually in the range of 30%-50%, which directly limits the maximum magnetoresistance ratio that spin valve devices can achieve [15]. In recent years, researchers have devoted themselves to developing new ferromagnetic materials with high spin polarizability. Semimetallic ferromagnets (such as Co_2MnSi , Fe_3O_4 , CrO_2 , etc.) theoretically have spin polarizability close to 100% [16], making them ideal electrode materials for high-performance spin valves. However, semi-metallic properties are often extremely sensitive to defects and surface/interface states, making it difficult to give full play to their theoretical advantages in practical applications. Perovskite-type materials (such as $\text{La}_{0.7}\text{Sr}_{0.3}$). The TMR [17,18] ratio based on LSMO/STO/LSMO structure can reach 1800% at low temperature, but the spin polarization decreases sharply with the increase of temperature, and the performance decreases greatly at room temperature.

The interface treatment technology of electrode materials also significantly affects the performance of spin valves. For example, proper annealing of CoFeB electrode can promote diffusion of B atoms into barrier layer, improve crystal quality of CoFe/MgO interface, and effectively improve TMR ratio. As shown in Figure 3, different ferromagnetic electrode materials and their treatment processes have a significant impact on the magnetoresistance ratio of spin valves, in which the interface optimized CoFeB/MgO/CoFeB structure can achieve a TMR ratio of more than 500% at room temperature, much higher than traditional material systems [19]

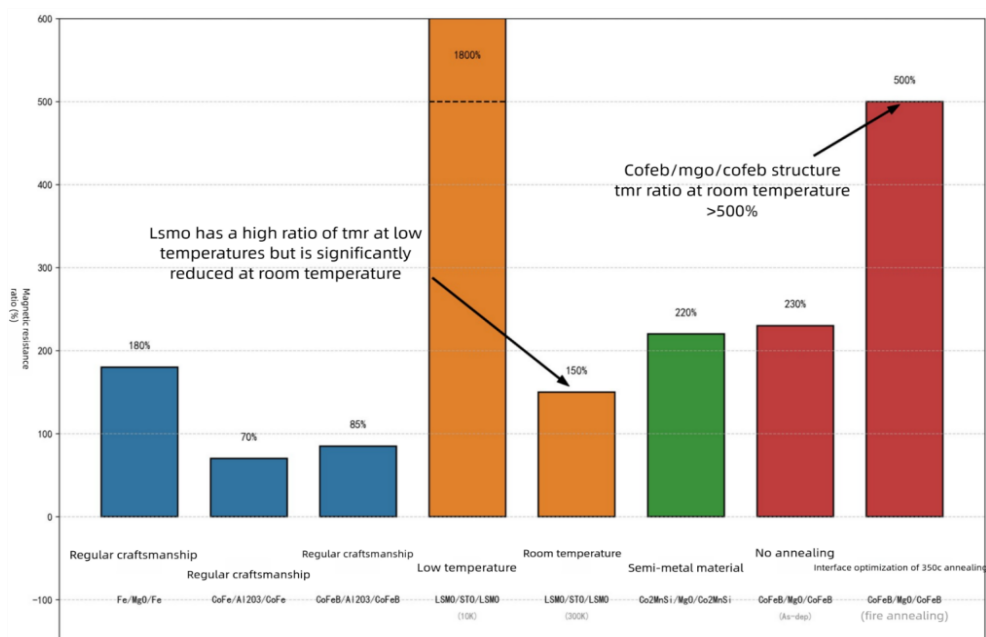


Figure 3. Comparison of magnetoresistive ratios of different ferromagnetic electrode materials

2.1.2 Organic semiconductor materials

Organic semiconductor materials provide a new development direction for spin valves due to their advantages such as flexibility, light weight, and solution processability. Xiong et al. first realized an

organic spin valve based on LSMO/Alq/Co structure by using Alq₃(tris (8-hydroxyquinoline) aluminum) in 2004^[5], and obtained a magnetoresistance ratio of about 40% at room temperature, which opened the study of organic spintronics. Li Dan et al. systematically studied the influence of molecular length and spin-orbit coupling effect on spin transmission performance in organic materials^[20], which promoted the development of subsequent new organic semiconductor materials, including small molecular materials (such as C₆₀, CuPc, etc.), π conjugated polymers (such as P3HT, PFO, etc.) and two-dimensional organic frameworks (COFs). Recently, Liu et al. successfully fabricated organic spin valves based on acid-stripped two-dimensional covalent organic frame films^[21,22], which have achieved significant improvements in spin diffusion length and carrier mobility.

