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Oxides for spintronics: A review of engineered materials for spin injection

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ABSTRACT

In this article we have reviewed the role of oxides in spintronics research, and specifically how these materials stand to further improve the efficiencies and capabilities of spin injection for active spintronic device development. The use of oxides in spintronics is advantageous in that they are stable in air, can be easily modified, and can possess a wide variety of properties which are beneficial to spintronics applications. This paper delineates the progression of spintronics and shows how applying oxide systems, in the form of half-metallic LaSrMnO₃, the diluted magnetic semiconductor ZnO:Co, and diluted magnetic dielectrics CeO₂:Co and Sm₂O₃:Co, has influenced and improved spintronics capabilities. An outline of the future potential for oxides in the realm of organic spintronic devices is also given. Copyright © 2014 VBRI press.

Keywords: Spintronics; diluted magnetic semiconductors; diluted magnetic dielectric; ferromagnetism.







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Introduction

It has long been postulated that through the utilization of spin degrees of freedom in addition to charge degrees of freedom, electronic devices have the potential to pass limitations currently posed by quantum effects [1-3]. Moore's law, a prediction which has correctly projected the biennial two-fold increase in the number of transistors in a single chip, will soon fail, as devices approach sizes bordering on atomic scales [1-4]. Due to these everlooming limits on device size, the development of spintronics has come to the forefront of research.

There are two main categories of spintronics devices, passive and active [5-7]. Passive spintronic devices make use of the spin degrees of freedom, whereas active spintronic devices employ both the spin and charge degrees of freedom. Current technology only utilizes passive spintronics. In passive spintronics, like magnetic RAM and magnetic read heads, the net polarization of the spins in a material is used for data storage and retrieval; active spin manipulation is not necessary (Fig. 1a) [8, 9]. In contrast, active spintronic devices rely on manipulation to generate versatile devices with the capabilities of both traditional electronics and passive spintronics (Fig. 1b) [5, 10]. The first step in the operation of a spintronic device is the insertion of charge carriers into a semiconductor. However, limitations in spin injection in semiconductor channels have hindered device efficiencies and stand as a barrier to active spintronics development.

Four main methods of spin injection have been investigated, which are shown in **Fig. 2**. The first involves

using a metallic ferromagnet to inject polarized spins into a semiconductor [11, 12]. The second method entails using a tunneling barrier to reduce impedance mismatches between the ferromagnet and the semiconductor [13]. The third method addresses materialistic mismatches by having the ferromagnetic injector itself be semiconducting [14-18]. The fourth method suggests use of a ferromagnetic tunnel barrier with a non-magnetic metal injector to promote enhanced polarization and efficiency [19-22]. Due to the versatility, stability, and ease of modification, various oxides have been adapted to use in these roles.

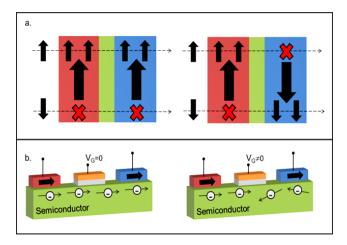


Fig. 1. (a) Schematic illustrating the two states (left-low resistance and right-high resistance) of a traditional passive spintronics device such as a magnetic read head. The resistance level is determined by the relative orientation of the two magnetic layers to one-another. If the two channels are aligned parallel to one another (left), electrons with parallel spin orientation can pass, whereas electrons anti-aligned cannot. For the two channels anti-aligned (right), both orientations of electrons are impeded in their motion through the junction. (b) Schematic showing the two states of an active spintronics device. In this structure, spin is injected and passes through a semiconductor base. If the spins align with the orientation of the magnetic contact at the drain, they can easily pass into it, acting like an "on" state in the transistor.

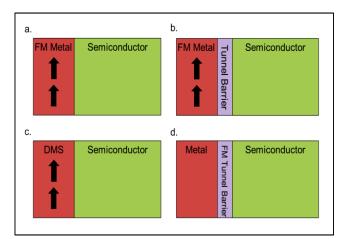


Fig. 2. (a) Spintronic device using a ferromagnetic metal and a semiconductor. Theoretically, polarized spin injection should result in a net polarized spin current injected into the semiconductor. (b) Using a half-metal where only one type of spin is present, injection through a tunnel barrier should be ideal. (c) Using a dilute magnetic semiconductor as the means of injection should reduce materialistic mismatches which cause problems in the metal-semiconductor structures. (d) Using a traditional metal, spin polarization can be completed using a dilute magnetic dielectric tunnel barrier, which preferentially allows for tunneling dependent on the spin orientation.

