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Oxide nanowires for spintronics: materials and devices

Yufeng Tian, Saidur Rahman Bakaul and Tom Wu*

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Spintronics, or spin-based data storage and manipulation technology, is emerging as a very active research area because of both new science and potential technological applications. As the characteristic lengths of spin-related phenomena naturally fall into the nanometre regime, researchers start applying the techniques of bottom-up nanomaterial synthesis and assembly to spintronics. It is envisaged that novel physics regarding spin manipulation and domain dynamics can be realized in quantum confined nanowire-based devices. Here we review the recent breakthroughs related to the applications of oxide nanowires in spintronics from the perspectives of both material candidates and device fabrication. Oxide nanowires generally show excellent crystalline quality and tunable physical properties, but more efforts are imperative as we strive to develop novel spintronic nanowires and devices.

1. Introduction

Since the discovery of the giant magnetoresistance (GMR) effect in 1988,^{1,2} the spin-based electronics or spintronics has emerged into an active research area. Spin-based data storage and manipulation possess several advantages over the conventional charge-based counterpart, including ultra-low power consumption, higher data processing speed, nonvolatility, improved scalability and so on.^{3,4} However, there are several prominent obstacles down this road of spin-based technology.

There exists the grand material challenge: so far there is no ferromagnetic (FM) semiconductor exhibiting both strong magnetism and high transition temperature, suitable for room temperature device applications. The most thoroughly studied and best understood FM semiconductor is Mn-doped GaAs, but it suffers from low Curie temperatures capped at about 190 K.^{5,6} A breakthrough is the theoretical prediction of above room temperature ferromagnetism (RTFM) in Mn-doped GaN and ZnO,⁷ which jump-started a flurry of activities aiming to find suitable candidates in doped wide-bandgap oxides, but experimentally observed magnetism is usually very weak.^{8,9}

Transition metal oxides exhibit rich physical properties ranging from insulators, semiconductors, to metals, and even superconductors. Their bandgaps span from the visible to the ultraviolet (UV) regimes, and their magnetic properties range

Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, 21 Nanyang Link, 637371, Singapore. E-mail: TomWu@ntu.edu.sg; Fax: +65 6794 1325; Tel: +65 6514 1047



Yufeng Tian

Yufeng Tian received his bachelor degree from Shandong University (China) in 2003. He worked as a visiting scholar in University of Idaho (America) from 2007 to 2008. After receiving his PhD from Shandong University in 2009, he worked as a postdoctoral scientist in Prof. Tom Wu's group at Nanyang Technological University (Singapore). His research focuses on the magnetic and transport properties of various spintronics oxide materials and devices.



Saidur Rahman Bakaul

Saidur R. Bakaul received his BS in Electrical and Electronic Engineering from Bangladesh University of Engineering and Technology (2005) and his PhD in Electrical and Computer Engineering from the National University of Singapore (2011). Since 2011, he has been working as a postdoctoral researcher in the group of Prof. Tom Wu. His research interests include spintronic devices, oxide ferromagnets and electronic transport properties of superconductor ferromagnet hybrid devices.

from diamagnetic, paramagnetic, to FM and antiferromagnetic (AFM). They can also possess notable dielectric properties both in the low- k and high- k regimes, and can be ferroelectric and piezoelectric. Surface chemical reactivity of oxides can be tailored from being highly active to inert. All these functionalities stem from the strong and complex coupling between charge, spin, orbital, and lattice degrees of freedom in the material systems (Fig. 1).¹⁰

Driven by the needs for miniaturization in many areas of modern technology, more-and-more attention is paid to the nanoscale systems. This is enabled by the significant breakthroughs in deposition technologies, lithography processing, and characterization tools, which are accompanied by new insights in theory and computation (Fig. 1). It is recognized that nanoscale systems are not merely the miniature form of the bulk counterpart, instead they can bring about many “emergent phenomena” such as quantum confinement and proximity effects.¹¹ Nanostructures including nanowires, nanotubes, nanorods, and nanoparticles represent an exciting and rapidly expanding research area that crosses the borders between physics, chemistry, biology, engineering, and materials science.^{12–14} These

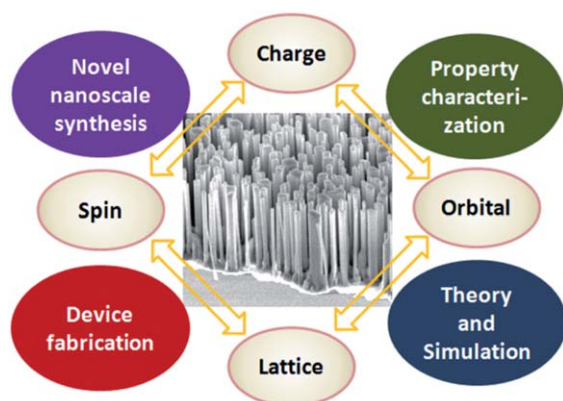


Fig. 1 Schematic diagram showing the intricate coupling between charge, spin, orbital and lattice in transition metal oxides, as well as the close relationship between various research fronts.



Tom Wu

Tom Wu obtained his bachelor degree from Zhejiang University (China) in 1995. After receiving his PhD from University of Maryland in 2002, he worked as a postdoctoral scientist in Argonne National Laboratory. In 2006, he joined Nanyang Technological University (Singapore) as an assistant professor. His research group focuses on synthesizing a wide range of oxide nanomaterials and thin films, and fabricating novel field effect and memory devices.

bottom-up nanostructures are potential building blocks for the nanoscale electronics and photonic devices.^{15–21}

Here, we focus on the perspective of applying oxide nanowires in spintronic applications. On the material aspect, we will discuss the physical properties of nanowires made of wide-bandgap oxides, colossal magnetoresistance oxides, and other half-metallic oxides. On the device aspect, we will review some notable examples of spintronic devices, the advantages of incorporating bottom-up nanowires, and the associated challenges.