Ilaria et al. found that oxygen impurity in organic spin valve is closely related to bistable state and magnetoresistance^[6]. The spin transport mechanism in organic materials has not been fully clarified. The existing theories include direct tunneling mechanism, hopping conduction mechanism and double spin filtering mechanism. Ding Mingyi et al's research based on organic single crystals^[23] shows that the "abnormal" magnetoresistance effect observed in organic spin valves (resistance decreases with temperature) implies that the organic layer not only acts as a spacer layer, but also may participate in the spin filtering process, which provides a new idea for designing high performance organic spin valves.

2.1.3 Tunneling barrier/spacer materials

The choice of tunneling barrier/spacer materials directly affects the spin-dependent transport efficiency^[24]. Liu Shuai et al. showed that GMR spin valves often use Cu as a nonmagnetic spacer layer^[25], while TMR spin valves initially use amorphous Al₂O₃ as a tunneling barrier, and their room temperature TMR ratio is usually about 70%. Butler et al. predicted theoretically in 2001 that MgO tunneling barrier can greatly improve TMR ratio through coherent tunneling effect of Δ band, while Yuasa et al. experimentally confirmed that MgO based tunnel junction can achieve TMR ratio above 180% at room temperature in 2004. In recent years, Li Chexin et al. have systematically studied the application prospect of two-dimensional materials as new spacer/barrier layers^[13,17], and found that single-layer or few-layer graphene can significantly reduce interface scattering and improve spin transfer efficiency due to its excellent conductivity and atomic-level flat surface^[26]. Xu Ziming et al. confirmed that h-BN, MoS₂ and other two-dimensional materials also show unique tunneling barrier characteristics^[27]. Table 1 compares key performance parameters for different spacer/barrier materials, including critical thickness, tunneling resistance area (RA), and maximum magnetoresistance ratio achieved^[6]

Table 1. Comparison of performance parameters of different spacer materials

Material	Working mode	Critical thickness (nm)	RA($\Omega \cdot \mu\text{m}^2$)	Maximum MR ratio (%)	characteristic
Cu	GMR	2-3	0.1-1	10-20	High electrical conductivity
Amorphous Al ₂ O ₃	TMR	1-1.5	10-100	70-80	Good tunneling characteristics
MgO	TMR	1-2	1-10	180-600	Coherent tunneling effect
Single-layer graphene	GMR/TMR	0.34	0.5-5	10-15	Atomic-level flat interface
Multilayer graphene	GMR/TMR	1-2	1-10	20-30	Adjustable tunneling characteristics
h-BN	TMR	0.7-1	10-50	50-120	Excellent tunneling barriers
MoS ₂	TMR	0.6-1	5-30	40-100	Semiconductor properties
MgO/Al ₂ O ₃ bilayer	TMR	1.5-2.5	5-20	120-250	Optimized the UI status

Composite barrier structures (e.g. MgO/Al₂O₃ bilayer, MgO sandwich structure, etc.) further optimize the spin-dependent transport performance by adjusting the interface state and band matching. For

example, inserting an ultra-thin Ta or Ru layer at the CoFeB/MgO interface can effectively control the magnetic anisotropy of the interface and improve the thermal stability of the spin valve. Ferromagnetic electrode materials determine the upper limit of spin polarization, spacer/barrier materials control spin-dependent transport efficiency, and organic semiconductor materials open up a new direction of flexible spintronics^[28]. Future material optimization will focus more on interface engineering, composite material design and multifunctional material integration to achieve higher performance spin valve devices.

2.2. Structure optimization

On the basis of material optimization, the structure design of spin valve also has important influence on its performance. By multilayer structure design, interface engineering and micro-nano structure optimization, spin transfer process can be effectively controlled, and magnetoresistance ratio, thermal stability and reliability of spin valve can be further improved.

2.2.1. Multi-layer structural design

Traditional spin valves usually use a simple sandwich structure (magnetic layer/nonmagnetic layer/magnetic layer), while multilayer structure design introduces more complex interlayer arrangement and interaction on this basis, effectively enhancing spin filtering and regulation capabilities. Double spin filtering effect is one of the core concepts of multilayer structure design, that is, to form more efficient spin-selective transmission channels by utilizing the differences in band matching and scattering mechanisms between two or more ferromagnetic layers. For example, Hu Yujie's team achieved double filtering in CoFeB/MgO/CoFeB/Ru/CoFeB/MgO/CoFeB double tunnel junction structure, and the magnetoresistance ratio can be increased to more than 600% at room temperature^[29]

Multi-interface synergy is another key strategy, which makes multiple interfaces participate in the spin regulation process by precisely controlling the interface characteristics between functional layers. The results show that MgO interface provides coherent tunneling channel and Fe/Cu interface enhances spin filtering in Fe/MgO/Fe/Cu/Co structure. The two mechanisms cooperate to make the device exhibit higher magnetoresistance ratio than that of single mechanism.