Ferromagnetic metals for spin injection

The first spintronics devices were relatively simple in design, consisting of a ferromagnetic (FM) metal contact used to inject spins into a semiconducting material (Fig. 2a) [11]. However, attaining high enough levels of injected spin polarization proved challenging. Efficiencies were often limited to less than five percent, even at temperatures below 10 K, due to low polarization and materialistic mismatches [8]. The failure of these devices can be accurately described by theoretical predictions made by Schmidt et al. [23]. In these estimations, Schmidt was able to describe the spin polarization within the semiconductor (α_2) as a function of spin polarization in the ferromagnetic metal (β). In this equation, eqn. (1), it shows that, even for relatively large spin polarization in the ferromagnet (sixty percent for example), spin polarization in the semiconductor is greatly limited and degrades quickly as a function of penetration depth,

$$\alpha_{2} = \beta \frac{\lambda_{FM}}{\sigma_{FM}} \frac{\sigma_{SC}}{\lambda_{SC}} \frac{2}{\left(2 \frac{\lambda_{FM} \cdot \sigma_{SC}}{x_{0} \cdot \sigma_{FM}} + 1\right) - \beta^{2}}$$
(1)

In fact, for spin polarizations less than 90%, spin injection depth will be limited to less than 100 nm [23]. Because this degree of spin polarization is typically impossible in traditional ferromagnetic metals, half-metals were proposed as a potential solution. Half-metals contain only one type of spin at the Fermi energy (Fig. 3), therefore 100% polarized spin injection should be feasible [24]. However, typical half-metals such as Heusler alloys are less than desirable due to their tendency to get oxidized and lose their half-metallic nature. As such, a half-metal oxide is highly desirable. One such oxide is half-metallic La_{0.7}Sr_{0.3}MnO₃ (LSMO). However, this setup did not prove exceptionally efficient, as impedance mismatches between the ferromagnet and semiconductor significantly hindered spin injection and, because LSMO is only half-metallic below room temperature, its use for commercial devices is extremely limited.

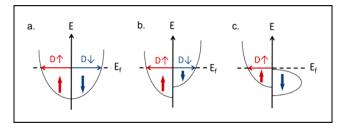


Fig. 3. (a) Density of spin states for a traditional metal with no spin imbalance. (b) A ferromagnetic metal where there is a difference of spins. (c) A half-metal where only one type of spin-state is available for injection.

In order to overcome this, tunnel barriers were introduced [13]. Often MgO or another insulating oxide such as aluminum oxide, tunnel barriers function as a conduit between the ferromagnetic metal and the semiconductor (Fig. 2b). By bypassing the impedance mismatch, more highly polarized spin injections can be achieved. However, maximum injection polarization was still completely reliant on the polarization of the injector. Another downside to a tunnel barrier device is a greatly diminished injected spin current. In order to avoid the need for a tunnel barrier, it was suggested that using a ferromagnetic semiconductor would remove impedance mismatch and enhance polarized current injection density.

Dilute magnetic semiconductors

In order to satisfy the need for a magnetic semiconductor for spin injection where none exist naturally, extrinsic magnetic doping of semiconductors using transition metals has been used for decades. These materials, also referred to as dilute magnetic semiconductors (DMS), can be formed by uniformly doping a semiconductor with a small concentration of a metal throughout the lattice in order to attain an intrinsically ferromagnetic material, ideally with room temperature functionality. With an initial influx of interest in the realm of spintronics, novel materials such as GaAs doped with Mn [18], ZnO doped with Ni [25] and Mn [26], GaN doped with Mn [27], and Mn doped InP [28] have been fabricated. However, in order for these materials to be of use in spintronic devices, room temperature ferromagnetism is a necessity. Theoretical predictions made by Dietl et al. [29] have indicated that both GaN as well as ZnO have the potential to be ferromagnetic at room temperature.

Because of the materialistic advantages of oxide-based systems; ZnO has become one of the most widely studied DMS based systems. Early reports of a ZnO:Co were given by Ramachandran et al. in 2004 [16]. They reported the formation of phase pure thin films grown by pulsed laser temperature deposition which showed room ferromagnetism. The critical formation of phase pure materials was supported by high resolution TEM (HRTEM) and X-ray diffraction measurements. By illustrating the formation of a pure ferromagnetic semiconductor and providing a delineation of the mechanism behind its ferromagnetic behavior, it has been made possible to further research and study DMS systems for spintronics application.

As shown in **Fig. 2c**, these materials are advantageous in that they allow a simplified device consisting of only the spin injector and the active semiconducting layer. Because the injector and active layer are both semiconductors, the impedance mismatch between the two is greatly diminished. This allows a possibility for nearly unhindered polarized spin current to be injected into the active layer. However, DMS have low coercivity, which will require application of an external magnetic field for device operation.