2. Materials candidates for spintronic nanowires

2.1 Wide-bandgap dilute magnetic oxides

Since the theoretical prediction of a hole-mediated RTFM in Mn-doped ZnO at a carrier concentration around 10^{20} cm⁻³,⁷ transition metal doped wide-bandgap oxides such as ZnO, TiO₂, In₂O₃, SnO₂, and (In, Sn)₂O₃ (ITO) have been regarded as promising candidates for high Curie temperature dilute magnetic oxides (DMOs). Although extensive experimental investigations have been performed on both undoped and transition-metal-doped DMOs,^{22–27} the complex magnetic behavior is found to sensitively depend on the concentrations of doped cations, carriers, defects, and synthesis details.²⁷

In many cases, the formation of metallic clusters or magnetic secondary phases,²⁸ even artefacts and contaminations are responsible for the observed magnetic signals.²⁹ In terms of intrinsic mechanisms, for DMOs with high carrier concentrations, long-range FM order can be Ruderman–Kittel–Kasuya–Yoshida (RKKY) or double exchange type.^{10,30,31} For highly insulating DMOs, the long range FM exchange is mediated by localized carriers in the impurity band to form bound magnetic polarons.^{27,32} RTFM was also observed in undoped oxides, known as d^0 ferromagnetism, which is closely related to the defects.³³ Coey *et al.* proposed a general charge transfer model, where the FM order is linked to itinerant carriers associated with structural defects and TM dopants.³⁴ Despite the large number of studies in this field, the mechanisms of ferromagnetism in DMOs remain unclear, and the present status is discussed in several recent reviews.^{10,35}

Nanowires are often single phase and highly crystalline, and they are ideal single-domain building blocks for constructing spintronic nanodevices. High quality ZnO,^{36–38} In₂O₃,^{39,40} and TiO₂⁴¹ nanowires can be routinely synthesized. Their morphology can be well controlled by catalyst,^{37,42} precursor,⁴³ pressure, and geometry⁴⁴ during synthesis, also by post-growth treatments such as annealing⁴⁵ and coating.^{45–48} Pioneer works indicate that ferromagnetism can be stabilized in various wide-bandgap oxides nanowires or nanorods doped with transition metals, including ZnO doped with Co,^{49–52} Mn,^{52–54} Ni,⁵⁰ Cu,^{55–57} and FeCo;⁵⁸ TiO₂ doped with Fe,⁵⁹ Co,^{60,61} and Cr;⁶⁰ SnO₂ doped with Co,⁶² Mn,⁶³ Ni,⁶⁴ and Cu,⁶⁵ and even in oxide nanowires doped with nonmagnetic elements, such as ZnO doped with C⁶⁶ and N.⁶⁷ The large surface-to-volume ratios of nanowires can greatly enhance ferromagnetism through surface defects⁶⁸ or structural/compositional inhomogeneity.^{55,56} As an example, the nanostructure and magnetic properties of well aligned Cu-doped ZnO nanowires were shown in Fig. 2.

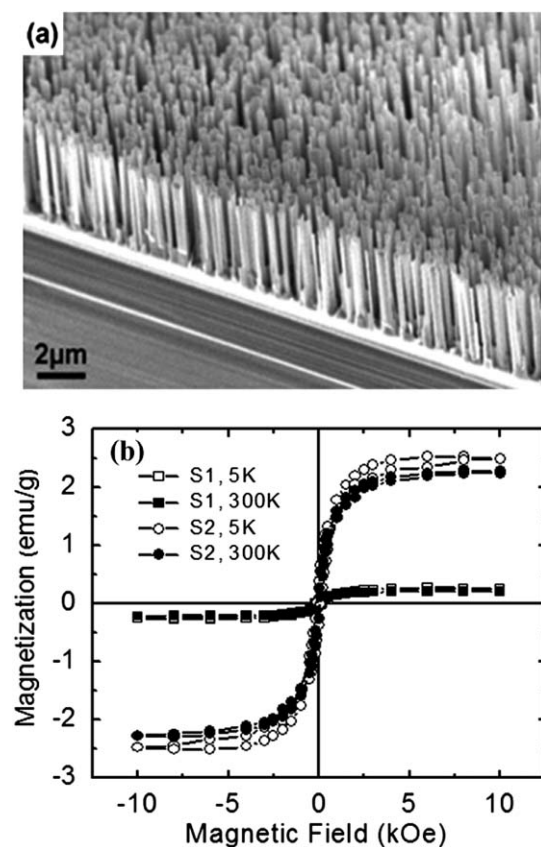


Fig. 2 (a) SEM image of vertically aligned Cu-doped ZnO nanowires grown on a sapphire substrate. (b) Magnetic hysteresis loops measured at 5 K and 300 K for Cu-doped ZnO nanowires with homogeneous (S1) and inhomogeneous (S2) dopant distributions. Copyright 2008 Wiley-VCH, reproduced from ref. 55 with permission.

Nanowires exhibit multiple functionalities such as gas sensing,^{69–71} photovoltaic devices,^{72,73} and transparent electrodes.⁷⁴ Because of symmetry-breaking surfaces, nanowires of SnO₂ and In₂O₃ show UV light emission and even lasing which are absent in the bulk counterparts.^{75–79} Furthermore, three dimensional epitaxial heterostructures with tailored optical properties were synthesized,⁸⁰ and interface-related properties can be explored.

Nanowires have been successfully incorporated into nanoelectronics,⁸¹ and naturally they are envisaged as ideal building blocks for nanoscale spintronics. Qiu *et al.* showed that a metal–semiconductor hybrid nanostructure can provide a unique opportunity to stabilize RTFM because metal decoration can tune the Fermi level of oxide nanowires and promote the carrier transfer.⁸² Furthermore, the Curie temperature of FM semiconductor in laterally patterned heterostructures was manipulated,⁸³ and this nanolithography-based approach may be also applicable to DMOs. In terms of devices, a theoretical study suggested that the magnetic properties of DMO nanowires can be tuned by an electrical field,⁸⁴ which may enable the electric control of spin polarized current.