Superlattice structure can realize quantum size effect and new spin transport phenomenon by alternately stacking ultra-thin magnetic layers and nonmagnetic layers to form artificial periodic structure. Typical superlattices such as [Co/Pt]*n* multilayer films have perpendicular magnetic anisotropy, which is suitable for developing high density vertical spin valve memory cells, while [CoFeB/Ta]*n* superlattices exhibit more stable magnetization switching behavior and larger magnetoresistance changes by regulating interlayer coupling^[12]

As shown in Figure 4, a typical multi-layer spin valve structure achieves more efficient spin filtering and transport through carefully designed combinations of functional layers. Synthetic antiferromagnetic structure (SAF) and interlayer coupling optimization are important means to improve the performance of spin valve, which can effectively reduce flux leakage, enhance the stability of magnetization direction and improve the response sensitivity to external magnetic field.

2.2.2. Interface engineering

Interface is the key region for the transfer of spin electrons from one material layer to another. Interface characteristics directly affect the transfer efficiency of spin-polarized current. Intercalation layer technology is an important means of interface engineering. Ultrathin (usually 0.5-2nm) transition layers can be inserted between the main functional layers to significantly improve interface characteristics. For example, the insertion of Ta with a thickness of 1-2 atomic layers at the CoFeB/MgO interface can promote the diffusion of B atoms into the Ta layer, improve the crystalline quality of the CoFeB layer and the flatness of the MgO interface, and thus improve the TMR ratio. Similarly, inserting LiF or AlO_x ultra-thin layers into organic spin valves can effectively adjust energy level matching, reduce interface scattering, and achieve more efficient spin injection.

Interface roughness control is another key technology. Rougher interfaces can enhance spin scattering and reduce spin coherence length, which will adversely affect device performance. The results show that the interface roughness can be controlled below 0.3nm by optimizing the film growth kinetics (such as adjusting the deposition rate, substrate temperature and gas pressure), and the performance of spin valve can be improved significantly. Sun Wenchao's team found that, especially in CoFeB/MgO/CoFeB systems, interface atomic-level flatness is critical to achieving high TMR ratios^[30]

Polarization matching optimization aims to achieve optimal matching of spin channels by adjusting the band structure of materials at the interface. For example, introducing CuI or vanadium dioxide at the LSMO/organic layer interface can adjust the Fermi level of organic semiconductors, reduce spin scattering, and improve injection efficiency.

2.2.3. Size effect and micro/nano structure

As the device size decreases to nanometer scale, quantum confinement effects and interface effects become more pronounced, which have a profound impact on the performance of spin valves. Nanoscale spin valves (transverse size <100nm) exhibit magnetization reversal behavior different from macroscopic devices. The results show that when the area of CoFeB/MgO/CoFeB tunnel junction is reduced from 1 μm^2 to 50 nm \times 50 nm, the TMR ratio can be increased by 15%-30%, which is mainly due to the reduction of domain walls and vortex structures in the magnetic layer, and the realization of more uniform magnetization switching ^[31]

Lateral and vertical structures are two main spin-valve configurations. The traditional lateral structure (current parallel to film plane) is simpler in process implementation, but there is a limit of size reduction; the vertical structure (current perpendicular to film plane) is more suitable for high density integration and can make full use of tunneling magnetoresistance effect. Comparative studies show that vertical structures can usually achieve higher magnetoresistance ratio and lower critical current density for the same material system.

Patterned spin valve arrays are fabricated by photolithography or electron beam etching techniques to form spin valve cells into array structures with specific shapes and arrangements, suitable for high-density storage or logic devices. Especially in spin transfer torque (STT) devices, patterning elliptical or annular structures can effectively control the magnetization direction and reduce the critical current.

2.3. Process optimization

In addition to material and structure design, the fabrication process also has a decisive influence on the performance of spin valves. Optimization of thin film deposition process and subsequent processing technology is the key to realize high performance spin valve devices.

