Materials for a Sustainable Microelectronics Future: Electric Field Control of Magnetism with Multiferroics

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Abstract | This article is written on behalf of many colleagues, collaborators, and researchers in the field of complex oxides as well as current and former students and postdocs who continue to enable and undertake cutting-edge research in the field of multiferroics, magnetoelectrics, and the pursuit of electric-field control of magnetism. What I present is something that is extremely exciting from both a fundamental science and applications perspective and has the potential to revolutionize our world, particularly from a sustainability perspective. To realize this potential will require numerous new innovations, both in the fundamental science arena as well as translating these scientific discoveries into real applications. Thus, this article will attempt to bridge the gap between fundamental materials physics and the actual manifestations of the physical concepts into real-life applications. I hope this article will help spur more translational research within the broad materials community.

Keywords: Energy efficiency in computing, Multiferroics, Magnetoelectric coupling, Thin films, Spin–orbit coupling

1 Introduction

Before I get into the specific topic of this paper, it would seem to make sense to give a broader perspective, especially given the sustainability theme of this special issue. In 2010, I was asked by Energy Secretary Steve Chu to join him in the US Department of Energy to lead and articulate the DOE Sunshot Initiative. The name "Sunshot" was coined to bring back memories of the original "Moonshot" Initiative, which led to the Apollo program and galvanized the U.S. into action in the space race. Sunshot was meant to galvanize a different generation, to focus on clean energy and in doing so solve the biggest problem of our lifetime, namely Climate Change. In 2010, solar electricity was about 5X more expensive than electricity from fossil fuel (for example, the levelized cost of electricity in the U.S was ~ 5 c/kWh in the wholesale market; compared to that solar was ~ 27 c/kWh, leading to the factor of 5X difference). The Sunshot challenge, therefore, was this: how does one bring the cost of solar electricity down by a factor of 5X? I note that a 5X reduction of the cost of any technology in a commodity market with small margins, is extremely difficult; indeed, this is one of the main reasons that the incumbent technologies are so difficult to displace since they have already built up the economies of scale and captured market share. Furthermore, the challenge from Secretary Chu was that we should use the power of Science and Technology to solve this rather than revert to policy pathways such as the "Feed-in Tariff" approach that was being implemented in Europe. Thus, we needed a holistic approach, bringing together innovations in the hardware side as well as in market transformations and manufacturing. The cost of solar electricity has come down dramatically over the last decade, reaching the Sunshot target well ahead of the originally set 2020 target. Of course, this dramatic drop in the cost of solar electricity was aided by the corresponding drop in the prices of solar panels due to large-scale manufacturing in China. Sunshot was thought to be a success of

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the Steve Chu administration, perhaps mainly for the fact that an aggressive target was set by the federal government and a clear game plan was articulated and executed. A broader impact of the Sunshot Initiative manifested itself during the recent transition to the Biden–Harris administration. I had the opportunity to participate in the transition team, focused on the Department of Energy. In doing so, I had the privilege of working with some amazing scientists, engineers, policymakers in the Energy Transition Team, on identifying the top priorities, given the administration's focus on Climate Change, eradicating the COVID-19 pandemic, creating a more environmentally just investment as well as build back jobs and manufacturing. Based on our learning from the Sunshot Initiative, we proposed a set of "Earthshots", now focused on large, tough problems in Energy and Climate Change. One of them focused on what the world of microelectronics could look like in a decade, specifically from an energy consumption perspective. The rest of this article tries to capture the science technology challenges required to bring the energy consumption down in Microelectronics. Of course, this is still a personal perspective, in terms of what materials can enable.

1.1 The Macro-systems Perspective

We begin the discussion from a broad, macro-systems perspective. Microelectronics components and systems form an ever-increasing backbone of our society. Computing devices have pervaded many parts of our daily life, for example through a host of consumer electronics systems, providing sensing, actuation, communication, and processing and storage of information. All of these are built upon an approximately $470B/year global market that is growing at a steady pace of 10–15% annually[1,2]. Many of these innovations started as materials research ideas, often first discussed within the hallways of the many physics and materials conferences worldwide. The emergence of a few new global phenomena will change this landscape dramatically. The first among them is the notion of the "Internet of Things" (IoT), which is the network of physical devices, vehicles, home appliances, and other items embedded with electronics, software, sensors, actuators, and connectivity which direct integration of the physical world into computer-based systems, resulting in efficiency improvements, economic benefits, and reduced human exertion,[3] illustrated schematically in Fig. 1[4]. Thus, it is not inconceivable that every modern building will be outfitted with
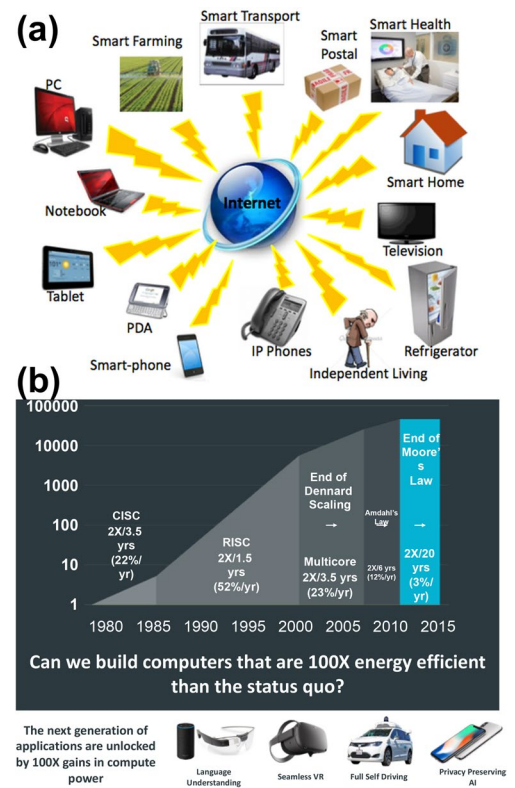

Figure 1: a A schematic illustrating the emergence of the "Internet of Things" and Machine Learning/Artificial Intelligence as macroscale drivers for the Beyond Moore's Law R&D[4]. **b** The leveling off the various scaling laws as a function of time, leading to the end of Moore's Law.

millions of sensors and actuators that can dynamically optimize the energy consumption dynamics of that building. Similarly, a modern automobile has many sensing, communicating components embedded. While still in its infancy, it is possible that driver-less automobiles, for example, will be a routine aspect of our life 20 years from now.

The second major phenomenon is the field of machine learning (ML)/artificial intelligence (AI) that is taking the technology world by storm. It uses a large amount of statistical data analytics which, in turn, provides the computing system the ability to "learn" and do things better as they learn, not unlike normal human beings. While there are several scientific disciplines that come into play, of relevance to us is the fact that microelectronic components are critical underpinnings for this field.

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J. Indian Inst. Sci.|VOL 102:1|489–511 January 2022|journal.iisc.ernet.in

(a) A new physical phenomenon is needed

Every ~ **20 years** a computing revolution is launched by a **physics & materials** driven 100x improvement in energy consumption.
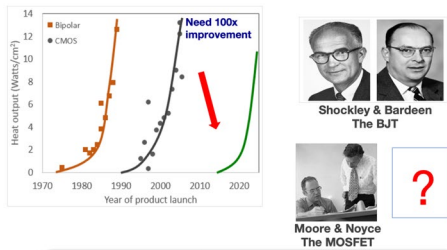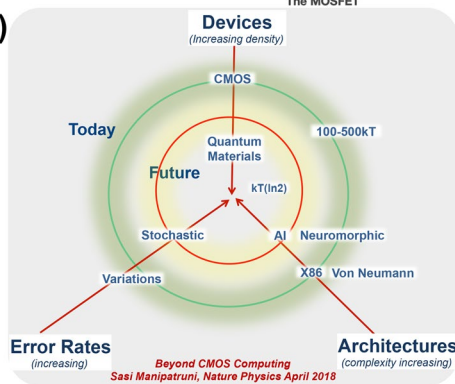

(b)

Figure 2: a The evolution of semiconductor electronics, from Bipolar to CMOS. As the technology has evolved, so has the heat output from the technology, which then leads to a new technology base. These innovations came from Shockley, Bardeen and then Moore and Noyce; the question mark poses the challenge: what comes after this; **b** three complementary pathways for Beyond CMOS electronics[1].

IT Challenge & Opportunity : Moore's Law, IoT & AI
Market size ~ $470B in 2019; growing rapidly

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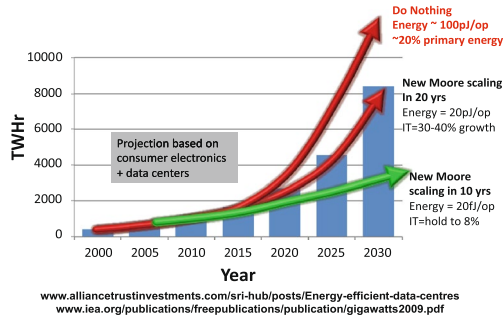www.iea.org/publications/freepublications/publication/gigawatts2009.pdf

Figure 3: Estimation of the total energy consumed in all of Microelectronics by 2030, if nothing is done to reduce the energy consumption/operation from the ~ 100pJ/operation level, while the number of microelectronic components is growing exponentially due to the emergence of the "Internet of Things" and Artificial Intelligence/Machine Learning (RED curves). A New Moore's Law at 20 femtoJoule/operation (GREEN curve) will enable us to keep the energy consumption level at the ~ 8% level[13].