Dilute magnetic dielectrics

The fourth approach proposed for injecting spin into an active spintronic device eliminates the need for a metallic ferromagnetic contact. By using a ferromagnetic tunnel barrier, spin filtration will occur on tunneling from the metal to the semiconductor. For this tunnel barrier, illustrated in **Fig. 2d**, a dielectric material doped with transition metal is used. This unique class of material, known as dilute magnetic dielectrics (DMD), involves

doping dielectric material with ferromagnetic transition metal, and creates a final barrier which is both electrically insulating as well as intrinsically ferromagnetic.

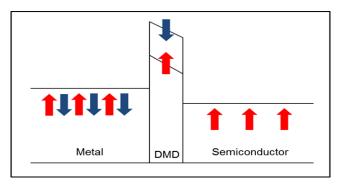


Fig. 4. Schematic illustrating the function of a DMD tunnel barrier for polarized spin injection. The energy required for tunneling is dependent on the spin orientation, thereby enabling polarized spin injection for a non-polarized stream of electrons.

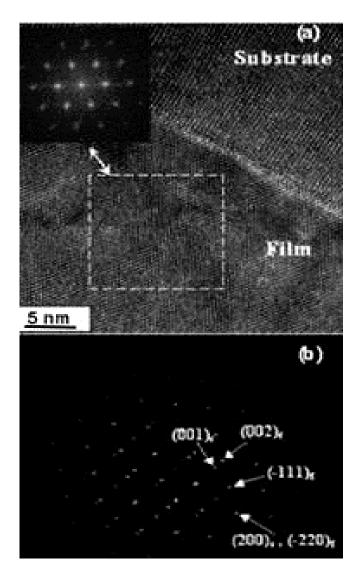


Fig. 5. High resolution TEM image of CeO₂:Co thin films showing no cluster formation [19]. [Reprinted with permission from "Tiwari, A.; Bhosle, V. M.; Ramachandran, S.; Sudhakar, N.; Narayan, J.; Budak, S.; Gupta, A. Appl. Phys. Lett. 2006, 88, 142511." Copyright 2006, AIP Publishing LLC.].

The goal of a DMD is not to act as a spin injector, but rather to function as a spin filter for polarized injection. This works because the energy barrier for tunneling through DMDs varies directly with spin orientation as is illustrated schematically in **Fig. 4**. As both spin orientations of electrons reach the DMD from the metal, only those with the lower energy requirement for tunneling are capable of passing through into the semiconductor. Because of this, spin polarization and efficiency can increase drastically compared to other devices.

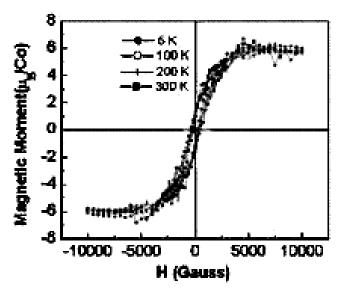


Fig. 6. Magnetization loop for temperature between 5 and 300 K **[19]**. [Reprinted with permission from "Tiwari, A.; Bhosle, V. M.; Ramachandran, S.; Sudhakar, N.; Narayan, J.; Budak, S.; Gupta, A. Appl. Phys. Lett. 2006, 88, 142511." Copyright 2006, AIP Publishing LLC.]

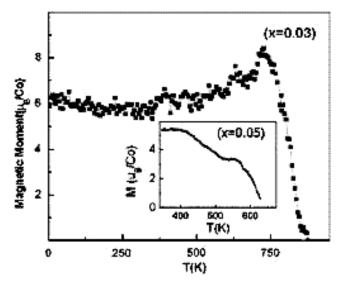


Fig. 7. Saturation magnetization vs. temperature for CeO₂:Co thin film [19]. [Reprinted with permission from "Tiwari, A.; Bhosle, V. M.; Ramachandran, S.; Sudhakar, N.; Narayan, J.; Budak, S.; Gupta, A. Appl. Phys. Lett. 2006, 88, 142511." Copyright 2006, AIP Publishing LLC.]

One of the most researched materials is rare-earth oxide CeO_2 , which is dilutely doped with transition metal element. As reported by Tiwari *et al* . [19], a ferromagnetic doped sample has been achieved using pulsed laser deposition. Through use of TEM, as shown in **Fig. 5**, it was shown that the samples were phase-pure.

Preforming subsequent magnetic (SQUID) measurements, illustrated as in **Fig. 6**, it was shown that at room temperature, these materials possessed a giant magnetic moment of $\sim 6.8 \mu_B$. Magnetic measurements taken as a function (**Fig. 7**) of temperature gave a Curie temperature of approximately 725 K [19].

Much of the current interest surrounding DMDs is regarding the mechanism behind their ferromagnetism. The desired state is one that results in intrinsic ferromagnetism. This kind of magnetism is characteristic of a phase-pure material, where the magnetic material has substituted uniformly throughout the lattice. However, extrinsic magnetic behavior can also occur. This magnetism indicates that ferromagnetic dopant metal has not substituted into the lattice, but has formed magnetic clusters. In order to reiterate that the behavior seen is due to intrinsic doping, not extrinsic, Prestgard *et al* . [**30**] has reported the formation of non-phase-pure CeO₂:Co thin films.