2.3.1. Thin film deposition techniques

Magnetron sputtering is one of the most popular techniques for spin valve fabrication, which has many advantages such as high efficiency, good uniformity and wide range of materials. The magnetron sputtering process optimization for spin valve mainly focuses on several aspects: one is to control the growth kinetics of thin films by adjusting sputtering power and pressure to form high-quality crystal structures; the other is to reduce oxidation and impurity contamination by using ultra-high vacuum system (background vacuum <10 Torr); and the third is to introduce auxiliary ion beam assistance or substrate bias technology to promote film densification and interface flatness. The results show that for CoFeB/MgO/CoFeB structure, the best interface quality and magnetoresistance properties can be obtained by sputtering at low power (50-100 W) under argon pressure of 0.5-1 Pa and maintaining substrate temperature in the range of room temperature to 200 °C.

Molecular beam epitaxy (MBE) plays an important role in the study of high quality spin valves because of its precise atomic-level growth control capability. The Fe/MgO/Fe single crystal grown by MBE exhibits ultra-high TMR ratio (>1000% at low temperature) close to the theoretical

prediction, which verifies the coherent tunneling theory. However, MBE technology is inefficient and costly, and is mainly used for basic research and prototype validation ^[32]

Organic material evaporation process is the key technology for preparing organic spin valves. Organic layers are typically deposited using vacuum thermal evaporation or organic molecular beam epitaxy (OMBE) techniques, with key parameters including evaporation source temperature control, substrate temperature, and evaporation rate. The results show that for small molecular materials such as Alq, high quality amorphous films can be formed by controlling the evaporation rate in the range of 0.1-0.5 Å/s and the substrate temperature from -30 °C to room temperature, reducing molecular agglomeration and interface defects.

2.3.2. Heat treatment and micro-nano processing technology

Heat treatment, particularly annealing at 250-350 °C for 30-60 minutes, is crucial for optimizing CoFeB/MgO/CoFeB spin valve performance by improving crystallization and interfaces, though excessive temperatures cause detrimental element diffusion and interface degradation^[33]. [33]. Applying a 0.5-1 T magnetic field during annealing (MFA) induces magnetic anisotropy, significantly boosting the magnetoresistance ratio (TMR) from ~150% as-deposited to over 380%. Controlling harmful interface diffusion requires diffusion barriers (e.g., Ru, Ta) and precise thermal conditions. Complementary nanofabrication techniques, including optimized lithography, etching (e.g., RIE/IBE to minimize edge damage), and contact electrode engineering (e.g., Ta/Au, Ti/Au to reduce resistance), are essential for realizing high-performance devices. Overall, meticulous optimization of heat treatment and fabrication parameters decisively governs spin valve performance.

Table 2. Effect of heat treatment conditions on magnetoresistance ratio

Device structure	Annealing temperature (°C)	Annealing time (min)	Magnetic field strength (T)	Diffusion barrier layer	Initial TMR (%)	TMR after annealing (%)	Lift the ratio (%)	Main mechanism
CoFeB/MgO/CoFeB	No annealing	30	0	None	150	150	0	Pristine state
CoFeB/MgO/CoFeB	250	30	0	None	150	220	47	Improved crystallinity
CoFeB/MgO/CoFeB	300	30	0	None	150	280	87	Significant improvement in crystallinity
CoFeB/MgO/CoFeB	350	30	0	None	150	320	113	Optimal crystallization temperature
CoFeB/MgO/CoFeB	400	30	0	None	150	280	87	Elemental diffusion
CoFeB/MgO/CoFeB	350	30	0.5	None	150	350	133	Magnetic fields induce anisotropy
CoFeB/MgO/CoFeB	350	30	1.0	None	150	380	153	Strong magnetic field for best results
CoFeB/MgO/CoFeB	350	60	1.0	None	150	360	140	Diffusion increases
CoFeB/Ta/MgO/CoFeB	350	30	1.0	Ta	150	400	167	Ta promotes diffusion of B and blocks other elements
CoFeB/Ru/MgO/CoFeB	350	30	1.0	Ru	150	390	160	Ru diffusion blocking effect
CoFeB/MgO/CoFeB	350	30	1.0	None	150	395	163	Rapid cooling inhibits diffusion

Process optimization is the key link to realize high performance spin valve devices from theoretical design to practical application. Thin film deposition technology determines the basic quality of materials and interfaces, while heat treatment and micro-nano processing technology further enhance and solidify device performance. Only by combining material optimization, structure optimization and process optimization can the potential of spin valve devices be fully utilized to achieve high magnetoresistance ratio, low power consumption and high reliability.

2.4. Field regulation

In addition to optimizing the performance of spin valves from the perspectives of materials, structures and processes, external field control techniques have shown unique advantages in recent years, providing the possibility of real-time, reversible and non-contact tuning of spin valve performance. Electric field, strain and optical field modulation have become important directions in spintronics.