1.2 Do We Need a New Paradigm for Computing?

We can now ask the question: how do these global phenomena relate to microelectronics and, more importantly, to new materials? Or stated in a different way, what can *materials physics* do to enable this coming paradigm shift? To put this into perspective, we now need to look at the fundamental techno-economic framework that has been driving the microelectronic field for more than five decades. This is the well-known "Moore's Law", which underpins the field of microelectronics through the scaling of CMOS-based transistors (Fig. 2). Broadly, it states that the critical dimensions of the transistor shrink by 50% every 18–24 months. At its inception, CMOS transistors were "macroscopic" with the critical dimension well over 1 μm. In 1974, a path to shrinking such transistors, at constant power density was proposed[5–7] and was followed for the next 30 + years. Today, however, this so-called Dennard scaling is no longer sustainable

while the critical dimensions of modern transistors are rapidly approaching sub-10 nm scales; the point at which both the fundamental science (*i.e.*, classical electron dynamics) no longer more suffices to adequately understand operation and ever more complex manufacturing issues must be addressed. In the past 5–8 years, there has been an ever-increasing sense that something has to be done about this issue.[8–12]

1.3 Energy Efficiency in Computing

As if this combination of challenges was not enough, we have yet to introduce perhaps the single most important aspect into consideration: energy consumption (Fig. 3)[13]. Of the many issues modern technologists must address, the one we highlight here has the potential to be the most impactful from a sustainability perspective, namely energy. The energy consumed per logic operation, which in today's CMOS transistor is of the order of 50–100 pJ/logic operation (note that this actual number may be debated, but it remains that the energy consumed is of the order of pJ/operation). For the sake of discussion, assume that there is no change to this number soon, but, at the same time, the demand for and consumption of microelectronic components in IoT and AI/ML will grow exponentially. Consequently, it is quite conceivable that the total energy consumption in all of microelectronics could grow
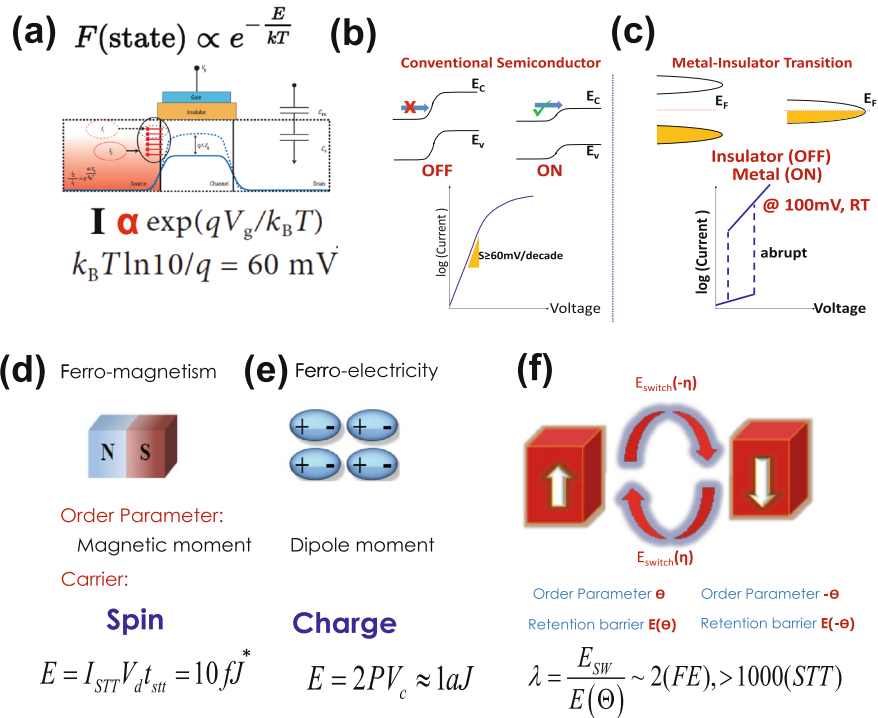
(a) $F(\text{state}) \propto e^{-\frac{E}{kT}}$

$$I \, \alpha \, \exp(qV_g/k_B T)$$
$$k_B T \ln 10/q = 60 \text{ mV}$$

(b) Conventional Semiconductor

OFF ON

$S \geq 60 \text{mV/decade}$

(c) Metal-Insulator Transition

Insulator (OFF)
Metal (ON)

@ 100mV, RT

abrupt

(d) Ferro-magnetism

Order Parameter:
Magnetic moment
Carrier:
Spin

$$E = I_{STT} V_d t_{stt} = 10 \, fJ^*$$

(e) Ferro-electricity

Dipole moment

Charge

$$E = 2PV_c \approx 1aJ$$

(f) $E_{switch}(-\eta)$

$E_{switch}(\eta)$

Order Parameter θ Order Parameter $-\theta$
Retention barrier $E(\theta)$ Retention barrier $E(-\theta)$

$$\lambda = \frac{E_{SW}}{E(\Theta)} \sim 2(FE), > 1000(STT)$$

Figure 4: a Schematic description of the Boltzmann distribution function for electrons in the CMOS channel, leading to the 60 mV/decade of current as the limit, which is known as the "Boltzmann Tyranny", shown in **b**[16]; **c** a possible manifestation of metal–insulator transition as the base for the next generation of logic; **d**, **e** the emergence of correlations (spin in (**d**) and dipolar in (**e**)) that can then be used to reduce the energy consumption in a memory-logic device; **f** schematic description the ratio of the energy required to switch a ferroelectric element compared to the barrier height[1].

to ~ 20% of primary energy by 2030[14]. Today, it is of the order of 5% and thus is not of great concern, especially in contrast to sectors such as buildings, which consume ~ 38% of the total energy consumption, or transportation which consumes ~ 24% (fractions noted here are for the United States). At the scale of ~ 20% of primary energy, microelectronics would become a serious component of the worldwide energy consumption mix and thus deserves to be addressed from the energy efficiency perspective as well. Thus, these three global phenomena, namely the emergence of IoT and AI/ML as well as the end of Moore's Law (including aspects of dimensional constraints and total energy consumption in microelectronics) form the backdrop for our discussion as we ask: what can we do with new materials physics?

1.4 The Opportunity for New Materials Science Leading to Technology

The microscopic behavior of the electronic charge in a CMOS transistor is governed by the Boltzmann distribution (Fig. 4)[15]. A quick analysis shows that the current changes exponentially with voltage, with a slope of 60 mV/decade of current,[16] termed as the "Boltzmann Tyranny"[1,11] since the Boltzmann physics is imposed on the functioning of the actual device. In real transistors, this voltage slope is typically larger. This fundamental behavior is central to the performance of the transistor, both in terms of the voltage required and the energy consumed in the process of operating the transistor. In recent years, there has been the realization that the Boltzmann Tyranny needs to be addressed—thus the need for new materials and materials phenomena. One proposed pathway is to use materials exhibiting a metal-to-insulator transition, such as in correlated-electron systems. Under ideal conditions, such a metal-to-insulator transition can be very abrupt. Another key realization, which is described in a recent review,[1] identifies the broad class of quantum materials as possible candidates to overcome this tyranny, mainly through the insertion of additional, internal interaction energies into the Boltzmann distribution. For example, this could
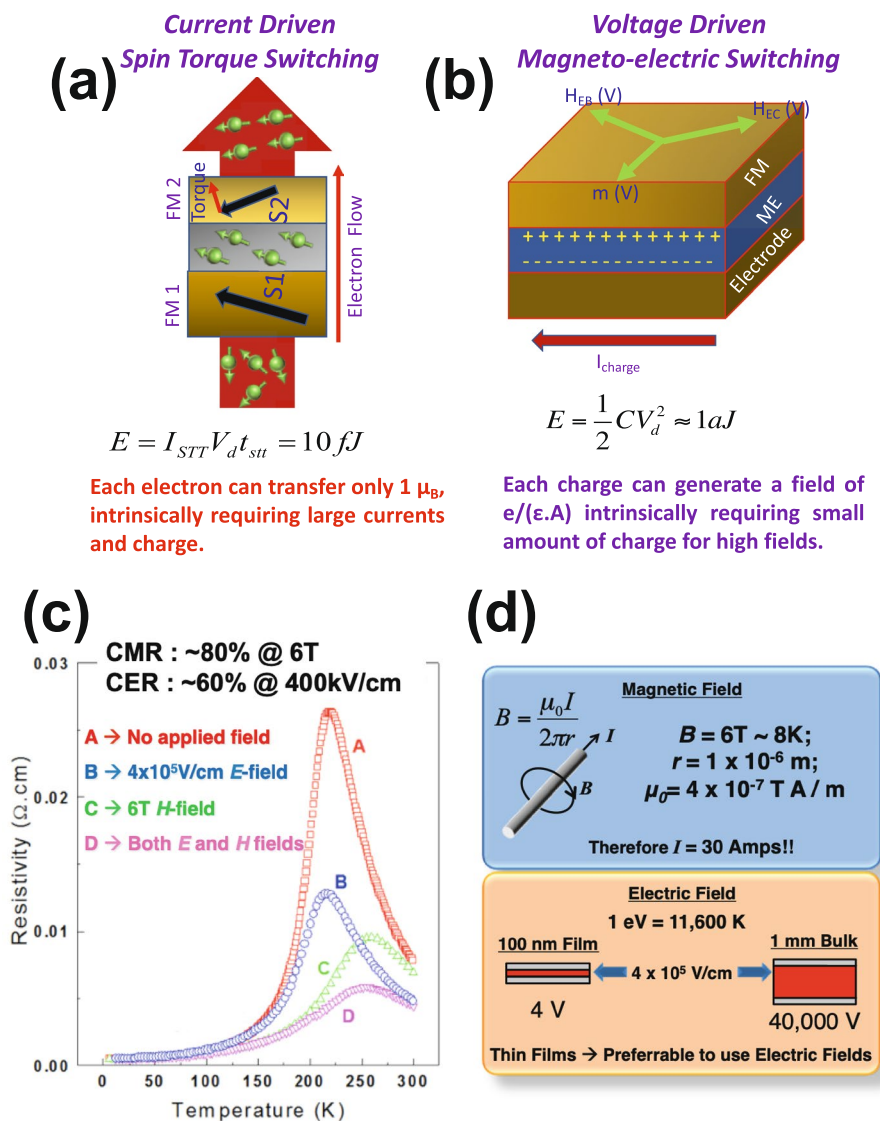
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(a) Current Driven Spin Torque Switching

$$E = I_{STT}V_d t_{stt} = 10\,fJ$$

Each electron can transfer only 1 μ_B, intrinsically requiring large currents and charge.

(b) Voltage Driven Magneto-electric Switching

$$E = \frac{1}{2}CV_d^2 \approx 1aJ$$

Each charge can generate a field of e/(ε.A) intrinsically requiring small amount of charge for high fields.

(c)

CMR : ~80% @ 6T
CER : ~60% @ 400kV/cm

A → No applied field
B → 4x10⁵V/cm *E*-field
C → 6T *H*-field
D → Both *E* and *H* fields

(d)

Magnetic Field

$$B = \frac{\mu_0 I}{2\pi r}$$

$B = 6T \sim 8K;$
$r = 1 \times 10^{-6}\,m;$
$\mu_0 = 4 \times 10^{-7}\,T\,A/m$

Therefore $I = 30$ Amps!!