Magneto-transport properties of individual oxide nanowires have been studied,^{85–87} where s–d exchange induced spin splitting of conduction band results in low-field positive magnetoresistance (MR), while suppression of the weak localization effect is

responsible for the negative MR at high fields (Fig. 3).⁸⁶ Furthermore, the magneto-transport of nanowires can be modulated by applying a gate voltage.⁸⁶

Combining the oxide nanowire channel (ZnO, In₂O₃ and so on) with the ferroelectric gate dielectrics (Pb(Zr_{1–x}Ti_x)O₃, for example) in ferroelectric transistors has been explored.^{88,89} Memory effects and enhanced performance were observed. Potentially, novel nanowire-based devices with multiferroic components can be developed.

Another notable area is p–n junctions and light emitting diodes (LED) based on oxide nanowires.^{90,91} Good rectification and blue-light-emitting LED have been demonstrated. By replacing non-magnetic nanowires with FM nanowires and/or combining with FM electrodes, we can envisage nanowire-based spin LED.

2.2 Mixed-valence manganites

Mixed-valence manganites have been intensively investigated because of the “colossal magnetoresistance” (CMR) and the complex magnetic phase transitions.⁹² In manganites, the magnetic, electronic, and structural degrees of freedom interact with each other through “double exchange” and Jahn–Teller interaction. As an example, the rich phase diagram of La_{1–x}Sr_xMnO₃ is shown in Fig. 4.⁹³

In terms of applications, the large MR and the great tunability of CMR oxides are promising for magnetic recording, spin valve devices, and magnetic tunnelling junctions.^{94–98} For example, Zhao *et al.* reported an electric field modulation of MR in a La_{0.9}Sr_{0.1}MnO₃ junction from –70% to 80% near room temperature.⁹⁹ Complementary magnetic and electric field tunings were observed in devices with phase-separated manganites as channel,¹⁰⁰ and the transition temperature was tailored electrically.^{101,102} In narrow bridges of manganites with macroscopic phase separation, negative differential resistance has been observed as a result of reversible metal-to-insulator transition in nanoscale filaments.^{103,104}

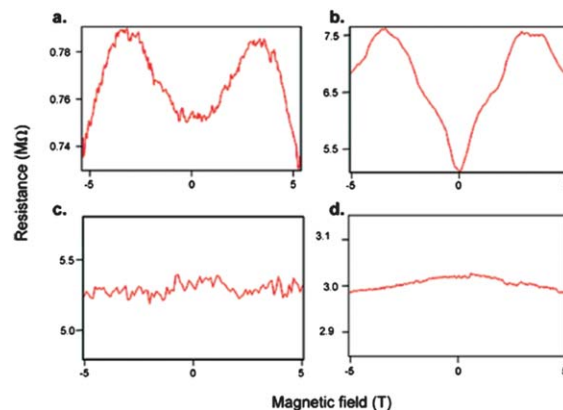


Fig. 3 MR measured at 10 K on devices made of (a) Co-doped ZnO nanowire contacted by Co electrodes; (b) Co-doped ZnO nanowire contacted by Au electrodes; (c) pure ZnO nanowire contacted by Co electrodes, and (d) pure ZnO nanowires contacted by Au electrodes. Copyright 2006 American Chemical Society, reproduced from ref. 86 with permission.

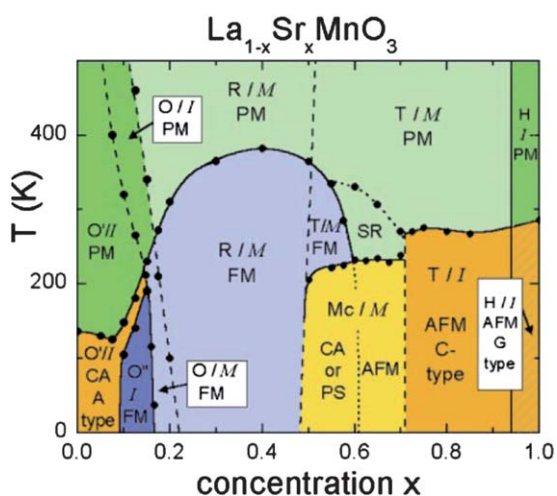


Fig. 4 Phase diagram of $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$. The crystal structures (Jahn–Teller distorted orthorhombic, O' ; orthorhombic, O ; orbital ordered orthorhombic, O'' ; rhombohedral, R ; tetragonal, T ; monoclinic, Mc ; and hexagonal, H) are indicated as well as the magnetic structures (paramagnetic, PM ; short range order, SR ; canted spin structure, CA ; anti-ferromagnetic structure (either A - or C -, or G -type), AFM ; ferromagnetic, FM ; phase separated, PS) and the electronic state (insulating, I ; metallic, M). Copyright 2009 American Physical Society, reproduced from ref. 93 with permission.

However, there are several obstacles related to manganites in nanodevice applications. First, the spin polarization of manganites decays rapidly with temperature. Second, the defect chemistry and the stoichiometry–property correlation in manganites are quite complex.^{105,106} Third, the physical properties of interfaces in manganite-based devices remain elusive.^{107,108} Finally, there is the urgent need for developing suitable device processing techniques.

Compared to the wide-bandgap oxides, bottom-up synthesis of manganite nanowires is more challenging. Nevertheless, high quality single crystal nanocubes¹⁰⁹ and nanowires¹¹⁰ of manganites have been reported. But so far there has been few studies on spintronic devices based on individual manganite nanowires.

For manganites, nanowires and related heterostructures hopefully can enhance the Curie temperature¹¹¹ and the low-field MR.^{112,113} Another promising research topic is anisotropic magnetoresistance (AMR) which is a result of spin-orbital interaction and has important applications in magnetic field detection and data storage.¹¹⁴ Significant magnetic anisotropy has been observed in nanowires.¹¹⁰ In addition, the morphology of nanowire can be modified by annealing or growing on engineered substrates,¹¹⁵ which could significantly affect the properties. We look forward to more efforts on exploring the anisotropic transport in manganite nanowires and gaining deeper understanding on low-dimensional spin-dependent transport.