2.4.1. Electric field regulation

Electric field control via gate structures effectively modulates spin valve magnetoresistance and magnetization through three key mechanisms: altering ferromagnetic surface electronic configuration/spin polarization, adjusting spin channel carrier concentration/distribution, and modifying interface magnetic anisotropy^[27,31]. For example, applying an electric field of ± 0.5 V/nm to the CoFeB/MgO structure can change the magnetic anisotropy of the interface by 40%, which significantly changes the magnetization switching characteristics of the CoFeB layer.

The change of interface polarization induced by electric field is another important mechanism. In multiferroic/ferromagnetic heterostructures (e.g. BiFeO/CoFe)^[34], electric fields (2-5 V/ μ m) induce ferroelectric switching, which reversibly controls the adjacent ferromagnetic layer's magnetization direction via exchange coupling. This approach achieves magnetization reversal with $\sim 1/100$ th the energy of current-driven methods, enabling ultra-low-power magnetic memory applications.

The effect of electric field on spin lifetime is mainly realized by adjusting spin-orbit coupling strength and scattering mechanism. In semiconductor or organic materials, the gate voltage can adjust the strength of the Rashba effect in the conduction channel, which in turn changes the spin relaxation time. It has been proved that the spin lifetime can be nearly doubled and the magnetoresistance ratio of spin valve can be greatly increased by appropriate gate voltage in organic semiconductor Alq. As an efficient and low-energy spin control method, electric field modulation provides a new way to optimize the performance and extend the function of spin valve devices, especially suitable for the development of new spin electronic devices with low power consumption and multiple functions.

2.4.2. Strain regulation

Strain control technology utilizes the influence of mechanical strain on magnetic and electronic structure of materials to realize dynamic adjustment of spin valve performance. Wang Shaoting et al's study on PMN-PT/magnetic multilayer structure shows^[35] that the influence of mechanical strain on magnetic anisotropy is the core mechanism of strain regulation, which is mainly realized through magnetoelastic effect and magnetostriction effect. When ferromagnetic materials are strained, their lattice structure deforms, resulting in electron orbital recombination and spin-orbit coupling changes, thus adjusting the magnetic anisotropy energy of the materials. Wang Mengxi et al. found in the study of stress regulation of spintronic devices^[29,36] that applying 0.1% uniaxial tensile strain on CoFeB thin films can make its magnetic anisotropy energy change more than 50%, which significantly affects its magnetization switching characteristics.

Yang Huali et al. systematically reviewed the research progress of flexible magnetic thin film materials and devices^[37], and pointed out that the study of spin valve performance on flexible substrates provides a practical application platform for strain regulation. Chen Xia et al. successfully fabricated Co/Cu/ γ -Fe₄N spin valves based on mica flexible substrates^[31], demonstrating that CoFeB/MgO/CoFeB structures fabricated on polyimide (PI) or polyethylene terephthalate (PET) flexible substrates can withstand bending strains up to 2% while maintaining functional integrity, exhibiting excellent mechanical flexibility. More importantly, by controlling the bending degree of the substrate, the magnetoresistance ratio can be reversibly adjusted within the range of $\pm 25\%$, which lays the foundation for the development of wearable and deformable spintronic devices.

Liu Mengli et al's study on multiferroic heterojunction^[16,31] reveals that piezoelectric-magnetolectric coupling effect provides another way to realize strain regulation. The spin valve is integrated on piezoelectric materials (such as PMN-PT, PZT, etc.) to form multifunctional heterostructures. The

strain state of piezoelectric materials is controlled by voltage, and then the magnetization configuration of spin valve is controlled. Fang Mei et al.'s study of ferroelectric-spintronic devices shows ^[18] that this "electric-mechanical-magnetic" coupling transfer chain allows nonvolatile control of spin valves with low voltages (usually <10 V). Typical Fe/MgO/Fe/PMN-PT structure can increase or decrease the magnetoresistance ratio by more than 45% on the basis of the original value under the applied voltage driving, showing excellent multi-field coupling regulation ability.

2.4.3. Light field regulation

Optical field modulation technology utilizes the interaction of light and spin to realize contactless and ultra-fast modulation of spin valve performance. Lu Wentian et al. systematically clarified the basic mechanism of photoexcited spin states for the first time in their study of laser-induced spin valve structures ^[24]. They found that when ferromagnetic materials are irradiated by laser light, photon energy can excite electrons to transition from the ground state to the excited state, changing the electronic structure and spin distribution of the material, and thus affecting its magnetization characteristics. Pan et al. subsequently carried out magneto-optical effects of chiral and achiral perovskites ^[26], demonstrating that pulsed laser light (wavelength 780 nm, power density 10-100 mJ/cm²) applied to CoFeB films can induce magnetization direction changes in picoseconds, much faster than traditional magnetic field control.