Electric Field

1 eV = 11,600 K

100 nm Film ← 4 x 10⁵ V/cm → 1 mm Bulk

4 V   40,000 V

Thin Films → Preferrable to use Electric Fields

Figure 5: A set of schematics illustrating the energy consumption for nominal devices. **a** On the left is a current-driven spin torque switching device and **b** is a voltage driven magnetoelectric switch[18]. **c** The original data for the colossal magnetoresistance effect of ~ 80% at 6 T and a ~ 60% colossal electroresistance effect at an electric field of 400 kV/cm; **d** a simple calculation of the current required to create a magnetic field of 6 T at a distance of 1 μm from the center of the current-carrying wire while the bottom shows the calculation of the voltage required to create the 400 kV/cm electric field. This voltage scales with the dimensions of the object, while the magnetic field shows not scale with the dimensions of the object[19].

be the exchange interaction in a ferromagnet or the dipolar interaction in ferroelectrics. In its simplest form, such an interaction can be represented by an additional term in the Hamiltonian that represents the exchange interaction energy for a magnet given by $E_{ex} = -J \bullet S_1 \bullet S_2$, where J is the exchange integral and S_1 and S_2 are the two neighboring spins (or the corresponding dipolar energy in the case of a ferroelectric). This term then becomes the key component within the Boltzmann distribution function, and it modifies the energy landscape. In simpler terms, the exchange energy (or the dipolar energy in a ferroelectric) makes the spins (or the dipoles) align collectively *without the need* for an external source of energy. Thus, if one could use spin or a spontaneous dipole as the primary order parameter rather than merely the electronic charge in a CMOS device, one could take advantage of such internal collective order

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to reduce the energy consumption. Indeed, this is the premise behind two recent research articles,[1,10] where the rudiments of a possible magneto-electric spin orbit (MESO) coupled memory-logic device are discussed. While many parts of this device require further detailed study and innovations, one aspect that we will focus on, pertains to advanced materials and electric-field control of magnetism.

1.5 Energy Consumption at the Microscopic Scale

We can begin this discussion with a question: why would one use an electric field to control magnetism, when it would rather be straight-forward to use a magnetic field instead? The answer is energy. In fact, one can potentially reduce the energy consumption by as much as a few orders of magnitude using electric fields rather than magnetic fields. To illustrate this, we explore two possible scenarios. The first describes how a moving electron can create a spin torque, of interest in spin transfer torque (STT) based memory devices. The key is that each electron carries with it a moment of 1 μ_B and, therefore, generating a large enough spin torque to move domain walls or switch the magnetization requires many electrons (i.e., a large current), which, in turn, requires an appropriate current source (e.g., a battery). For a nominal device dimension (*e.g.*, 10×10 nm lateral dimensions) one can estimate the energy consumed in this process to be on the order of a few fJ (10^{-15} J); (Fig. 5a)[17]. One can contrast this to a capacitive device, with an electric field modulating the charge, where for a similar 10×10 nm device with a dielectric constant of ~100 (which is reasonable for ferroelectrics), one can generate relatively large fields of the order of 10 kV/cm with just about an aJ of energy (10^{-18} J, Fig. 5b)! Thus, although this looks like simple physics on the surface, there can be significant impacts for computing when one is either in an Artificial Intelligence (AI)/ Machine Learning (ML) or High-Performance Computing (HPC) environment.

We can now look at another interesting aspect of electric and magnetic fields, using the colossal magnetoresistive manganites as an example, Fig. 5c, d[18,19]. Figure 5c shows the resistivity–temperature plot for the La–Ca–Mn–O system, both as a function of magnetic field (6 T), (leading to colossal magnetoresistance) and electric field of 400 kV/cm, (leading to colossal
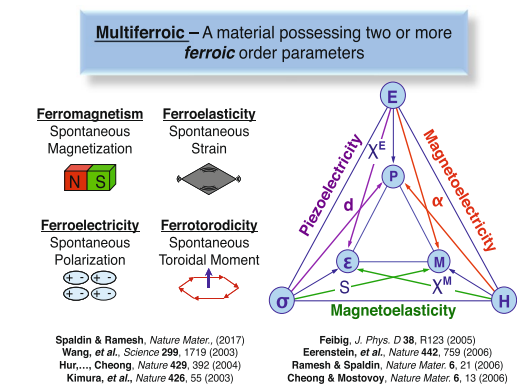

Figure 6: A schematic illustrating the 4 symmetry-based order parameters in solids. On the right is the "Nye-diagram" showing the coupling between the intrinsic and extrinsic thermodynamic variables[18].

electroresistance)[20]. Figure 5d schematically describes an estimate of the current required to generate this magnetic field of 6 T at a distance of 1 μm from a wire (in the BLUE box, using Ampere's Law), which is of the order of a few Amperes, pointing to a large energy consumption. In contrast, the electric field of 400 kV/cm can be generated by an applied voltage of 4 V across a 100 nm thick film (ORANGE box). The key to note is that the magnetic field does not scale with the size of the object (i.e., the field of 6 T is the same, whether the sample is a single macroscopic crystal or a thin film/nanostructure) while the electric potential (which is of greater interest from a practical perspective) required to get to a specific electric field scales with the dimensions of the object.

2 Multiferroics and Magnetoelectrics

With this as background, let us now explore the prospects and progress in electric-field control of magnetism with special attention to multiferroic and magnetoelectric materials. The pace and breadth of the work in this field means that it will be impossible for one manuscript to cover all the developments. Furthermore, this article is also somewhat of a personal perspective of the field; therefore, I would direct the reader to several excellent recent reviews[20–26] on this subject for complementary information on other aspects of such approaches. The manipulation of magnetic properties by an electric field in magnetoelectric multiferroic materials has driven significant research activity with the goal of realizing their transformative technological potential. Here, I
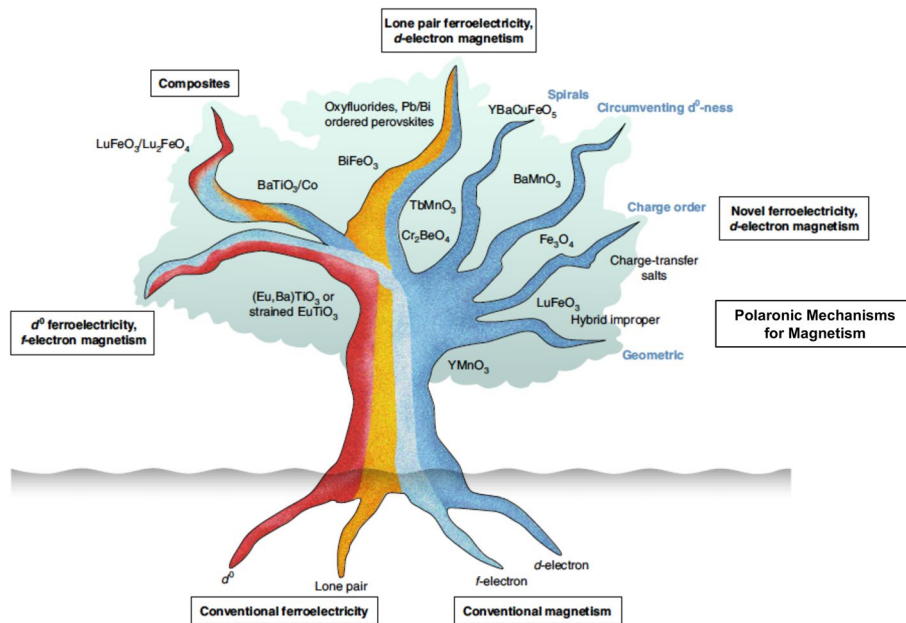
Figure 7: The "Multiferroic Tree" that depicts how one can design multiferroics from the basic elements of bringing together magnetic species (for example, ions with f/d-electrons) and polar species (i.e., chemical species that lead to the emergence of a spontaneous polarization). Each branch depicts exemplar multiferroic systems; the boxes on the outside identify the dominant mechanism responsible for the formation of multiferroics[21].

review progress in the fundamental understanding and design of new multiferroic materials, advances in characterization and modeling tools to describe them, and explore devices and applications. Focusing on the translation of the many scientific breakthroughs into technological innovations, I identify the key open questions in the field where targeted research activities could have maximum impact in transitioning scientific discoveries into real applications.

2.1 Symmetry and Fundamentals of Magnetoelectric Coupling

Before delving into specific magnetoelectrics and multiferroics and their applications, it is prudent to describe the symmetry basis for such materials (Fig. 6)[18]. Magnetism, for example, breaks time-reversal symmetry. A simple way to visualize this is a classic Amperean experiment in which a current is flowing through a metal wire wherein the corresponding magnetic field (B) is then given by the right-hand rule, as described in basic physics textbooks. The key symmetry breaking attribute of a magnet is that reversal of the current direction (which is also represented as reversing time), leads to a reversal of the sense of the magnetic field. A ferroelectric, on the other hand, breaks

spatial inversion symmetry (*e.g.*, a mirror plane is lost during the phase transition). In contrast to these two, a ferroelastic (such as martensite) breaks neither time nor spatial inversion symmetry. The apparent lack of a fourth component to complete this picture was recognized in the early years of multiferroics research,[27] leading to the addition of a fourth order parameter, namely a ferrotoroidic order (i.e., one which breaks both time and spatial inversion symmetry).

2.2 Multiferroic and Magnetoelectric Materials

Multiferroics exhibit more than one primary ferroic ordering (*i.e.*, ferromagnetism, ferroelectricity, ferroelasticity, or ferrotoroidicity) in the same phase[28]. This terminology is usually extended to include other types of order such as antiferromagnetism as well as composites of individual ferroics and is most often used today to refer specifically to magnetoelectric[29] materials combining ferroelectric and magnetic behavior. Importantly, the combination of ferroic orders in multiferroics can lead to coupling between them, so that one ferroic property can be manipulated with the conjugate field of the other, with particular focus on the prospect of switching the orientation of magnetization using an electric field[30,31].

Table 1: This table complements Fig. 7.

Pathway	Fundamental mechanism	Example systems
A-site driven	Stereochemical activity of lone pairs on A-site leads to ferroelectricity; magnetism from B-site	$BiFeO_3$; $BiMnO_3$
Geometrically driven	Long range dipole–dipole interactions and oxygen rotations breaks inversion symmetry	$YMnO_3$; $BaNiF_4$
Charge ordering	Non-centrosymmetric charge ordering leads to ferroelectricity in magnetic materials	$LuFe_2O_4$
Magnetic ordering	Ferroelectricity is induced by a lower symmetry ground state that lacks inversion symmetry	$TbMnO_3$; $DyMnO_3$
Atomically designed superlattices	Still under investigation; likely lattice mediated	$LuFeO_3$–$LuFe_2O_4$

It summarizes the various identified mechanisms for creating multiferroics

This coexistence and control of simultaneous ferroic order is challenging, requiring design of the electronic structure at the most fundamental level, new materials chemistries to implement them, the development of new tools to compute and characterize the novel properties associated with the coupled behaviors in parallel with new approaches to synthesize such materials with atomic-scale precision. When this is successful, it presents possible routes to entirely new device architectures, as exemplified by the MESO[10] device. This review focuses on the recent developments in these three aspects: basic science, experimental and theoretical methods, and applications of magnetoelectric multiferroics[3].