2.3 Half-metallic binary oxides

Half-metallic ferromagnets exhibit conducting states at the Fermi level for the majority spin electrons, but a semiconductor gap for the minority spin electrons.¹¹⁶ Direct experimental

evidence to confirm such spin-dependent band structures have been obtained by using techniques like spin-resolved photo-emission¹¹⁷ and point contact Andreev reflection.¹¹⁸ As a result of the high spin polarization ratio ($\sim 100\%$), half-metallic binary oxides such as Fe_3O_4 , CrO_2 , and EuO are attractive for spintronics applications.¹¹⁹

Magnetite (Fe_3O_4) is probably the most studied half metal because of its high Curie temperature (~ 858 K), which is critical for device applications.¹²⁰ Moreover, the lack of toxicity in Fe_3O_4 also promises applications in medical and biological areas, such as drug delivery and magnetic resonance imaging.¹²¹ Previously, Fe_3O_4 nanowires,^{120,122} nanotubes,^{123,124} and core/shell nanowires^{125,126} have been synthesized, and their magneto-transport properties were studied. Ferromagnetism of magnetite nanowires remains above room temperature, and the Verwey transition temperature is around 120 K.¹²⁰ In a magnetic tunnel junction made of Fe_3O_4 nanocrystals, the sign of tunnel MR was found to switch from negative to positive at the Verwey transition.¹²⁷ Liao *et al.* demonstrated a spin filter effect in Fe_3O_4 nanowires contacted with normal metallic electrodes, where only the minority spin carriers can transport through the Fe_3O_4 nanowires due to the negative spin polarization (Fig. 5).¹²² Furthermore, the room temperature positive MR can be modulated by the bias voltage.¹²²

Exchange bias is widely used in the present commercial technologies such as spin valve and magnetic tunnel junctions, and it is regarded as an effective way to overcome the superparamagnetic limit in magnetic storage media.¹²⁸ The exchange coupling and exchange bias effect in Fe_3O_4 based branched¹²⁹ and core–shell nanocrystals¹³⁰ have been studied, and novel properties including higher saturation magnetization, enhanced coercivity, exchange bias, and improved thermal stability were observed.

As another notable spintronic material, CrO_2 has a high spin polarization ($>95\%$)¹¹⁸ and a Curie temperature of 395 K. Fascinating transport properties have been revealed in CrO_2 thin films.^{119,131} For instance, the CrO_2 based tunnelling junction shows an enhanced low field MR,¹¹⁹ and the Josephson supercurrent through the CrO_2 may have a spin triplet nature.¹³¹ However, CrO_2 is metastable in ambient conditions, and its synthesis is rather challenging. Recently, single crystal CrO_2 nanorods¹³² and nanowires¹³³ have been reported. Spin-dependent tunnelling transport was achieved in individual CrO_2 nanorod devices contacted with nonmagnetic Ti/Au metallic electrodes.¹³² Unfortunately, the magnitude of the spin dependent MR decreases rapidly with increasing temperature and bias voltage, which may be a result of spin independent inelastic hopping across the contact barriers.¹³²

EuO is an attractive material as it may be the only magnetic binary oxide that could be thermodynamically stable in contact with silicon.¹³⁴ Schmehl *et al.* demonstrated that the spin polarization ratio of EuO exceeds 90%, and the half metallic behavior remains even when highly doped.¹³⁴ More interestingly, giant Zeeman splitting and obvious magnetic circular dichroism (MCD) response confirm the ferromagnetic nature of polycrystal EuO nanorods synthesized *via* a three step reaction process.¹³⁵ In addition to its strong magnetism ($6.9 \mu_B$), these EuO nanorods have one of the largest MCD responses of any materials, highlighting their potential in magneto-optical applications.¹³⁵

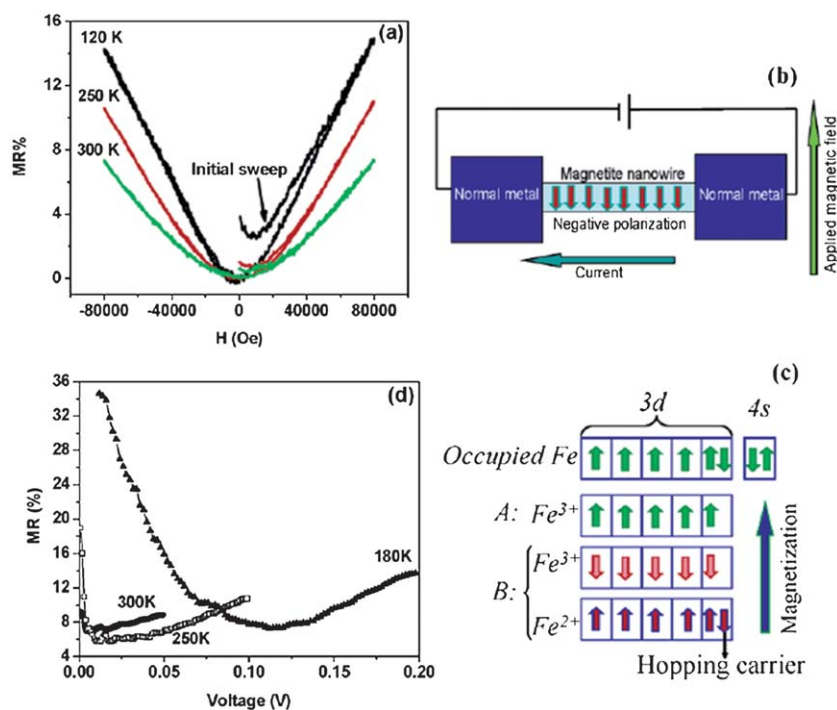


Fig. 5 (a) MR of individual Fe₃O₄ nanowires measured at different temperatures. (b) Schematic of the device. (c) FM order of Fe₃O₄, where the minority carriers are responsible for the transport. (d) Bias dependence of MR for the Fe₃O₄ nanowire-based devices. Copyright 2006 American Chemical Society, reproduced from ref. 122 with permission.