These films containing cobalt clusters were intentionally fabricated via a pulsed laser deposition technique. The existence of these clusters was supported through x-ray absorption spectroscopy (XAS) and x-ray photoelectron spectroscopy (XPS) measurements, the results of which are reported in **Fig. 8**. In order to determine the degree of cluster formation as compared to substitutional cobalt, the resulting data were compared to standards of Co^0 , Co^{2+} , and Co^{3+} . This comparison was used to generate a linear combination fit (LCF), also shown in **Fig. 8**, which mapped the contribution and indicated that almost 86% of the cobalt was in the form of clusters and the remaining 14% existed substitutionally in the lattice.

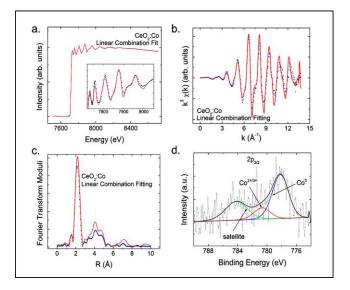


Fig. 8. (a) XAS data for the Co K-edge of the CeO₂:Co film (as data points). The linear combination fitting model is shown as a line. This fitting indicated that ~86% of the cobalt was in metallic cluster form, and ~14% was Co^{3+}/Co^{2+} form (12% Co^{3+} , 2% Co^{2+}). Inset shows a close up of the XAS fine-structure. (b) Illustration showing the K-space transformation of **Fig. 8** a. (c) The R-space Fourier transform of the above data (both the fit and the experimental data). (d) XPS data showing the presence of mixed valence Co (2+/3+) as well as metallic cobalt.

XPS measurements supported these findings through investigation of the $2p^{3/2}$ Co state. Subsequent SQUID measurements were performed on these films, shown in

Fig. 9, which is exhibited superparamagnetic behavior with a saturation magnetization of $2.4\mu_B/Co$ [30]. This result is in direct contradiction to the ferromagnetic response seen in previously reported phase-pure samples. These results not only showed that previous ferromagnetic behavior in DMD systems is due to intrinsic magnetism, but also illustrated the unique superparamagnetic behavior in extrinsically-magnetic clustered systems.

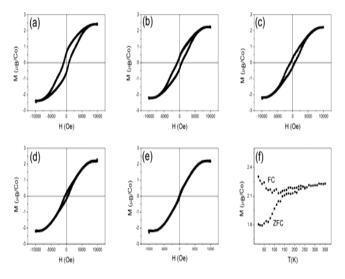


Fig. 9. Magnetization loop for CeO₂:Co thin films with cobalt clustering, (a) at 10K, (b) 50K, (c) 100K, (d) 200K, (e) 300 K. (f) shows the magnetization versus temperature in the zero field cooled (ZFC) and the field cooled (FC) sweeps [30]. [Reprinted with permission from "Prestgard, M. C.; Siegel, G.; Ma, Q.; Tiwari, A. Appl. Phys. Lett. 2013, 103, 102409. "Copyright 2013, AIP Publishing LLC.]

In a related system, Sm₂O₃, the first observance of dynamic superparamagnetism has been reported [31]. Similar to the clustered CeO₂ system, these Sm₂O₃ thin films have illustrated behavior which is superparamagnetic in nature. However, TEM experiments have shown that no clustering of the transition metal dopant is occurring. Rather, there is clustering of polarons around defect sites, which has resulted in this unique dynamic superparamagnetic behavior. Again, these results have shown that transition metal doping of rare-earth oxides will exhibit superparamagnetic behavior when clusters form and ferromagnetic behavior when their doping is uniform.

Prospects for future research

Spintronics devices may hold the key to advancing the computing power of electronics long beyond the limitations of Moore's law. Oxides will provide an important role in this advancement due to their capacity to fulfill any role in a spintronics device. By achieving a better understanding of oxide systems and further improving current oxide capabilities, spintronics technologies have the potential to transition from the laboratory to full-scale production in the near future.

Current research in the realm of spintronics has also begun to include organic materials. Organic materials have several advantages over traditional inorganic materials such as lower cost, higher flexibility, and even transparency. Though the research on organic spintronics has just started, quite significant progress has already been made in this field. For example Xiong *et al.* [32] reported giant magnetoresistance in organic spin valves using π -conjugated organic semiconductors (OSEs). Spin organic LEDS (spin-OLED) have likewise been developed [33, 34]. Despite all this progress, current organic spintronics devices lack room temperature capabilities which seriously inhibit their prospect for real life applications [35]. If recent research is a good indicator, further exploration in this field using oxide materials may hold the key to room temperature functionality of spintronic devices.

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