There are now many established routes to circumvent the "contradiction" between ferroelectricity (typically associated with ionic species with empty *d*-orbitals) and magnetism (typically associated with partially filled *d*-orbitals)[21,32]. Such a "bottoms-up" design is described in the "multiferroic-tree" (Fig. 7). A quick perusal of this figure shows that although there are several multiferroics, there is still a dearth of technologically viable multiferroics, *i.e.*, those that can be manipulated at room temperature. Thus, a more diverse palette of new materials with robust room temperature coupling of ferromagnetism and ferroelectricity is still urgently needed and indeed should be the focus of future interdisciplinary research. Table 1 provides a summary of the top five physical principles that have led to the discovery of several multiferroics. Of these, the two most studied are multiferroics in which the polar order comes from one of the crystal sites and the magnetic order is built into the other chemical site, as is the case in $BiFeO_3$ and $BiMnO_3$. The second type, which has received considerable interest from the physics community, is based on a polar order emerging because of a magnetic transition
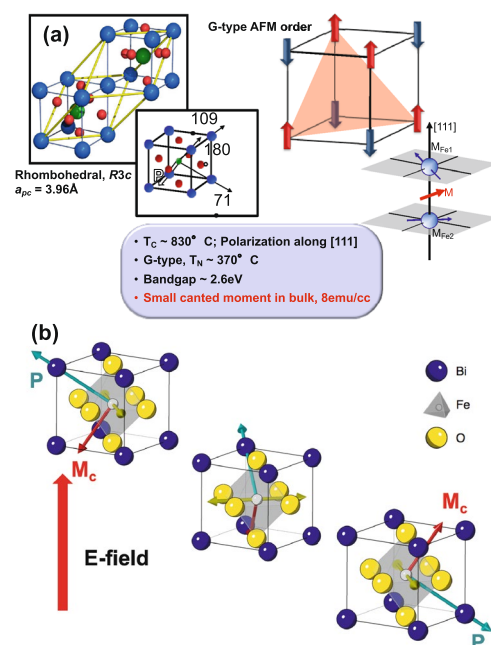

Figure 8: **a** A schematic illustration of the rhombohedral crystal structure of $BiFeO_3$ as well as the G-type antiferromagnetic order and the canted moment arising because of the Dzyaloshinskii–Moriya coupling; **b** schematically illustrates the 180-degree switching of the polar axis in 2 steps and the associated changes in the canted moment direction[36].

as, for example, in the manganites. An emerging third pathway is via the power of heteroepitaxy and superlattice design. We will use these as examples to explore both the fundamental materials physics of coupling as well as the potential for future applications.

Of the known multiferroics, bismuth ferrite, $BiFeO_3$, remains arguably the most important, and certainly the most widely studied, with

more than 6000 papers published over the last decade. The establishment of its large (~ 90 μC/cm^2) ferroelectric polarization, combined with magnetic ordering persisting well above room temperature[33] spawned an intense research effort that continues to unveil fascinating new physics and potential new applications[34] of this rhombohedrally distorted perovskite. (Fig. 8)[35]. While there was considerable debate regarding the spontaneous polarization (which was experimentally measured in epitaxial thin films and predicted theoretically to be ~ 90 μC/cm^2,[36]), this value of the spontaneous polarization in the rhombohedral phase is now well validated. Magnetism in BiFeO$_3$ is complex. Although the ground state is a robust G-type antiferromagnetic structure (which can be described by spins in {111} that are ferromagnetically coupled in-plane but antiferromagnetically coupled out of plane along the[109]), the magnetic structure is quite a bit more sophisticated. Due to the antisymmetric Dzyalozhinskii–Moriya coupling (which is governed by the crystal symmetry and is allowed for the $R3c$ crystal symmetry of BiFeO$_3$), a small, canted moment arises, which lies in the {111} (i.e., perpendicular to the spontaneous polarization direction). A further complication arises in single crystals in which this canted moment spirals about the [1-10] so that it does not exhibit a macroscopically measurable magnetic moment until this spin spiral is broken, either by elastic strain (for example, through epitaxial thin-film growth) or through the application of a magnetic field of ~ 16–18 T. Furthermore, domain walls can play a key role in the emergence of a magnetic moment, which typically manifests in the form of a spin glass.

2.3 Chemical Substitutions in Bismuth Ferrite

The role of isovalent and aliovalent chemical substitutions at both the bismuth and iron sites has been extensively studied. Outside of the fundamental understanding of the polar and magnetic order, these studies have also focused on the possibility of creating phase boundaries (much like the morphotropic phase boundary in the PbZr$_{1-x}$Ti$_x$O$_3$ family of ferroelectrics) that can lead to large piezoelectric responses and allow for tuning of the ferroelectric switching behavior[37,38]. Chemical substitutions at the Fe^{3+} site have mainly attempted to manipulate the antiferromagnetic nature (for example, manganese or cobalt substitution has been shown to enhance the magnetic moment)[39]. While these studies have indicated a certain degree of success, detailed studies of the magnetoelectric coupling in such alloyed BiFeO$_3$ materials are just emerging[40]. Indeed, a grand challenge would be to discover pathways to enhance the magnetic moment to ~ 50 emu/cc (the canted moment in pure BiFeO$_3$ is only ~ 6 emu/cc) while at the same time demonstrating magnetoelectric coupling (this issue is discussed more in the section on magnetoelectric coupling). Aliovalent substitutions at the Bi-site (for example with Ca^{+2})[41] destabilizes the polar state and the accompanying oxygen vacancies causes large field-dependent conduction and electro-coloration phenomena.

Thin-film synthesis of BiFeO$_3$ has been a very fruitful pathway to study the materials physics of magnetoelectric coupling as well as pointing the way to possible applications. The perovskite symmetry and lattice parameters (pseudocubic lattice parameter of 3.96 Å) means that many oxide-based substrates are available for epitaxial synthesis (albeit with varying degrees of lattice mismatch). Thin films with thicknesses down to just a few unit cells and as large as a few microns have been synthesized by physical-vapor deposition (e.g., pulsed laser deposition, sputtering, molecular beam epitaxy), chemical-vapor deposition,[42] and chemical-solution deposition. Many studies have used conducting perovskite electrodes (such as SrRuO$_3$, La$_{1-x}$Sr$_x$MnO$_3$, La$_{1-x}$Sr$_x$CoO$_3$) as bottom electrodes to both template the perovskite phase as well as provide a bottom contact for electrical measurements. These synthesis studies have led the way to enable a wide range of materials physics studies including thickness-size effects down to just a few unit cells[43]. Consistent with other perovskite ferroelectrics, a suppression of the magnitude of the polar order is observed, although both theory and experiments indicate that a polar state is stable down to even a couple of unit cells.

Like many ferroelectrics, BiFeO$_3$ is also quite susceptible to strain. Compressive strain, can be imposed, for example, through a substrate with a lattice parameter smaller than that of BiFeO$_3$ (for example, SrTiO$_3$ and LaAlO$_3$). Under a large (~ 5%) compressive strain, a super-tetragonal-like (T-like) structure, with an enhanced c/a ratio and an almost square pyramidal iron-oxygen coordination, has been shown to be the stable phase[44–46]. Both ab initio theory[47] and experiments[48] point to a huge spontaneous polarization of ~ 150 μC/cm^2, oriented along or close to the [001]. Intriguingly, when BiFeO$_3$ is grown on LaAlO$_3$, a mixed phase ensemble of a strained version of the parent rhombohedral (R)-like and super-tetragonal

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T-like structures is stabilized. This two-phase system has a giant extrinsic piezoelectric response because of applied electric fields changing the relative fractions of the two phases[49]. In addition, the R-like component has an enhanced magnetic moment that is not yet well understood[50]; a microscopic understanding of its magnetism would be valuable in further optimizing the magnetic properties of BiFeO$_3$. Under biaxial tensile strain, an orthorhombic phase, with ferroelectric polarization in the in-plane,[108] has been stabilized by growing BiFeO$_3$ on orthorhombic NdScO$_3$ (110) substrates[51]. This structural flexibility of BiFeO$_3$ reflects the large number of competing low-energy polymorphs[52–54] and points to further exploration of the rich *bulk* crystal chemical phase space in the Bi–Fe–O system[55]. It is likely that several other phases, with different ratios of the parent Bi$_2$O$_3$ and Fe$_2$O$_3$ oxides are "hidden" within this chemical phase diagram, and could be accessed, for example, by exploiting as-yet unidentified stacking sequences of Fe–O and BiO layers[56]. With on-going improvements in layer-by-layer synthesis techniques, I anticipate the identification of additional phases within this rich phase diagram over the next few years. In addition to its structural and magnetoelectric richness and large ferroelectric polarization,[57] several other intriguing and unexpected behaviors have been reported. These include a photovoltaic effect,[58] photocatalysis,[59] photostriction,[60] electrochromism,[61,62] and gas-sensing behavior[63].

While first-principles density functional theory (DFT) calculations remain the gold standard for understanding and predicting the properties of ferroelectrics and multiferroics, second-principles calculations are proving increasingly valuable in the study of larger scale systems, for example heterostructures, domain walls and defects, as well as longer timescales in molecular dynamics[64]. In second-principles calculations, an effective model Hamiltonian is constructed by a judicious choice of the essential physics, and the parameters of the model are extracted by fitting to DFT. Such effective models have been used successfully applied for many years to describe structural phase transitions of prototypical ferroelectrics[65,66] and recent extensions to include additional lattice degrees of freedom,[67] as well as magnetic interactions,[68] have extended their applicability to multiferroics. One length scale larger, a Landau–Ginzburg thermodynamic potential that includes both polar and antiferrodistortive distortions and their coupling to the magnetism has been successful in reproducing the bulk behavior of BiFeO$_3$ and offers great
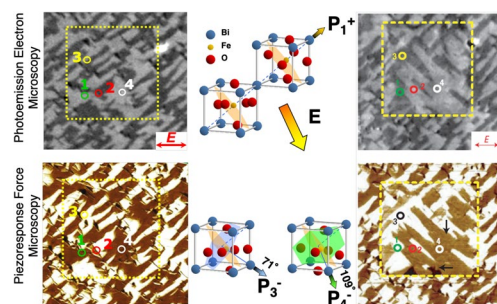

Figure 9: Electric field control of antiferromagnetism probed using XLDPEEM. On the left bottom is piezoforce microscope image showing the ferreoelectric domain structure before switching; the corresponding XLD-PEEM image (probing antiferromagnetism) is shown at the left top and the yellow box outlines the electrically switched area. The corresponding PFM/PEEM images after switching are shown to the right.

promise for predicting properties in thin film heterostructures and nanostructures[69].