2.4 Ferroelectric and multiferroic oxides

Nanowires of perovskite ferroelectric oxides, *e.g.*, BaTiO₃, have been used in energy harvesting devices,^{136,137} photonics, and nanoelectromechanical systems.¹³⁸ Molten-salt method is effective to produce nanostructures of complex oxides, *e.g.*, nanoparticles, nanocubes, nanoblets, and nanoflowers.¹³⁹ Using a scanned probe microscope, Yun *et al.* demonstrated that nonvolatile electric polarization can be reproducibly induced and manipulated on single crystal BaTiO₃ nanowires.¹⁴⁰ The ferroelectricity is retained in nanowires as thin as 0.8 nm, which can be attributed to surface adsorbates such as OH and carboxylates.¹⁴¹

Multiferroic materials show simultaneous ferroelectric and magnetic orders, promising novel applications.¹⁴² Among them, BiFeO₃ (BFO) has received the most attention, due to its high ferroelectric Curie temperature (~1103 K) and the AFM Neel temperature (~647 K). Periodic ferroelectric domains¹⁴³ and electrical control of AFM domains¹⁴⁴ have been realized in BFO. Different from the AFM order in bulk BFO, weak ferromagnetism and superparamagnetic behaviour at low temperature were observed in polycrystalline nanowires, indicating significant size effects on the magnetic order of BFO.^{145,146} Synthesis of single crystalline BFO nanowires has not been reported yet, which is also the status for many other complex oxides. Ground breaking efforts are needed to realize high quality nanowires of complex perovskite oxides with controlled composition and morphology. A promising approach towards well ordered complex oxide nanostructures is to combine lithography and epitaxial thin film growth,¹⁴⁷ which will hopefully facilitate future studies.

3. Spintronic devices based on oxide nanowires

3.1 Fabrication techniques: top down vs. bottom up

Perhaps the most realistic approach to realize practical nanowire-based spintronic devices is the “top-down” technique, in which some form of lithography, such as electron beam/optical lithography,^{148,149} nanoimprint lithography,¹⁵⁰ or μ -contact printing^{151,152} is used to pattern desired nanostructures. Besides, focused ion beam technique^{153,154} or chemical etching in conjunction with suitable masks can also be used to carve out nanowires from an oxide film. In addition, Suzuki *et al.* invented a unique combination of atomic force microscopy lithography and Mo lift-off technique to create oxide nanowires with dimensions down to 100 nm.¹⁵⁵

The lithography process has the advantage of excellent controls over position and shape of the nanowires, and making contacts and multilayers always involve lithography. However, top-down fabrication often entails expensive equipment and complicated processing. Furthermore, most of the oxide materials need high deposition temperature, and the typical electron beam and photo resists are incompatible with this requirement.

The bottom-up synthesis mainly refers to the self-assembly of nanowires without involving any lithography. It has advantages in terms of mass production and cost-effectiveness. In addition, the bottom-up nanowires are single crystalline and free from defects such as edge roughness, which is a major challenge in lithographically patterned nanowires. So far there has been few work on using self-assembled nanowires in spintronic devices, and a series of baby steps need to be taken, such as rational nanoscale synthesis, controlled positioning of nanowires, making multilayer structure, and creating good electrical contacts.

In some cases, combining top-down and bottom-up techniques have more advantages.^{156,157} Lithographically patterned resists can be used as masks and then directed growth of oxide nanowires is achieved by spin coating of sol-gel precursors. Recently, there have been a few reports on creating oxide nanowires by using laser interference techniques.^{158–160} In this process a periodic interference pattern can be generated by using a laser source and prisms with different numerical apertures, creating regular arrays of oxide nanowires.

Guided synthesis and assembly have been demonstrated in nanowire-based devices. Dai showed that the locations of carbon nanotubes can be controlled by using lithographically patterned catalytic nanoparticles, and their growth directions can be determined by van der Waals self-assembly forces and applied electric fields.¹⁶¹ The electrical-field-controlled alignment of nanowires was also demonstrated for semiconductor nanowires,^{162,163} and Langmuir-Blodgett (LB) technique was used for similar purposes.¹⁶⁴ In addition, structures of crossed nanowires, suitable for memory devices and logic circuits, have also been realized by using a combination of top-down and bottom-up approaches.^{165,166} Such fabrication techniques should be feasible for making spintronic devices with oxide nanowires.

3.2 Spin valve and magnetic tunnel junctions

Spin valve may be the most influential spintronic device which has already found applications in magnetic data storage industry. Its basic working principle is the GMR effect,^{1,2} where the resistance of a FM/NM/FM multilayer depends on the relative alignment between the two FM layers; In general, an antiparallel alignment gives rise to larger resistance than the parallel alignment. Most GMR research focuses on transition metals, but spin valves have also been realized in oxides, in particular FM manganites.^{167,168}

A related concept is the magnetic tunnel junction (MTJ)^{169,170} which also has vast applications in nonvolatile magnetic memory devices. The basic difference between spin valve and MTJ lies in the middle spacer layer, which needs to be insulating for MTJ, whereas for spin valve, this layer is conducting. A number of efforts have been devoted to create oxide based MTJ devices so far, especially due to the 100% spin polarization in several FM oxides, such as $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ (LSMO) with $x \sim 0.33$, CrO_2 and Fe_3O_4 . Lu *et al.*¹⁷¹ and Sun *et al.*¹⁷² first fabricated all-oxide MTJ device with LSMO and SrTiO_3 as FM and insulating layer, respectively. Subsequently, a record tunneling magnetoresistance (TMR) ratio of 1850% was reported in 2003 by Bowen *et al.*¹⁷³ Despite these promising results, a major issue for the oxide based MTJ devices is that the working temperature is often lower than the room temperature, generally ascribed to the degraded interfaces.^{174,175}

Nanowire-based spin-valve devices were first realized in metallic systems. Piraux *et al.*¹⁷⁶ and Blondel *et al.*¹⁷⁷ reported on such devices based on NiFe/Cu and Co/Cu by using electro-deposition in nanoporous polymer templates. In 2000, Evans *et al.*¹⁷⁸ synthesized multilayered (Co–Ni–Cu/Cu) nanowire spin valves in anodic aluminum oxide (AAO) membranes which showed very large GMR effect (55% at room temperature and 115% at 77 K). Among other works, Dubois *et al.* used permalloy (Py) and Cu stacked in a vertical template,¹⁷⁹ whereas Castaño

*et al.*¹⁸⁰ grew in-plane nanowire stacks of Py, Cu and Co using interference lithography and ion milling. Core-shell nanowires were also demonstrated recently, which act as integrated spin valves (Fig. 6).¹⁸¹ The fully functional devices contain a chemical-vapor-deposited Ni core nanowire which was surrounded by sequential shell layers of CoO(10 nm)–Co(5 nm)–Cu(5 nm)–Co(5 nm) deposited by sputtering, and a MR effect of 9% was reported in this structure.