Wenting et al. first realized the large-amplitude non-volatile regulation of magnetoresistance in organic spin valves in 2019 ^[10], and developed photoelectric spin valves combining photoelectric effect and spin valve function. Pan Ruiheng et al. further studied the spin transport and magnetic field effect in organic-inorganic hybrid perovskites ^[38,39], and found that in LSMO/ZnO/Co heterostructures ^[33], light can generate photogenerated carriers in ZnO layer, change its conductivity and spin transport characteristics, and realize optical control of magnetoresistance ratio. More importantly, by adjusting the wavelength and intensity of light, fine control of the magnetoresistance ratio can be achieved. In 2021, Pan Ruiheng and Wang Kai showed ^[8] that in the organic spin valve LSMO/P3HT/Co structure, ultraviolet irradiation can increase the magnetoresistance ratio by more than 40%, while visible light irradiation can reduce it by 25%, showing multi-mode optical response characteristics.

Ultrafast optical control is a frontier field of optical field regulation. Ultrafast spin dynamics can be controlled by femtosecond laser pulses. Different from the slow magnetization change caused by thermal effect, Huayan et al. revealed in the angle-dependent magnetoresistance study carried out in 2021 that ultrafast optical control can realize direct regulation of spin orientation at sub-picosecond level through non-thermal effects such as inverse Faraday effect or photoinduced spin transfer ^[40]. The experimental setup and results of the spin valve controlled by the optical field verify the advantages of high temporal resolution and spatial selectivity of the optical control technology, providing a new idea for the high frequency operation of the spin valve.

Because of its advantages of non-contact, ultra-fast and low loss, optical field modulation technology is not only an important tool for studying spin dynamics, but also a new way to realize high-performance spin functional devices, which has broad application prospects in the development of spin electronics in the future.

2.5. Other Emerging Methods

In addition to the mainstream technologies described above, a number of emerging approaches have shown unique potential in recent years to improve spin valve performance. This section focuses on spin-orbit coupling engineering and topological materials applications.

2.5.1. Spin-orbit coupling engineering

Spin-orbit coupling (SOC), as a bridge between electron spin and orbital motion, is a key factor in spin injection, transmission and detection. Spin-orbit coupling engineering improves spin valve performance by modulating SOC strength and characteristics. Heavy metal doping is an effective

method to enhance SOC ^[13], doping high atomic number elements (such as Pt, Ta, W, etc.) into ferromagnetic materials or interface layers can significantly enhance SOC intensity. The results show that doping 5% Pt into CoFeB can increase the spin Hall angle by nearly 5 times and greatly improve the spin current conversion efficiency ^[20,41]. In Ta/CoFeB/MgO structure, Ta layer can generate effective spin-orbit torque due to its strong SOC characteristics, which makes the critical current density of driving CoFeB magnetization flip lower than 10 A/cm², providing a new scheme for low power spin-transfer torque devices.

The enhancement of Rashiba effect is another important strategy, which mainly breaks the spatial inversion symmetry by designing asymmetric interface structures or applying vertical electric fields to generate or enhance Rashiba-type SOC. The typical Pt/Co/AlO_x or Pt/Co/MgO structure produces strong Rashiba effect due to the asymmetry of the interface, which locks the spin and orbital momentum and realizes efficient current-spin conversion. The spin-orbit torque efficiencies of 0.1-0.3 are observed in these structures, which are much higher than those of conventional spin-polarized current drives (usually <0.1).

The spin Hall effect is an important application of SOC engineering. The spin Hall effect makes the current spin in the vertical direction, which provides a new way for spin injection. Heavy metal materials (Pt, Ta, W) and topological insulators exhibit large spin Hall angles (up to 0.1-0.3) and can efficiently convert charge flow into spin currents. The spin-orbit moment magnetic tunnel junction based on W/CoFeB/MgO structure can realize ultra-low power magnetization switching, and its critical current density is one order of magnitude lower than that of traditional structure.