3 Magnetoelectric Coupling
3.1 Magnetoelectric Coupling and Heterostructures

With this basic understanding of the order parameters and symmetry in systems such as BiFeO$_3$, we can now ask perhaps the most important question: how does magnetism couple to an electric field such that the state and direction of magnetism can be manipulated through the application of an electric field? In nature, this coupling between electricity and magnetism occurs through electromagnetism. However, to be able to dramatically change the state of magnetism with an electric field, it is desirable for the magnetoelectric coupling to be significantly stronger than what is available in nature.

Understanding the potential for electric-field control of antiferromagnetism requires probing the magnetism using X-rays or neutrons since direct magnetometry studies are not effective for antiferromagnets. Such studies of BiFeO$_3$ have shown that when the polarization state switches with the application of an electric field, there is a corresponding rotation of the magnetic order[70,71]. This can also be spatially probed using a combination of piezoresponse force microscopy (PFM) to image the ferroelectric order and X-ray magnetic linear dichroism (XLD) photoemission electron microscopy (PEEM) to image the antiferromagnetic order, (Fig. 9). It is interesting to note that there has been little detailed work on a
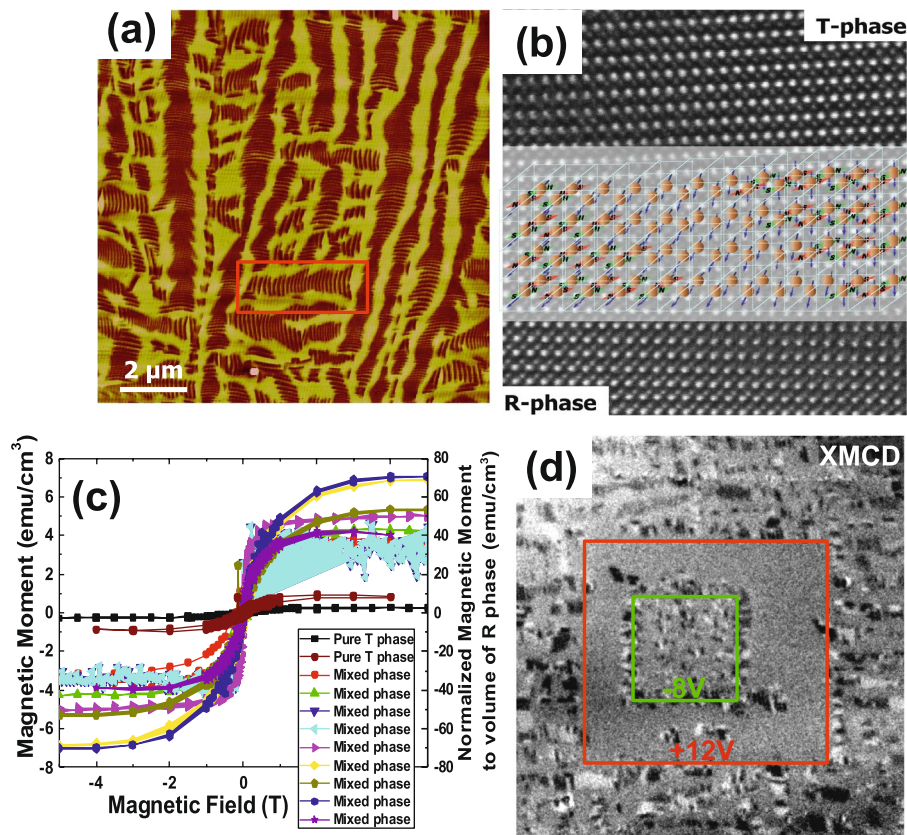
Figure 10: **a** An AFM image of the mixed phase $BiFeO_3$ on a $LaAlO_3$ substrate showing the striped of the rhombohedral (R) phase (in dark red) embedded epitaxially in a matrix of the tetragonal (T) phase (in yellow); **b** the corresponding atomic resolution TEM image of the interface between the T and R phases with a superimposed spin structure in the two phases; **c** the experimentally measured magnetic moment for a set of samples which exhibit the mixed phase behavior; also shown is data for a pure R and T phase film; the right side y-axis shows the moment normalized to the volume of the R-phase, indicating an enhancement in the moment of the constrained R-phase; **d** a Fe-XMCDPEEM image showing the switching of the magnetic moment in the R-phase.

full understanding of how the antiferromagnetic state is manipulated by an electric field with most studies assuming the magnetic order merely follows that of the polar order, but not clarifying that pathway. This is particularly surprising, since the antiferromagnetic resonance frequencies are in the several hundred GHz range and $BiFeO_3$ has ferroelectromagnons in the 700 GHz to 1 THz range. Given the current surge in interest in antiferromagnetic spintronics, I expect that such insulating multiferroics which exhibit electrical field controllable antiferromagnetism would also garner more interest, specifically from the high-frequency community.

Multiferroic coupling has been shown even in the mixed-phase versions of $BiFeO_3$ described above (Fig. 10a, b). This two-phase system has a giant extrinsic piezoelectric response because of applied electric fields changing the relative

fractions of the two phases[49]. Using X-ray magnetic circular dichroism-based photoemission electron microscopy coupled with macroscopic magnetic measurements, it was found that the spontaneous magnetization of the rhombohedral-like phase in these mixed-phase samples is significantly enhanced above the canted antiferromagnetic moment in the bulk phase, because of a piezomagnetic coupling to the adjacent tetragonal-like phase and the epitaxial constraint (Fig. 10c)[72]. Subsequently, reversible electric-field control and manipulation of this magnetic moment at room temperature was also shown (Fig. 10d).

3.2 Electric Field Control of Mixed Magnetic States and Nanocomposites

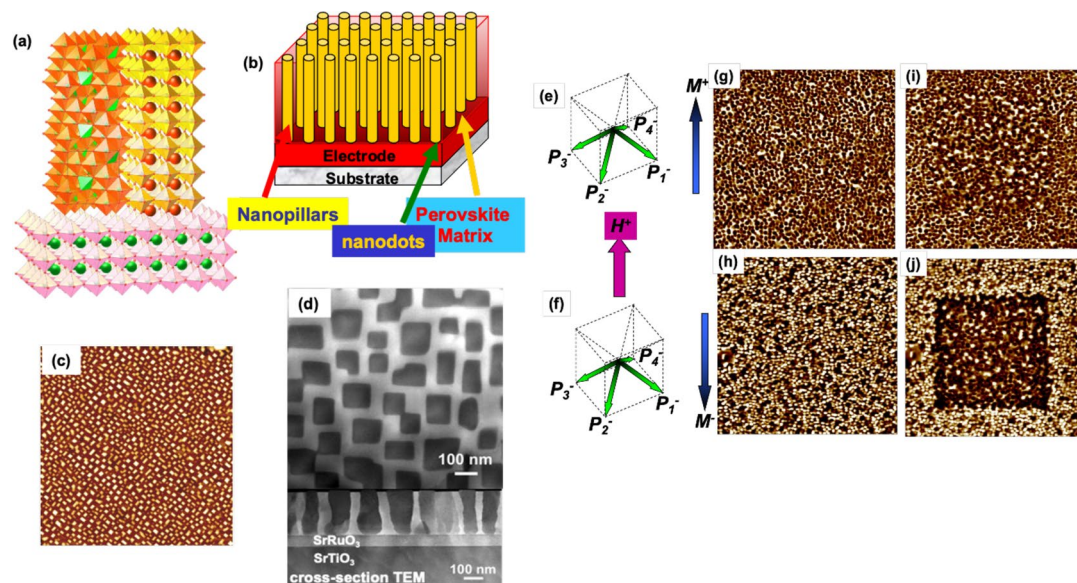It was realized early on[73] that nanocomposites comprised of ferrimagnets embedded (in many

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Figure 11: a A schematic of the 3-D vertically epitaxial magnetoelectric nanocomposite[73]; **b** AFM image of the ferrimagnetic CoFe2O4 nanopillars (in bright contrast embedded in a ferroelectric BiFeO$_3$ matrix (in dark contrast); **c** AFM image of the nanopillars in the BFO matrix; **d** TEM images (planar and cross section) of the interface between the spinel ferrimagnet and the perovskite ferroelectric; **g** magnetic force microscopy (MFM) images after magnetization at − 2 T (nanopillars in dark contrast), and **h** after magnetizing at 2 T, in which the ferrimagnetic nanopillars appear in bright contrast; **i** is the corresponding MFM image after the matrix was switched with a -16 V applied with an AFM tip[74].

cases epitaxially) in a ferroelectric/piezoelectric matrix could lead to efficient magnetoelectric coupling controlled by interfacial epitaxy, very much like structural alloys (e.g., Ni-based super-alloys). Such nanocomposites, exemplified by single crystalline nanopillars (Fig. 11a, b) of ferrimagnetic spinels (e.g., CoFe$_2$O$_4$) embedded epitaxially in a ferroelectric perovskite matrix (e.g., BiFeO$_3$), are illustrated in the AFM image (Fig. 11c). The epitaxial nature of the lateral interfaces is shown in corresponding planar section TEM images (Fig. 11d). Electric field-driven switching studies of such nanocomposites reveal perhaps the most interesting aspects of relevance to deterministic switching of the magnetic state with an electric field, that is, while the initially magnetized state (Fig. 11e, f) can be switched with an electric field, only ~ 50% of the magnetic nanopillars switch their state (for example, from magnetization pointing up to down; Figs. 11g-j). Detailed analysis of these data[74] revealed that this is indeed true and arises from the fact that the electric field assists in manipulating the magnetic anisotropy of the ferrimagnetic nanopillar. However, the magnetic anisotropy of the nanopillar is the same whether it is magnetized up or down (along the long axis of the nanopillars), thus leading to an ~ 50/50 mixture of up/down states after

the electric field manipulation. This study also throws light on the most important physics of such coupling phenomena, namely that manipulating the magnetization direction in a deterministic fashion, for example by 180°, requires that there be a field that breaks time-reversal symmetry. Said another way the electric field and corresponding piezoelectric stress that is generated does not break time-reversal symmetry and thus cannot deterministically switch the magnetization direction. But application of a small magnetic field to the nanopillar arrays during the electric field induced switching event leads to a complete switching of their magnetic state. These results also point to the need for extreme care in doing such experiments and the need to have a coupling mechanism that is magnetic in nature, for example, interfacial exchange bias coupling, which we focus on next.