While nanowire based spin valves with transition metals have been extensively exploited, similar devices with oxide ferromagnets have been rarely reported. Recently, Gaucher *et al.*¹⁸² successfully fabricated $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ nanowires with the smallest width of 65 nm by combining electron beam lithography and ion beam etching. They showed that the electronic transport properties of these oxide nanowires are comparable to those of unpatterned films. Goto *et al.* demonstrated GMR effect in $(\text{Fe,Mn})_3\text{O}_4$ nanowires with constrictions, which serve as lateral spin valves with pinned domain walls.¹⁸³

Oxide nanowires are very promising to serve as building blocks in lateral spin valves or MTJs. One possible scheme starts with bottom-up synthesized FM oxide nanowire, and a small portion of the nanowire can be converted into nonmagnetic by using techniques like selective Ar^+ ion milling, resulting in FM-NM-FM lateral devices. Such nanowire-based spintronic devices are attractive, but the central modified region must be thin enough to retain the spin coherent transport. Recently, Dai *et al.* theoretically showed that by selective partial hydrogenation of C-doped ZnO nanowires, it is possible to create a nanowire-based MTJ (Fig. 7).⁶⁶ Electronic phase separation in manganites may be exploited to realize such lateral nanowire-based devices.

3.3 Magnetic domain walls and spin transfer torque in oxide nanowires

A magnetic domain wall separates two oppositely polarized magnetic regions, and a number of data storage schemes based

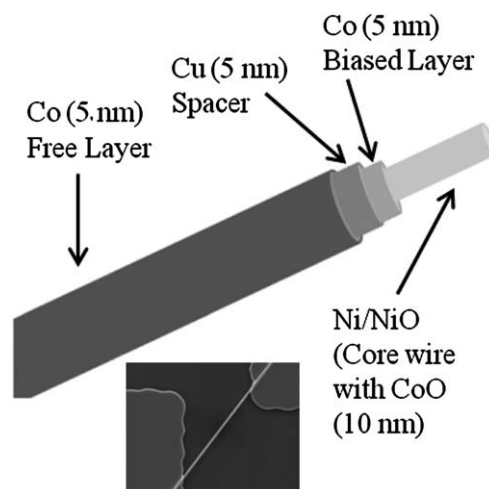


Fig. 6 Schematic of a core-shell nanowire-based spin valve device. The CoO shell biases the Co reference layer, and Cu acts as the nonmagnetic spacer. The inset shows the scanning electron microscopy (SEM) image of such a device. Copyright 2010 IEEE Magnetic Society, reproduced from ref. 181 with permission.

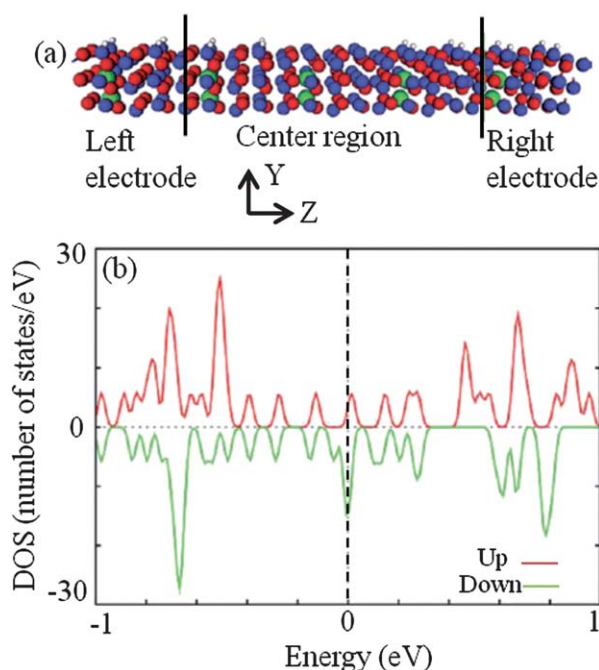


Fig. 7 (a) Atomic configuration of a TMR device made of a partially hydrogenated C-doped ZnO nanowire. Two electrodes are semi-infinite partially hydrogenated C-doped ZnO wires. The center region contains one unit cell of unhydrogenated wire and one unit cell of each electrode. (b) Theoretical calculation of the spin resolved total density of states of partially hydrogenated electrodes, showing a conversion from semiconducting to metallic characteristics. Copyright 2011 American Institute of Physics, reproduced from ref. 66 with permission.

on domain walls in magnetic nanowires have been proposed.^{184,185} In the race-track memory, each magnetic domain wall represents a data bit.¹⁸⁴ During the write operation, the domain wall is moved by external magnetic field or spin transfer torque.^{186–188} To read a bit, GMR or TMR type devices are used to detect the stray field from the domain wall. To utilize such a scheme, it is critical to controllably create domain walls. Magnetic nanowires with an artificial pinning center, such as notches,¹⁸⁴ bent conduits,¹⁸⁹ and narrow rings¹⁹⁰ can serve this purpose. Extensive research works have been performed to realize such memory devices using transition ferromagnetic metals such as Py, Co, and Ni.

