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Multiferroic Tunnel Junctions: The Future of Spintronic Devices?

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Abstract: *As the scaling limits of conventional CMOS technologies become increasingly evident due to issues like power dissipation and quantum effects, spintronic devices are emerging as viable alternatives for next-generation electronics. Among these, multiferroic tunnel junctions (MFTJs) have gained significant attention for their potential to realize high-density, energy-efficient, and non-volatile memory and logic systems. MFTJs integrate a ferroelectric barrier between two ferromagnetic electrodes, combining the tunneling magnetoresistance (TMR) of magnetic tunnel junctions (MTJs) with the tunnelingelectroresistance (TER) of ferroelectric tunnel junctions (FTJs). This dual functionality enables four non-volatile resistance states, controllable by electric and magnetic fields. Beyond this binary extension, strong magnetoelectric coupling at the ferromagnet/ferroelectric interface—driven by mechanisms such as spin-dependent screening and interfacial charge redistribution—offers voltage-based control of magnetic and spintronic properties. This review surveys the fundamental operating principles, experimental breakthroughs, and material systems enabling TMR, TER, and tunnelingelectromagnetoresistance (TEMR) in MFTJs. Emphasis is placed on interface engineering strategies and the performance metrics of various device architectures, including room-temperature-operable systems. The continuing development of MFTJs holds promise for multifunctional device applications in memory, logic, and neuromorphic computing.*

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I. Introduction

As the need for faster, more compact, and non-volatile electronic devices grows, existing technologies are encountering significant barriers to further miniaturization, mainly due to power dissipation, quantum mechanical limitations, and other constraints. Spintronics—particularly those leveraging multiferroic and magnetoelectric effects—has emerged as a promising alternative to address these challenges. This approach capitalizes on the electron's spin, in addition to its charge, for processing and storing information [1–7].

Among the innovative device concepts, multiferroic tunnel junctions (MFTJs) stand out as particularly promising. These devices incorporate a ferroelectric barrier sandwiched between two ferromagnetic electrodes, offering a path toward energy-efficient, high-density, non-volatile, and multifunctional memory and logic systems [8–12]. The MFTJ concept merges two ideas: the magnetic tunnel junction (MTJ), which consists of two ferromagnetic layers [13,14], and ferroelectric tunneling, which uses a ferroelectric rather than a conventional dielectric barrier [15–17].

In an MTJ, the spin-dependent tunneling current varies with the alignment of the magnetizations of the two electrodes, resulting in the tunneling magnetoresistance (TMR) effect. According to Julliere's model [13], the TMR ratio between parallel and antiparallel magnetic alignments is given by:

$$TMR = (R_{AP} - R_P)/R_P = 2P_1 P_2 / (1 - P_1 P_2), \quad (1)$$

where R_P and R_{AP} are the resistances in the parallel and antiparallel states, and P_1 and P_2 represent the spin polarizations at the two interfaces.

Conversely, in a ferroelectric tunnel junction (FTJ), the tunneling current is influenced by the direction of the ferroelectric polarization in the barrier, giving rise to the tunnelingelectroresistance (TER) effect under specific conditions [15–17]. The TER ratio is expressed as:

$$TER = |R_\uparrow - R_\downarrow| / \min(R_\uparrow, R_\downarrow), \quad (2)$$

where $R_\uparrow$ and $R_\downarrow$ correspond to the resistances of the ferroelectric polarization states pointing up and down, respectively.

In MFTJs, the coexistence of TMR and TER effects enables four distinct, non-volatile resistance states. These states can be controlled via external electric and magnetic fields, offering a novel and efficient way to realize high-density memory technologies.

Beyond simply offering more resistance states, multiferroic tunnel junctions (MFTJs) provide capabilities that go well beyond a simple combination of magnetic tunnel junctions (MTJs) and ferroelectric tunnel junctions (FTJs). Alongside the ability to regulate spin and charge tunneling currents through ferromagnetic and ferroelectric polarization, the interface between the ferromagnetic and ferroelectric layers can induce strong magnetoelectric coupling effects. These effects can stem from phenomena such as spin-dependent screening at the interface [18,19], modifications in bonding strength [20], or shifts in ionic oxidation states influenced by the ferroelectric polarization [21,22].

If this interfacial coupling is sufficiently strong, reversing the ferroelectric polarization can significantly impact magnetic properties, including magnetic anisotropy, coercivity, and even the magnetic structure at the interface. This strong magnetoelectric interaction opens up a low-power method to control magnetization through voltage—offering an alternative to traditional techniques like spin-transfer torque [23] or electric-field-tuned interfacial perpendicular magnetic anisotropy in MTJs [24,25].