3.3 Electric-Field Control of Magnetic Orientation Through Interfacial Exchange Coupling

Perhaps the most significant breakthrough in the past few years is the demonstration that the magnetization in conventional ferromagnets (e.g., Co$_{1−x}$Fe$_x$) can be rotated by 180° by

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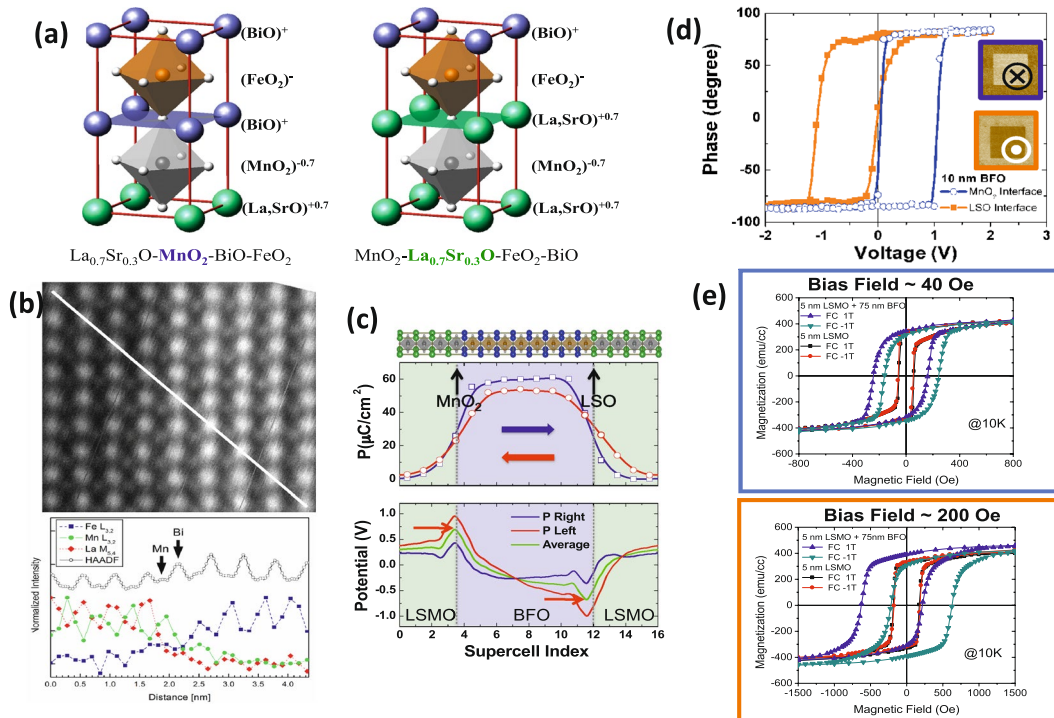
Figure 12: **a** Crystal structure model of the LSMO/BFO interface for the Bi–O and (La,Sr)–O interface termination; **b** an atomic resolution image of the LSMO/BFO interface with the corresponding EELS scan across this interface; **c** is a calculated plot of the polarization and the interface potential for the two types of interfaces; **d** piezoforce microscopy (PFM) phase angle as a function of voltage for the two types of interfaces, showing the build-up of an interface potential due to the termination[77]; **e** depiction of how the exchange coupling at the interface changes with the termination (measured at 10 K)[80].
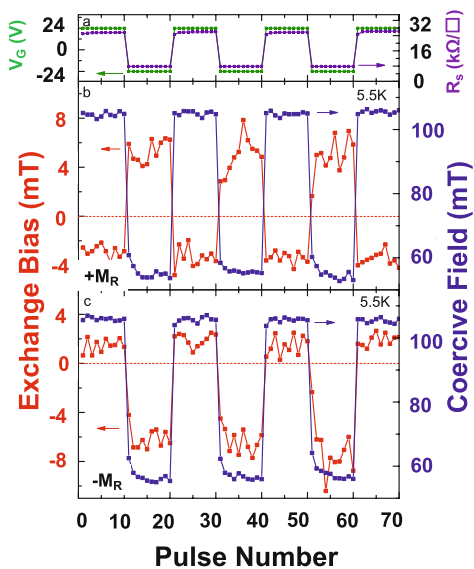
Figure 13: **a** A schematic of the bipolar voltage pulses imposed on a LSMO/BFO test structure; **b**, **c** bipolar modulation of the exchange bias field as a function of the electric field polarity; **b** is the LSMO set into the +MR state while **c** is for the LSMO set into the -MR state.

an electric field[75,76] when it is exchange coupled to BiFeO₃. The extension to all-oxide La₀.₇Sr₀.₃MnO₃/BiFeO₃ interfaces (Fig. 12), with chemically abrupt *A*-site termination,[77,78] allowed for electric-field control of exchange bias coupling, albeit at temperatures below 100 K[79,80].

Earlier work on this system has shown the ability to reversibly switch between two exchange bias states with the same polarity (unipolar modulation) and with opposite polarity (bipolar modulation) without the need for any magnetic field (Fig. 13).

Despite the exciting advances, an important open problem is the development of oxide ferro- or ferri- magnets with high T_c, a significant remanent moment and strong exchange coupling and Ohmic contacts with BiFeO₃ or another multiferroic. Spinels or double perovskites are promising candidates in this regard. In a complementary direction, the antiferromagnetic domain orientation in magnetoelectric Cr₂O₃, which can be controlled by an electric field, has been shown to affect the exchange-bias
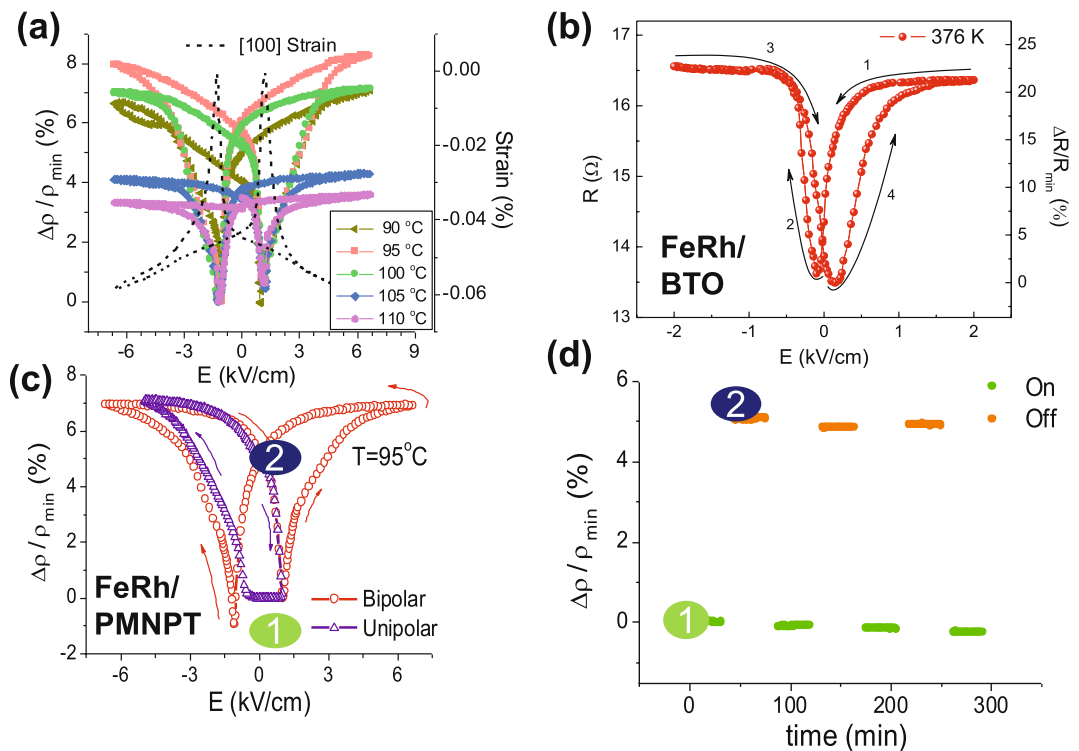
Figure 14: A Normalized resistivity change in the Fe$_{1-x}$Rh$_x$ layer as a function of electric field applied to the PMN-PT substrate; the corresponding strain in the PMN-PT layer is shown in dotted lines; **b** another example of the modulation of the Fe$_{1-x}$Rh$_x$ resistivity using a BaTiO$_3$ single crystal; **c** an example of the normalized change in the resistivity of the Fe–Rh layer showing the existence of two nonvolatile states that are captured in the time-dependent measurements in **d**[84–86].

coupling to a ferromagnetic overlayer[81] opening a pathway to electric-field switchable exchange-bias devices. If this could be achieved at room temperature, this would be truly revolutionary. Interfacial exchange coupling typically scales with the Curie temperature of the ferromagnet (generally the exchange bias vanishes at ∼ 0.25 Tc); for LSMO, with a Tc of 380 K, the exchange coupling vanishes around 100 K. Therefore there is currently a push to explore oxide ferromagnets with a Tc significantly higher than LSMO.

3.4 Electric-Field Control of Magnetic State

In parallel to these efforts to control the orientation of magnetization with an electric field using multiferroics such as BiFeO$_3$, there have been successes in electric-field manipulation of the magnetic state, for example, in changing between ferromagnetism and antiferromagnetism using composite systems as well. One example is electric-field modification of the magnetic exchange interactions in magnetic

Fe$_{1-x}$Rh$_x$ heterostructured with a piezoelectric $(1-x)$Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3 - x$PbTiO$_3$ (PMN-PT). Motivated by the long-known volume collapse at the ferromagnetic to antiferromagnetic transition in Fe$_{1-x}$Rh$_x$,[82,83] an electric field was used to drive the reciprocal effect, a ferromagnet-to-antiferromagnet transition induced by a structural deformation[84–86] (Fig. 14). Since the resistivities of the two magnetic phases differ, the magnetic transition is accompanied by a ∼ 25% change in film resistivity. Open challenges include reducing the optimal working temperature from around 100 °C to room temperature, tuning the chemical composition to optimize the strengths of the exchange interactions, achieving complete conversion between the ferromagnetic and antiferromagnetic phases, and reducing the required applied voltages. Other promising systems are the Mn–Pt intermetallics and half-doped perovskite manganites such as La$_{0.5}$Sr$_{0.5}$MnO$_3$, in which an electric-field-driven charge-ordered antiferromagnetic insulator to ferromagnetic metal transition could be possible.