2.5.2. Topological materials and spin valves

Topological materials, because of their unique electronic structure and spin properties, open up a new direction for the study of spin valves ^[42]. Topological materials offer novel pathways for spin valve research due to their unique electronic and spin properties. Topological insulators (e.g., Bi₂Se₃, Bi₂Te₃) provide natural spin-polarized currents through spin-momentum locked surface states, enabling efficient spin injection/detection in heterojunctions with ferromagnets, as evidenced by room-temperature spin Hall magnetoresistance in Bi₂Se₃/NiFe structures. Furthermore, Weyl semimetals (e.g., Co₃Sn₂S₂, Co₂MnGa) exhibit giant anomalous/spin Hall effects from linear dispersion and large Berry curvature near Weyl points; integration into spin valves achieves ultrahigh spin-charge interconversion, demonstrated by Co₂MnGa devices showing anomalous Hall angles up to 2000%·nm at room temperature. Additionally, topologically protected chiral spin textures (e.g., skyrmions) offer high stability and controllable dynamics for novel devices, such as FeGe/CoFeB/MgO-based skyrmion spin valves enabling single-skyrmion electrical detection and unique nonlinear magnetoresistance.^[23]

The combination of topological materials and spin valves represents a new paradigm for spintronics research. Combining quantum topological properties with spintronics functions is expected to achieve more efficient and stable spin control and reading, providing new design ideas and material choices for the next generation of spintronics devices ^[23,34]

3. Summary and Prospect

In this chapter, we summarize and compare all kinds of optimization methods of magnetoelectronic devices studied above, and discuss the future development trend based on the current research progress, which provides reference for the research direction of related fields.

3.1. Comparative Analysis of Various Optimization Methods

Through the study of material selection, interface engineering, structure design and external field control, the contribution of different methods to the performance improvement of magnetoelectronic devices and their applicable conditions can be systematically summarized. This section will

comprehensively evaluate the magnetoresistance ratio improvement effect, process complexity, stability, cost and other dimensions.

3.1.1. Comparison of material optimization methods

As a basic method to improve the performance of magnetoelectronic devices, material optimization has the advantage of directly increasing the tunneling magnetoresistance ratio by selecting materials with high spin polarization. Co-based semi-metallic alloys (e.g. Co_2MnSi , $\text{Co}_2\text{Fe}_{0.4}\text{Mn}_{0.6}\text{Si}$) and Fe-based alloys (especially Fe/MgO/Fe structures) exhibit excellent magnetoresistance properties at room temperature. However, material optimization methods face challenges such as complex preparation process and difficult interface quality control in practical application. Especially for semi-metallic materials, the theoretical prediction of 100% spin polarizability is difficult to be fully realized in practical devices, mainly limited by interface defects and thermal excitation effects.

In contrast, Heusler alloy-based MTJ devices can achieve room temperature TMR of up to 330%, while Fe/MgO/Fe-based structures can achieve magnetoresistance ratios of up to 180% through delta band filtering effects. However, Heusler alloys have strict requirements on preparation conditions, especially the control of annealing temperature and process directly affects the formation of their ordered structure, which in turn affects device performance ^[43]

3.1.2. Evaluation of interface engineering methods

Interface engineering can effectively improve TMR ratio without greatly increasing process complexity by inserting ultra-thin layer, surface passivation and interface atom rearrangement. It has been shown that the TMR ratio can be increased by 15-40% by inserting 1-2 atomic layers of appropriate materials (such as Mg, Al_2O_3 , etc.) between the ferromagnetic layer and the tunneling barrier layer. The advantage of this method is that it is relatively simple to implement and has high reliability, but the disadvantage is that the lifting effect is limited and the precise control of the thickness of the insertion layer is required.

The comparison of different insertion materials shows that Mg insertion layer performs best at Fe/MgO interface, while for CoFeB/MgO interface, the insertion of ultra-thin Ta layer can promote the crystallization and interface flatness of CoFeB more effectively. However, these methods place high demands on material growth equipment, requiring atomic-level precision thickness control capabilities.

3.1.3. Comprehensive comparison of structural design and external field regulation

From the point of view of device structure design, double-barrier MTJ, perpendicular magnetic anisotropy structure and novel three-terminal device design methods have their own characteristics. Perpendicular magnetic anisotropy structure has obvious advantages in high density integration and thermal stability, but the fabrication process is complicated. Double-barrier structures can provide a 20-35% performance improvement while maintaining a relatively simple process, but there is a problem with interlayer coupling control.

External field modulation methods (especially strain modulation and electric field effects) provide dynamic tunability of device performance, which is not available in other methods. Electric field regulation avoids heat loss caused by high current, and the energy efficiency advantage is significant, but the current regulation range is relatively limited (usually 15-25%). Strain regulation shows unique advantages in the field of flexible electronics. More than 45% TMR regulation range can be achieved through piezoelectric-magnetolectric coupling, but the system integration is low.