Furthermore, this interfacial coupling could allow the electrical control of spin polarization, which is a highly sought-after feature in spintronics. Since the tunneling magnetoresistance (TMR) is directly related to the spin polarization at the electrode/barrier interfaces (as defined by Eq. 1), Garcia et al. introduced the concept of tunnelingelectromagnetoresistance (TEMR) [26] to quantify how ferroelectric polarization reversal can influence spin polarization:

$TEMR = |TMR_{\uparrow} - TMR_{\downarrow}| / |\min(TMR_{\uparrow}, TMR_{\downarrow})|$,

where $TMR\uparrow$ and $TMR\downarrow$ refer to the TMR ratios for upward and downward ferroelectric polarization states, respectively.

In 2007, Gajek et al. [27] observed the TER effect in a $La_{2/3}Sr_{1/3}MnO_3/La_{0.1}Bi_{0.9}MnO_3$/Au spin filter junction utilizing a naturally multiferroic $La_{0.1}Bi_{0.9}MnO_3$ barrier, achieving TMR of ~81% and TER of ~20%. Then, in 2010, Garcia et al. [26] provided the first experimental evidence for ferroelectric control of spin polarization in a Fe-indented $La_{0.7}Sr_{0.3}MnO_3/BaTiO_3$ MFTJ, reporting TMR values of ~−45% and ~−19% for opposite polarization states, along with TER ~37% and a striking TEMR of ~450%.

Subsequent studies showed that the sign of the TMR could be reversed by switching the ferroelectric polarization in MFTJs that used 3d ferromagnetic metals (e.g., Co, NiFe) as top electrodes. Examples include $La_{0.7}Sr_{0.3}MnO_3/PbZr_{0.2}Ti_{0.8}O_3$/Co [28,29] and $La_{0.7}Sr_{0.3}MnO_3/BaTiO_3$/NiFe junctions [30]. $La_{0.7}Sr_{0.3}MnO_3$ is often chosen as the bottom electrode due to its good lattice compatibility with oxide ferroelectrics and its half-metallic nature [31]. The use of 3d ferromagnetic metals for the top electrode is largely driven by their ease of integration into nanoscale structures.

However, MFTJs using 3d metals typically display relatively low TMR values—often under 20% for one or both polarization states—mainly because of the low spin polarization of these metals and potential interdiffusion at the interface. As a result, such devices seldom exhibit the well-defined switching behavior typical of MTJ-based memory and are not yet ready for practical device applications.

Higher TMR values and more well-defined resistance (R) versus magnetic field (H) switching loops have been achieved in MFTJs that utilize two half-metallic perovskite electrodes. Examples include:

- $La_{0.67}Sr_{0.33}MnO_3/BiFeO_3/La_{0.67}Sr_{0.33}MnO_3$ (TMR ~ 69%, TER ~ 40%) [32],
- $La_{0.7}Ca_{0.3}MnO_3/Ba_{0.5}Sr_{0.5}TiO_3/La_{0.7}Ca_{0.3}MnO_3$ (TMR ~ 300%, TER ~ 160%) [33],
- $La_{0.7}Sr_{0.3}MnO_3/BaTiO_3/La_{0.5}Ca_{0.5}MnO_3/La_{0.7}Sr_{0.3}MnO_3$ (TMR ~ 100%, TER ~ 104%) [34],
- $La_{0.7}Sr_{0.3}MnO_3/BaTiO_3/La_{0.7}Sr_{0.3}MnO_3$ (TMR ~ 65%, TER ~ 125%) [21,35,36],
- $La_{0.7}Sr_{0.3}MnO_3/SrTiO_3/BaTiO_3/La_{0.7}Sr_{0.3}Mn_{0.8}Ru_{0.2}O_3$ (TMR ~ 30%, TER ~ 128%) [37],
- $La_{0.7}Sr_{0.3}MnO_3/Pb(Zr_{0.3}Ti_{0.7})O_3/La_{0.7}Sr_{0.3}MnO_3$ (TMR ~ 50%, TER ~ 100%) [22],
- $La_{0.7}Sr_{0.3}MnO_3/BaTiO_3/La_{0.84}Sr_{0.16}CuO_{3-x}/La_{0.7}Sr_{0.3}MnO_3$ (TMR ~ 125%, TER ~ 104%) [38],
- and $LaNiO_3/Pr_{0.8}Ca_{0.2}MnO_3/BaTiO_3/La_{0.7}Sr_{0.3}MnO_3$ spin filter MFTJ (TMR ~ 24%, TER ~ 100%) [39].