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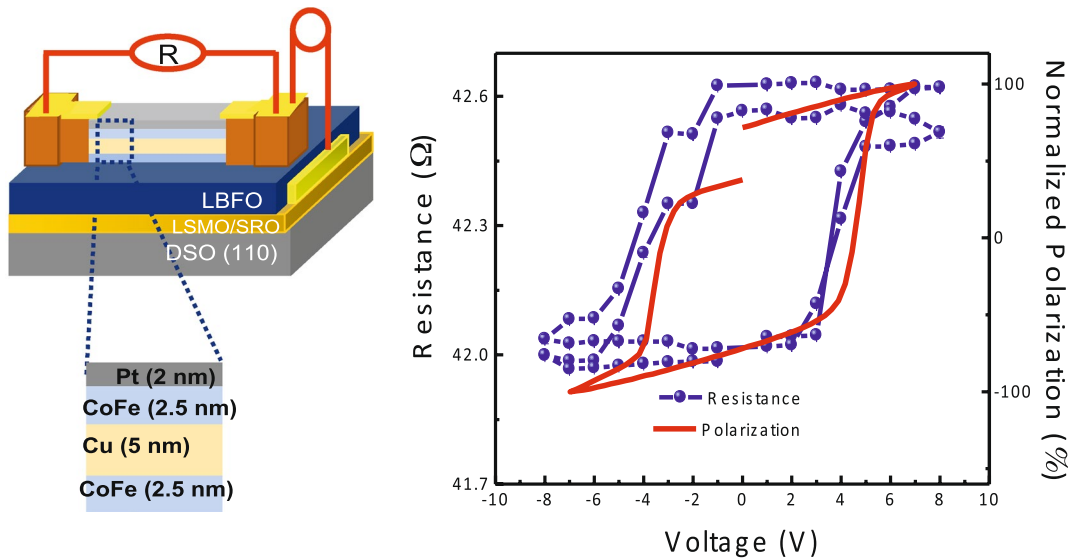J. Indian Inst. Sci.|VOL 102:1|489–511 January 2022|journal.iisc.ernet.in

Figure 15: a A schematic of the test structure used to probe the electric field-dependent manipulation of the magnetic state in a CoFe–Cu–CoFe spin valve; **b** the corresponding voltage-dependent magnetoresistance of the spin valve (BLUE), superimposed on the ferroelectric polarization-voltage curve for the BiFeO$_3$ layer (RED).
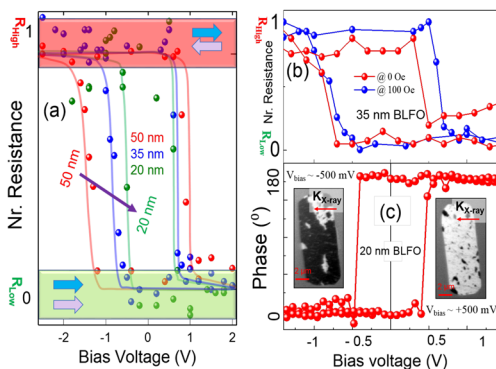

Figure 16: a Voltage-dependent GMR hysteresis as a function of La–BFO thickness from 50 nm down to 20 nm; **b** the normalized resistance of the GMR stack as a function of applied voltage at zero field (RED) and at 100Oe (BLUE); **c** the corresponding piezoelectric phase data showing switching of the polar state at ~500 mV for the 20 nm LBFO layer; also shown are the corresponding XMCD-PEEM (at the Co-edge) for a CoFe dot that has been switched (from BLACK to WHITE) with a bias of 500 mV.

4 Ultralow Power Logic-Memory Devices Based on Multiferroics

The push for ultra-low power logic-memory devices builds from seminal observations of the potential of magnetoelectric control using multiferroic-based devices the key being the ability to control magnetism with electric field at room temperature. An enabling observation was put forth in 2014, where deterministic reversal of the Dzyaloshinskii–Moriya (DM) vector and canted moment of BiFeO$_3$ using an electric field at room temperature was demonstrated[87,88]. First-principles calculations revealed a two-step switching process that enabled rotation of the magnetic order. This was exploited to demonstrate an energy-efficient control of a spin-valve device at room temperature (Fig. 15). The energy per unit area required for operation was approximately an order of magnitude less than that needed for spin-transfer torque switching. Building from such observations, a promising recently developed device architecture from Intel[89] combines two key recent discoveries, the inverse Rashba–Edelstein (spin-Hall) effect (IREE)[90–92,106] and electric-field control of magnetism. The resulting magnetoelectric, spin–orbit coupled logic device, MESO,[10] uses the IREE effect to convert spin to charge (or voltage) and the multiferroic to perform the opposite conversion of charge to spin. Success of the device rests on an increase in the IREE voltage output from current values of hundreds of μV to hundreds of mV as well as a reduction in voltage requirement for the magnetoelectric component from the current (~5 V) down to ~100 mV. Such breakthroughs could lead to a transformative 1 aJ (10^{-18} J) per memory bit or logic element.

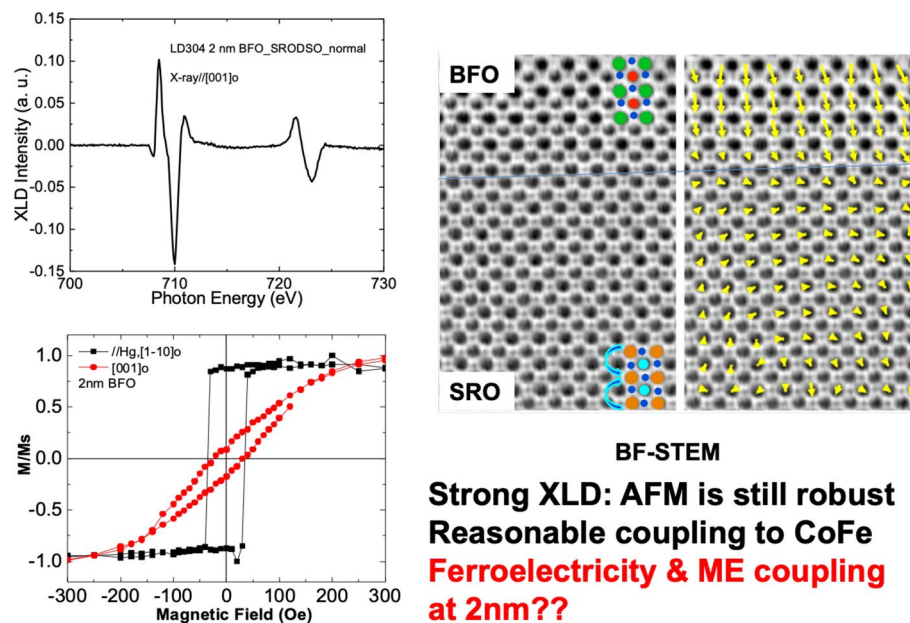Efforts in this direction are currently being undertaken. For example, magnetoelectric

Figure 17: a A X-ray linear dichroism plot between in-plane and out of plane xray polarization for a 2 nm BFO thin film, showing clear dichroism, indicative of a strong antiferromagnetic order; **b** the M–H loops for a CoFe layer that is in contact with the 2 nm BFO layer; the strong anisotropy between the orthogonal in-plane directions, [1-10]o and [001]o is indicative of strong exchange coupling; **c, d** HAADF-STEM images of the 2 nm (5 unit cells) BFO layer; the vector map (in yellow) shows that the polarization vector points towards the bottom SRO electrode.

switching of a magnetoresistive element was recently shown to operate at or below 200 mV, with a pathway to get down to 100 mV[93]. A combination of phase detuning is utilized via isovalent lanthanum substitution and thickness scaling in multiferroic $BiFeO_3$ to scale the switching energy density to $\approx$ 10 µJ cm^{-2}. This work provides a template to achieve attojoule-class nonvolatile memories. The key to this work was leveraging effects of lanthanum substitution is which known to both lower the polarization and the ferroelectric ordering temperature (and therefore the energy of switching) and to take advantage of innate thickness scaling effects (thinner films require smaller voltages for switching). In turn, the researchers showed that the switching voltage of the giant magnetoresistance (GMR) response can be progressively reduced from $\approx$2 V to 500 mV by a reduction of the film thickness down to 20 nm (Fig. 16a). Robust electric-field control of the magnetization direction in the bottom $Co_{0.9}Fe_{0.1}$ layer was shown in measurements both in a magnetic field of 100 Oe as well as in the remanent state (i.e., zero magnetic field) (Fig. 16b). The low-voltage magneto-electric switching in multiferroic $Bi_{0.85}La_{0.15}FeO_3$ was further probed by XMCD-PEEM imaging at the Co L_3 edge via studies (inset, Fig. 16c)

where application of $\pm$500 mV revealed contrast changes consistent with reversal of the in-plane magnetization. Further reducing the $Bi_{0.85}La_{0.15}FeO_3$ thickness to just 10 nm reduced the operating voltage to just $\pm$200 mV and maintained the robust switching behavior (Fig. 16d).

Despite this work, switching a ferroelectric state or a multiferroic state with a voltage as small as 100 mV remains a "grand challenge". Since the electric field scales with the dimensions of the ferroelectric, progression towards switching voltages of 100 mV automatically require that either the switching field be very low or that the switching behavior scales well with thickness (Fig. 17). As a direct consequence, it becomes critical to understand ferroelectric switching behavior in the ultrathin limit (< 20 nm). Quantitative studies of the switching dynamics at such a thickness are still lacking and should be a fruitful area of research in the immediate future. Indeed, I identify this as a *key* materials physics aspect that requires attention, especially on the experimental side. The top science and technology questions are also identified in Table 2.