Various optimization methods have their own advantages and disadvantages in terms of performance improvement effect, process complexity, stability, scalability and cost. Material optimization methods have the most significant effect in improving the absolute value of magnetoresistance ratio, while structural design and external field regulation have irreplaceable advantages in specific application scenarios (such as low power consumption, high integration and dynamic regulation).

3.1.4. Comprehensive optimization strategy analysis

In practical applications, the combination of multiple optimization methods can often produce synergistic effects. For example, the dual optimization of high spin polarizability materials combined with fine interface engineering can improve device stability while maintaining high magnetoresistance ratios. Similarly, the combination of electric field regulation and perpendicular magnetic anisotropy structures provides a viable path for low power, high density magnetic random access memories.

Comprehensive analysis shows that there are obvious differences in the optimal optimization strategies under different application scenarios: for laboratory research in pursuit of extreme performance, the combination of material optimization and interface engineering is the most effective; for industrial production, process simplification and cost control become key factors, and the combination of interface engineering and structure design is more practical; for future low-power, multifunctional devices, external field control methods (especially electric field effects) combined with advanced material systems show broad prospects.

3.2. Future development trend

In the context of the continued development of magnetoelectronic devices, several key trends deserve special attention, and these trends may lead to the development of the next generation of spintronic devices.

3.2.1. Exploration of new materials and structures

The application of two-dimensional materials (such as graphene and transition metal disulfides) as tunneling barriers or electrode materials is becoming a research hotspot. The unique electronic structure and atomic-level thickness control of these materials provide a new way to improve spin transfer efficiency. Especially, the appearance of two-dimensional magnetic materials (such as CrI₃, Fe₃GeTe₂, Dy/Pt/[Co/Pt]₃, etc.) creates the possibility for the realization of all-two-dimensional spin valve devices^[43]

Topological insulator materials also show great potential for applications as spin filters or spin current sources. The topologically protected state on the surface of these materials makes the electron spin and momentum strictly coupled, which is expected to realize nearly lossless spin transfer, providing a new idea for the design of ultra-low power devices.

In the aspect of structure design, superlattice structure and van der Waals heterojunction are expected to break through the traditional interface limitation and realize more freedom of performance regulation. These novel structures not only enable precise control of interface states, but also enable customized design of spin transfer properties through band engineering.^[18]

3.2.2. Prospects for multi-field coupling regulation

Multi-field coupling regulation (such as electric-magnetic-thermal-strain multi-field synergy) will become an important research direction in the future. Compared with single external field regulation, multi-field coupling can provide greater degree of freedom and amplitude of regulation. It has been proved that the efficiency of magnetic anisotropy modulation can be increased by 2-3 times by the synergistic effect of electric field and strain field, and the critical field strength required for inversion can be significantly reduced by heat assisted electric field modulation.

Especially noteworthy is the synergistic modulation of optical and electromagnetic fields. The spin dynamics induced by ultrafast laser pulses open up a new way for high-speed spin manipulation on the subpicosecond scale. This kind of control method has important application prospect in future high speed spin memory and logic device.^[5]

3.2.3. Possibilities of quantum spin technology

With the development of quantum information technology, quantum computing and quantum communication based on spin qubits have become the frontier research direction. Compared with traditional qubit implementations, spin qubits have the advantages of longer coherence time and easier integration. In particular, single-electron spin and nuclear spin hybrid systems show unique potential for quantum storage and quantum relay.

Spin-photon coupling system is an important bridge between quantum computing and quantum communication, and its research progress will determine the realization path of quantum networks in the future. At present, spin-photon interfaces based on NV color centers and quantum dots have made initial progress, but there are still great challenges in achieving strong coupling and high-fidelity conversion.

3.2.4. Challenges and opportunities in industrial applications

Although the performance of magnetoelectronic devices continues to improve, there are still multiple challenges from laboratory results to industrial applications. Reliability, consistency and cost control are key thresholds for industrial applications. Especially at the nanoscale, the sensitivity of device performance to process fluctuations increases greatly, which requires the development of more precise and controllable nano-fabrication technologies.

However, with the rapid development of 5G communication, Internet of Things and artificial intelligence, the demand for high-performance, low-power nonvolatile storage and computing devices has exploded, creating a broad market space for magnetoelectronics devices. Especially in the field of edge computing and AI chips, magnetoelectronic devices combining storage and computing functions are expected to achieve breakthrough improvements in computing power and energy efficiency.

Facing the future, interdisciplinary fusion innovation will become the core driving force for the development of magnetoelectronic devices. Cross-cooperation in materials science, micro-nano processing, quantum physics and electronic engineering is expected to lead to a new generation of disruptive spintronic devices and systems.

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