In manganites, various types of domain patterns such as stripes,¹⁹¹ bubbles,¹⁹² and checker-boards¹⁹³ have been reported. Wu *et al.* observed perpendicular stripe magnetic domains in LSMO nano dots.¹⁹⁴ Recently, Takamura *et al.* reported flower-shaped, flux closure domain and vortex structures in patterned manganites created by Ar⁺ ion milling (Fig. 8).¹⁴⁸ Mathews *et al.* reported successful fabrication of LSMO nanowires on NdGaO₃ substrate by using interference lithography.¹⁵⁹ It was demonstrated that not only the shape anisotropy but also the substrate induced anisotropy play important roles in determining the magnetic easy axis in these manganite nanostructures.

Controlled manipulation of domain walls in manganite nanostructures, which is essential for domain wall based memory devices, has not been thoroughly investigated. A few successes in this context came by using CrO₂. König *et al.*,¹⁹⁵ Biehler *et al.*¹⁹⁶

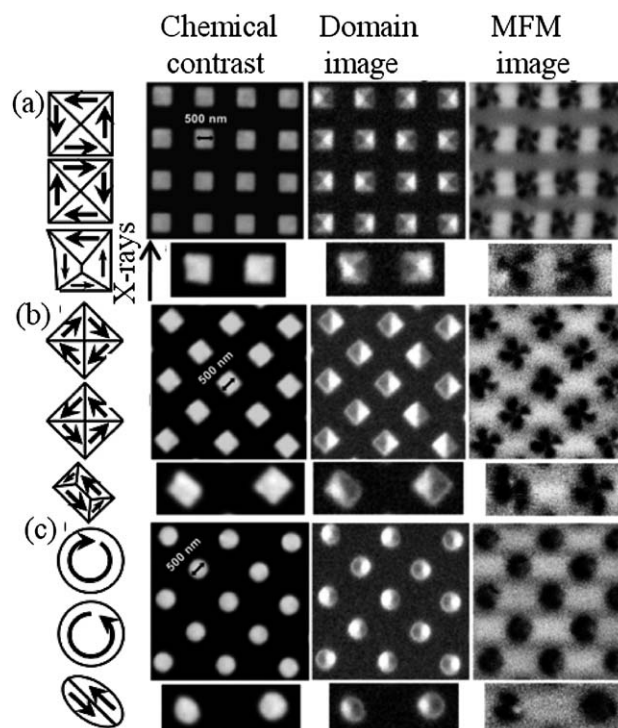


Fig. 8 Magnetic domain pattern in 500 nm diameter LSMO islands on (001) oriented STO substrates with (a) square, (b) diamond, and (c) circular shapes. The lower panel to each image shows the effect of distortions of the island shape. Copyright 2006 American Chemical Society, reproduced from ref. 148 with permission.

and Zou *et al.*¹⁹⁷ have performed pioneering experiments to create well defined magnetic domain walls in CrO₂ nanowires using Ar⁺ ion milling and selective area growth techniques. These works suggest that it is possible to pin magnetic domain walls at the notched areas and inject them controllably along the CrO₂ nanowires by using a nucleation pad and an external magnetic field.

In spite of challenges in controlling magnetic domain walls in oxide nanowires, several groups have reported current induced domain wall motion in oxide materials such as La_{0.7}Sr_{0.3}MnO₃, La_{0.67}Ba_{0.33}MnO_{3-δ}, CrO₂, and Fe₃O₄. By using focused ion beam (FIB) milling, Ruotolo *et al.*¹⁹⁸ and Céspedes *et al.*¹⁹⁹ patterned La_{0.7}Sr_{0.3}MnO₃ into nanowires containing notches as the domain wall pinning centers. The MR measurements confirm the current induced domain wall depinning with a critical current density of 10¹¹ A m⁻² (Fig. 9). Liu *et al.*²⁰⁰ reported current dependent low field MR effect in La_{0.67}Sr_{0.33}MnO₃ nanowires with constrictions and ascribed this effect to the spin polarized bias current. In a similar constricted La_{0.67}Ba_{0.33}MnO_{3-δ} nanowire, Pallecchi *et al.* observed magnetic field and DC bias current dependent asymmetric resistance hysteresis, which was also connected to the effect spin transfer torque.²⁰¹ Surprisingly, the threshold current was found to be in the range of 10⁷–10⁸ A m⁻², much smaller than the typical current needed for moving domain walls in metals (10¹¹ A m⁻²).²⁰² A number of possibilities, such as stronger spin torque due to half metallicity, Joule heating assistance and spin wave excitation may contribute to such a drastic reduction in the threshold current.

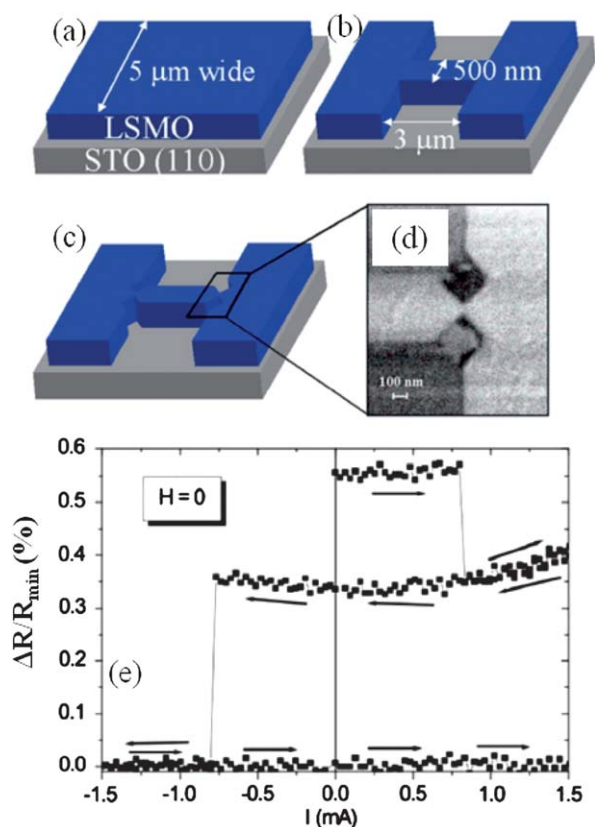


Fig. 9 Schematic of the fabrication process of a LSMO notched nanowire: (a) Ar^+ ion milling of $5\ \mu\text{m}$ wide tracks, (b) FIB milling of $3\ \mu\text{m}$ long, $500\ \text{nm}$ wide bridges, (c) FIB milling of nanoconstrictions at the bridge borders, and (d) SEM image of a patterned nanoconstriction. (e) Resistance change due to the current-induced DW displacement for a device with $50\ \text{nm}$ constriction. The arrows indicate the direction of the sweeping current. Copyright 2007 American Institute of Physics, reproduced from ref. 198 with permission.