While challenges at these voltage/energy, length, and time scales exist for all ferroelectric materials, special attention is now being

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Table 2: Key science and technology challenges.

| Materials physics	Translational
Discovery of new, room temperature multiferroics with robust coupling between magnetism and ferroelectricity, strong coupling, and magnetic moment larger than 50 emu/cc	Achieving thermal stability of ferroelectric and magnetic order parameters, as well as robust coupling between them, in 10 nm length-scales at room temperature. Thus, careful measurements of magnetoelectric and multiferroic phenomena at such length scales is critical
Developing new mechanisms for magnetoelectric coupling and understanding and approaching the limits of the strength of such phenomena	Reducing the voltage required for ferroelectric/magnetoelectric switching to ~ 100 mV
Atomic-scale design and layer-by-layer growth as an attractive pathway to discover and synthesize new room temperature multiferroics	A second key requirement for ultralow power electronics (e.g., an AttoJoule switch) would be designing proper ferroelectric multiferroics with small but stable spontaneous polarization of ~ 1–5 μC/cm²
Understanding the scaling limits, controlling and exploiting dynamics: magnetoelectric coupling at < 20 nm length scale; < 1nsec time scale; < 100kT energy scale	Integration and scale-up of synthetic approaches to enable manufacturing would be valuable
From a longer timescale perspective, reaching the theoretical Landauer limit for switching (kT(ln2) would be desirable and will require significant effort	Convergence of memory and logic
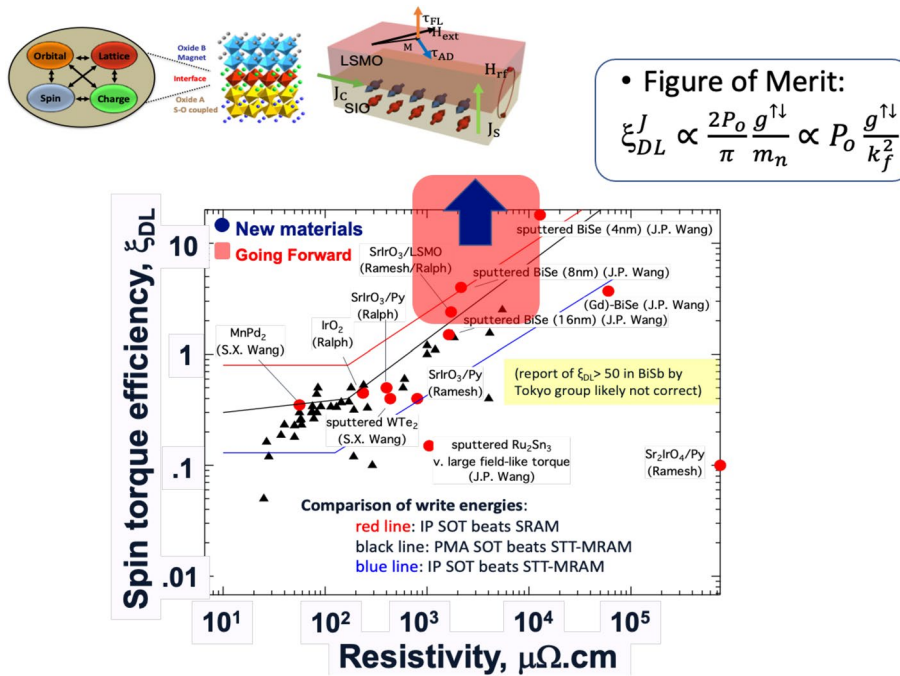
Figure 18: The top shows a schematic of the epitaxial oxide interface between an oxide ferromagnet and a S–O-coupled oxide such as SrIrO₃; to the right is shown a schematic of the spin to charge conversion, with the corresponding figure of merit, the spin torque efficiency; the bottom plot compares the spin torque efficiency vs. resistivity of a large number of metal-ferromagnet pairs. The BLUE arrow indicates the desired resistivity of the bilayer as well as the need for further enhancing the efficiency.

given to such responses in multiferroics such as BiFeO₃. The ferroelectric switching process in BiFeO₃ is believed to be limited by nucleation and growth of reverse domains[94] broadly captured by the Kay–Dunn model,[95] in which the coercive field scales as film thickness $d^{-2/3}$.

Consequently, progressively larger reductions in film thickness are needed to reduce the coercive voltage as it is pushed to smaller values. In BiFeO₃, lanthanum substitution has been shown[96] to reduce the switching energy by reducing the polarization,[97] although to an

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insufficient extent to date. Pushing $BiFeO_3$ close to a phase boundary between ferroelectric and antiferroelectric states or identifying materials without the robust octahedral rotations of $BiFeO_3$ could be an alternative pathway to smaller coercive fields. The challenges facing the spintronics community in enhancing the output of the inverse Rashba effect component by two to three orders of magnitude[98] are equally exciting. Figure 18 captures the current state of the art in spin-to-charge conversion (or the spin torque efficiency) using materials with large spin–orbit coupling. Among such materials topological insulators, such as Bi-Se and Bi–Te as well as other S–O-coupled materials which have exotic electronic band structures are favored. Correlated oxides such as $SrIrO_3$ have shown promise for large spin torque efficiencies, especially when juxtaposed epitaxially with an oxide ferromagnet such as LSMO.

5 Challenges and Opportunities

It seems inappropriate to write a concluding section when the exciting journey has just begun. Electric-field control of the magnetization direction at room temperature is now clear with the voltage required to accomplish this dropping down to just 0.5 V. As discussed recently, to get to an aJ switch, it is critical to reduce these switching voltages down even further (to ~ 100 m V) in conjunction with a switching charge density of ~ 10 $\mu C/cm^2$. How robust can this be, especially with respect to repeated cycling of the electric and magnetic states? In this regard, it appears that the community needs to increase the focus on the nature of the ferromagnet and its interface to the multiferroic. Prior experience with ferroelectric capacitors has shown that a conducting oxide contact yields a very robust capacitor; in a similar vein, we expect an oxide ferromagnet to form a more robust contact to the oxide multiferroic. Thus, there is an urgent need to discover and interface an oxide ferromagnet that couples magnetically to the multiferroic at room temperature. A template for this is already available from the work on $La_{0.7}Sr_{0.3}MnO_3/BiFeO_3$ interfaces, which display robust electric-field control of the magnetization direction, albeit at 100 K. In the same vein, there is an urgent need to discover more room temperature multiferroics so that one can explore multiple pathways to use these novel functionalities. Finally, we believe that exploring for new room temperature multiferroics would be very worthwhile pursuit for the materials community, especially when armed with the

computational discovery platforms such as the Materials Project and the Materials Genomics approach driven by ML pathways.

In this sense, tremendous progress has been made in understanding chemistry–structure–property relationships, and in engineering-specific atomic architectures, so that an era of "multiferroic materials by design" is within reach. Targeted functionalities, such as large magnetization and polarization and even exotic polarization topologies, are now within reach. Electric-field control of magnetism, while demonstrated in multiple implementations, must be optimized so that it can be achieved with smaller voltages, ideally below 100 mV. For multiferroic devices to be technologically competitive will, therefore, require precise growth of ultra-thin films guided by theoretical studies to exactly define the chemical compositions needed to optimize the polarization and coercive field. This will require improved fundamental understanding, which will be facilitated by improved first- and second-principles methods. Even with such a low-field-switching breakthrough, scale-up and integration, in particular compatibility with existing silicon processing methods, and integration with the appropriate peripheral electronics are key challenges. An oxide-based ferromagnet or ferrimagnet that couples strongly to $BiFeO_3$ and has a Curie temperature well above room temperature would be desirable.

Going forward, improved multi-scale approaches that allow treatment of the electronic and lattice degrees of freedom on the same footing[99] could lead to vastly enhanced system size and accuracy when combined with improved tools for generating effective potentials using input from first principles[100]. Modeling of the dynamics of ferroelectric switching[101] and its effect on magnetic order,[102] both of which are on time- and length-scales that are far outside the ranges accessible using density functional methods, now become feasible. Such models in combination with molecular dynamics start to allow calculation of dynamical magnetoelectric responses in the THz region,[103] which is particularly timely as it coincides with advances in experimental methods for generating THz radiation mentioned above. Finally, the ongoing development of new theoretical concepts, such as the magnetoelectric multipole as an order parameter for phase transitions that break both space-inversion and time-reversal,[104,105] as well as the production of practical computational tools for their calculation look very promising in terms

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of pushing the limits of computationally driven materials discovery[27,106–108].

I expect dynamical effects in multiferroics to increase in importance over the next years, driven by new experimental capabilities such as ultrafast X-ray sources[109] and closing of the so-called THz gap,[110] and the fundamental limits on the dynamics of spin-charge-lattice coupling phenomena will be established. Theoretical proposals of dynamical multiferroic phenomena, in which a time-dependent polarization induces a magnetization in the reciprocal manner from that in which spin spirals induce polarization[111] should be validated by careful experiments. At the same time, more work on antiferromagnetic resonance in multiferroics is required; while many studies were carried out in the 1960s[112] and 1970s on conventional antiferromagnets, activity with modern multiferroics, which typically have higher resonance frequencies (~ 700 GHz in $BiFeO_3$,[113] compared with ~ 350 GHz in other perovskite orthoferrites[112]), has been scarce.

The field of multiferroics and magnetoelectrics is poised to make further significant breakthroughs and I hope that this article will motivate additional research on this fascinating class of materials and their applications. While scientific interest in the field is beyond question, the need to identify market niches and enable pathways to products, so that multiferroics go beyond being an "area to watch" and address contemporary technological challenges. To achieve this, a shift of focus from fundamental materials discoveries to translational research and development will be needed, like that which occurred in the field of GaN-based light-emitting diodes 2 decades ago. The complexity of oxide-based material systems raises additional challenges, as we have seen for example in the colossal magnetoresistive manganites, making the active engagement of applied physicists and device engineers early in the research and development process even more essential. In this vein, the recent engagement of large microelectronic companies in the field of multiferroics[103,104] is particularly encouraging. While basic research in multiferroics is vibrant, the field would benefit from an injection of focused programs that address the transition to devices, scale-up and integration issues.

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Ramamoorthy Ramesh pursues key materials physics and technological problems in complex multifunctional oxides. Using conducting oxides, he solved the 30-year enigma of polarization fatigue in ferroelectrics. He pioneered research into manganites coining the term, Colossal Magnetoresistive (CMR) Oxides. His work on multiferroics demonstrated electric field control of ferromagnetism, a critical step towards ultralow power memory and logic elements. His extensive publications on the synthesis and materials physics of complex oxides are highly cited (over 65,000 citations, H-factor =144). He is a fellow of APS, AAAS & MRS and an elected member of the U.S. National Academy of Engineering and a Foreign member of the Royal Society of London as well as the Indian National Science Academy. His awards include the Humboldt Senior Scientist Prize, the APS Adler Lectureship and McGroddy New Materials Prize, the TMS Bardeen Prize and the IUPAP Magnetism Prize and Neel Medal. He was recognized as a Thomson-Reuters Citation Laureate in Physics for his work on multiferroics. He served as the Founding Director of the successful Department of Energy SunShot Initiative in the Obama administration, envisioning and coordinating the R&D funding of the U.S. Solar Program, spearheading the reduction in the cost of Solar Energy. He also served as the Deputy Director of Oak Ridge National Laboratory and the Associate Lab Director at LBNL. Most recently, he served on the Biden-Harris Transition Team for Energy.

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