Year	Substrate	Bottom electrode	Tunnellingbarrier	Topelectrode	Max(TMR, TER, TEMR)	T_C^{TMR}
2007 [27]	$SrTiO_3$	$La_{2/3}Sr_{1/3}MnO_3$	$La_{0.1}Bi_{0.9}MnO_3$(1.2nm)	Au	(81%,20%,n/a)at 3K	60 K

2010 [26]	$NdGaO_3$	$La_{0.7}Sr_{0.3}MnO_3$	$BaTiO_3$(1.2nm)	Fe	(-45%,37%, 450%) at4.2K	>4.2 K
2010 [32]	$SrTiO_3$	$La_{0.67}Sr_{0.33}MnO_3$	$BiFeO_3$(3nm)	$La_{0.67}Sr_{0.33}MnO_3$	(69%,40%,13%) at80K	>80 K
2011 [33,40]	$SrTiO_3$	$La_{0.7}Sr_{0.3}MnO_3$	$Ba_{0.95}Sr_{0.05}TiO_3$(2nm)	$La_{0.7}Sr_{0.3}MnO_3$	(1.1%,2%,8.5%) atRT	>RT
2011 [41]	$NdGaO_3$	$La_{0.7}Sr_{0.3}MnO_3$	$BaTiO_3$(1.2nm)	Co	(-20%,80%,82%) at4K	>4K
2012 [28]	$SrTiO_3$	$La_{0.7}Sr_{0.3}MnO_3$	$PbZr_{0.2}Ti_{0.8}O_3$(3.2nm)	Co	(7.5%,700%, 250%)at10K	250K
2013 [34,42]	SrTiO3	La0.7Sr0.3MnO3	BaTiO3 (3 nm)/ La0.5Ca0.5MnO3 (1-5 uc)	La0.7Sr0.3MnO3	(180%, 104%, 450%) at 80 K	180 K
2014 [43]	LSAT	La0.6Sr0.4MnO3	BiFeO3 (10 nm)	La0.6Sr0.4MnO3	(6.6%, 15.5%, 18%) at 10 K	100 K
2014 [44]	LSAT	La0.67Sr0.33MnO3	BaTiO3 (2.8 nm)	Co	(20%, 104%, 100%) at 10 K	> 10 K
2015 [29]	SrTiO3	La0.7Sr0.3MnO3	PbTiO3 (3.2 nm)	Co	(30%, 350%, 15%) at 5 K	140 K
2015 [30]	SrTiO3	La0.7Sr0.3MnO3	BaTiO3 (5 nm)	Ni0.81Fe0.19	(0.3%, 1500%, 300%) at 8 K	> 8 K
2015 [21,35]	SrTiO3	La0.7Sr0.3MnO3	BaTiO3 (3 nm)	La0.7Sr0.3MnO3	(82%, 125%, 127%) at 80 K	160 K
2015 [37]	SrTiO3	La0.7Sr0.3MnO3	BaTiO3 (6 uc)/SrTiO3 (4 uc)	La0.7Sr0.3Mn0.8Ru0.2 O3	(30%, 128%, 100%) at 10 K	> 10 K
2016 [38]	SrTiO3	La0.7Sr0.3MnO3	BaTiO3(6 nm)/La0.84Sr0.16CuO3-x (2 nm)	La0.7Sr0.3MnO3	(125%,104%, n/a)	n/a
2016 [22]	SrTiO3	La0.7Sr0.3MnO3	PbZr0.3Ti0.7O3 (4 nm)	La0.7Sr0.3MnO3	(40%, 100%, 170%) at 80 K	> 80 K
2016 [45]	SrTiO3	La0.6Sr0.4MnO3	PVDF (26 nm)	Co	(15%, 75%, 288%) at 10 K	120 K

2016 [39]	SrTiO3	La0.6Sr0.4MnO3	BaTiO3 (5 uc)/ Pr0.8Ca0.2MnO3 (9 uc)	LaNiO3	(24%, 100%, 167%) at 10 K	> 10 K
2017 [36]	SrTiO3	La0.7Sr0.3MnO3	BaTiO3(4.4 nm)	La0.7Sr0.3MnO3	(20%, 103%, n/a) at 14 K	> 14 K
2017 [46]	Si	Ni50.3Mn36.9Sb12 .8	SrTiO3 (2 nm)/PbZr0.52Ti0.48O3 (3 nm)	Ni50Mn35In15	(39%, 487%, 224%) at RT	>RT
2017 [47]	Si	Ni50.3Mn36.9Sb12 .8	BiFeO3 (4 nm)	Ni50.3Mn36.9Sb12.8	(13%, 33%, 17%) at RT	>RT

All these experimental studies on engineered multiferroic tunnel junctions exhibiting both TER and TMR effects are summarized in Table 1. This table also includes the peak values for TMR, TER, and TEMR, the temperature above which TMR and the four-state memory effect vanish (Tc_{TMR}), and notes on the availability of data (n/a indicating missing information). Room temperature results are marked as RT, and entries are sorted chronologically by publication date.
For comprehensive reviews on MFTJs, readers can refer to previously published works [8–12].

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