Apart from the manganite nanowires, current-induced domain wall motion has also been experimentally reported in constricted Fe_3O_4 ²⁰³ and CrO_2 ¹⁹⁶ nanowires with threshold current densities of $10^{13}\ \text{A m}^{-2}$ and $10^{10}\ \text{A m}^{-2}$, respectively. However, none of these experiments showed direct evidence of current-induced domain wall motion by using imaging techniques such as magnetic force microscopy. Overall, more efforts from the community are needed to shed light on the pinning and removal of domain walls in oxide nanowires, which is essential for future device applications.

In this research area of domain wall manipulation, oxide nanowires are very promising to present breakthroughs. The threshold current density needed for getting enough spin torque to move a domain wall in typical transition metals is very high ($\sim 10^{10}\ \text{A m}^{-2}$). To overcome this challenge, it can be advantageous to use half metallic oxides, such as LSMO, Fe_3O_4 and CrO_2 . Furthermore, a typical drawback of metals is their strong tendency for oxidation, which demands the incorporation of a protective capping layer in the device configuration. However, due to the spin flipping at the capping layer, the spin-torque-assisted domain wall motion and the magnetization switching process often become complicated.^{204,205} Oxide-based spintronic

devices can be much more stable. Finally, as the domain configuration in oxide materials depend sensitively on the strain state,^{206,207} strain may provide an extra handle to control and manipulate the domain walls.

3.4 Oxide nanowire based spin-field-effect transistor (spin-FET)

Since the invention of the spin valve, a number of novel device concepts have been proposed, including spin-field-effect transistor (spin-FET)²⁰⁸ and spin-LED.^{209,210} Datta and Das first proposed the concept of spin-FET, where instead of the charge degree of freedom, the spin state of the electron is used as the information carrier. It consists of similar structure to the typical metal-oxide-semiconductor FET device, except that the drain and source contacts are half-metallic ferromagnets which are magnetized in the channel current direction. The spin of the channel electrons and hence the resistance of the device, can be manipulated by pure Rashba type²¹¹ or Dresselhaus type spin-orbit coupling²¹² or a delicate mixture of them,^{213,214} with the assistance of an external gate voltage.

Two very important issues in designing spin-FET are the leakage current in the off-state and the spin relaxation in the channel. In this context, nanowires are suitable candidates as the channel (Fig. 10). 1D or quasi-1D nanowires may help suppress the leakage current as a result of their small cross section. Typically, semiconductor channels have mainly two types of spin relaxation mechanisms, *i.e.*, the D'yakonov-Perel mode²¹⁵ and the Elliott-Yafet mode.²¹⁶ In "narrow" nanowire-based channels these relaxation mechanisms are expected to be suppressed and new diffusion modes may emerge during the propagation of spins.²¹⁷⁻²¹⁹

In general, the operation of spin-FET relies on the controllability over channel electron spins through the spin-orbit interaction. Therefore, in order to realize oxide nanowire based spin-FET, one need to choose materials with a long spin de-phasing length (L_ϕ) and strong spin-orbit coupling. Indium tin oxide (ITO) nanowires could be a promising candidate in this regard. Recently Hsu *et al.* showed that L_ϕ in ITO nanowires can be as large as $520\ \text{nm}$ at low temperatures.²²⁰ Furthermore, it was shown that by modifying the level of disorder in these nanowires, the strength of spin-orbit coupling can be adjusted.

Apart from serving as channel materials, half-metallic oxides are promising candidate as the source and drain materials in a spin-FET. Due to the 100% spin-polarization and the possibility of controlling the magnetization by an external field, LSMO, CrO_2 and Fe_3O_4 can be incorporated as sources of

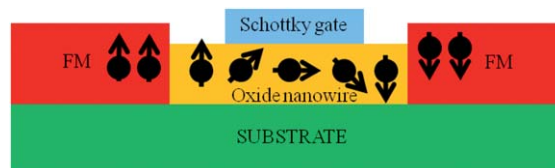


Fig. 10 Schematic of the proposed Datta-Das spin-FET²⁰⁸ (OFF state) with a nanowire channel. By tuning the spin-orbit interaction strength, the electron spin can be modulated and the device can be switched between the ON and OFF states.

spin-polarized carriers, which can eventually lead to all-oxide spin-FET devices.

4. Conclusions

Nanowires of wide-bandgap oxides, mixed-valence manganites, half-metallic oxides, and multiferroic oxides are promising candidates for potential applications in spintronics. Their physical properties can be genuinely different from the bulk counterparts due to excellent crystallinity, size effects, surface states, broken symmetry, and so on. With rational materials design, multiple functionalities such as enhanced ferromagnetism, improved low-field MR, and polarized light emission can be achieved in spintronic nanowires. Although there are many exciting progresses and potential applications of oxide-nanowire-based spintronic devices, considerable challenges remain to be resolved. The bottom-up nanowire synthesis with precise and reproducible controls on composition, morphology and physical properties is challenging for many key spintronic materials.

In terms of devices, there have been many revolutionary breakthroughs in spintronics, such as spin valves, magnetic tunnelling junctions, spin transfer torque devices, and spin field effect transistors. But there have been limited efforts on incorporating bottom-up oxide nanowires into spintronic devices. Difficulties in device fabrication include tailoring the interface spin transport, accurately positioning nanowires with respect to the other device components, and so on. The accelerating communications between communities of nanowires synthesis and spintronics hopefully can lead to the infusion of novel materials and device structures. Together with innovations in circuit design and system architecture, such endeavours may help overcome the challenges associated with the traditional electronics and lead to emerging spintronics technologies with better scalability, lower power dissipation, and increasing variability.

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