Chapter

Ferromagnetism in SnO_2 Doped with Transition Metals (Fe, Mn and Ni) for Spintronics Application: A Review of Experimental Status

Yared Worku, Dipti Ranjan Sahu and Vijaya Vallabhapurapu Srinivasu

Abstract

Due to their potential application in the field of spintronics, the discovery of various types of oxide-based dilute magnetic semiconductors (ODMS) materials that might work at practical room temperature ferromagnetism (RTFM) has recently attracted great attention. Among ODMS materials, transition metal™ doped tin oxide (SnO_2) compounds are important for the investigation of ferromagnetism due to its special important property such as high chemical stability, high carrier density, n-type behavior and trait long range ferromagnetism. However, the question of understanding the mechanism of ferromagnetism (FM) process is still not fully understood in these materials, due to unable to know exactly whether its FM property arises from the nature of the intrinsic property or secondary phases of the material. According to the results from many literature surveys, the mechanism of magnetic ordering responsible for magnetic exchange interaction in these materials is highly affected by oxygen vacancy, defects, dopant types and concentration, temperature, sample preparation method and so on. In this chapter, we reviewed the mechanism of ferromagnetism observed of Ni, Mn and Fe-doped SnO_2 materials.

Keywords: spintronics, diluted magnetic semiconductors (DMS), room temperature ferromagnetism (RTFM), TM doped tin oxide, oxygen vacancy and defect

1. Introduction

Current semiconductor-based electronics device uses only the electron charging property to perform a particular feature in which the electron spin degree is completely ignored [1]. The spin property of an electron which is associated with an intrinsic angular momentum of the electron provides new effects and new functionalities to electronics materials based on Spintronics principle [2, 3]. Spintronics deals with the role played by the spin of an electron associated with its magnetic moment, as well as the charge degree of electron [4]. Spintronics devices have several important applications compared to non-spin based electronics device, such as consume

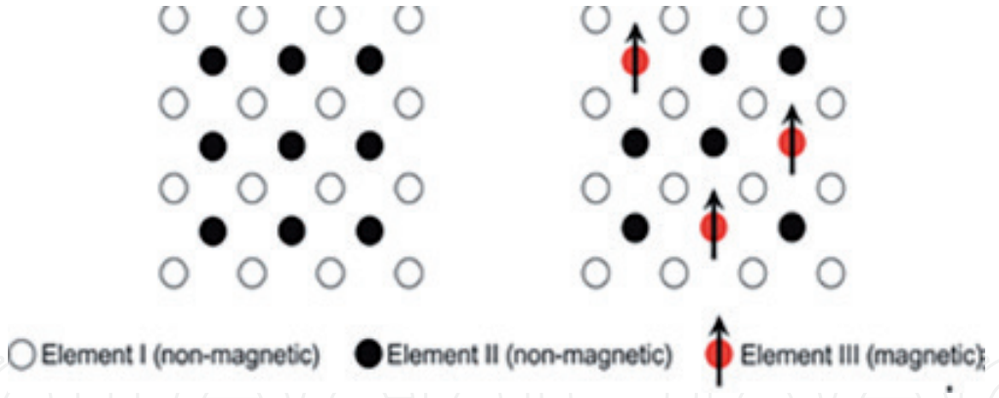

Figure 1.
Schematic view of a non-magnetic (left) semiconductor and a diluted magnetic semiconductor (right) [5].

less power of electricity, fast data processing speed, their memories are non-volatile [4]. Starting from metal-based technology, the research area of Spintronics shifted to the recent development of diluted magnetic semiconductors (DMS) materials which are compatible with standard semiconductor based electronics device**.** DMS are materials prepared through which a certain amount of the cations in a host semiconductor are partially replaced by transition metal ions (Mn, Ni, Co, Fe, Cr) as shown in **Figure 1** [5] as a result the materials attains both semiconducting and magnetic property which is makes these materials advantageous and applicable for Spintronics application. The total ferromagnetic behavior of these materials is linked to the interaction of the spin of the magnetic ions with the itinerant carriers [6–8]. DMS are important materials in the sense that logic, communications and storage operation can be achieved within the same materials technology [9, 10]. The property of achieving RTFM is one of the most important factors that determined DMS material to be used for practical spintronics application [7], The sp-d exchange mechanism between the d states of the TM doping and sp free carriers as well as the double exchange mechanisms are the main factor in the production of ferromagnetism in ODMS materials between d states of TM ions [11]. Among DMS materials oxide based DMS materials such as TM doped with HfO_2, TiO_2, ZnO and SnO_2 are more advantageous than normal DMS materials and have important magnetic properties arises from a large sp-d exchange interactions between the magnetic ion elements and band electrons [9, 10, 12, 13]. ODMS has important special properties such as having high n-type carrier concentrations wide band gap, light transparency, capability to be grown at low temperatures, ecological safety and cheap [14–16]**.** Due to its n-type semiconductor, good conductivity, high carrier density and high chemical stability, SnO_2 doped with TM is particularly promising materials for spintronic applications [17, 18]**.** SnO_2 naturally existing in cassiterite form and it has tetragonal rutile structure and its wide band gap is about 3.6 eV [19, 20]. SnO_2 has many technological applications, including gas sensors, solar cells, heat reflectors, lithium ion batteries and other optoelectronic devices [21–23].

2. Ferromagnetism in oxide-based DMS

In the recent years the research field of RTFM in O-DMS has got more attention and many kinds of compounds have been discovered [24]. However, the idea behind the original source of ferromagnetism in these materials is not well understood a not complete it becomes the most challenging area in solid state physics [25]. Several groups have stated that the mechanism behind ferromagnetism in most O-DMS materials is the material's intrinsic property itself or the direct and indirect

interaction between only magnetic impurities and magnetic impurity ions through oxygen vacancies [10, 26–30]. Recently, various experimental methods have been used to study the magnetic properties of DMS materials, in particular the vibrating sample magnetometer (VSM), the superconducting quantum interference device (SQUID), the physical property measurement system (PPMS) and the electron spin resonance (ESR) techniques. According to the results from many literature indicated that sample preparation, growth conditions, dopant type and concentration, co-doping effect, oxygen vacancies, defects and crystal structure has played a role for the magnetic behaviors observed in ODMS material [31–37]. Some scholars reported that vacancy-induced magnetism has been played a major role for the observed ferromagnetism in undoped SnO_2 [38]. In some cases SnO_2 thin films does not shows RTFM when doped with 3*d* cations rather show when doped with Mn, Cr, Fe, Co, or Ni [39–41]. Similarly undoped SnO_2 did not shows FM behavior. However, the doped SnO_2 shows FM behavior at higher doping level completely removes the ferromagnetic behavior of the doped one [31, 42]. As shown in **Figure 2** the improvement of magnetization by co doping (Ni-Mn, Fe-Co, Fe-Ni and Fe-Mn) in tin oxide has been reported and the mechanism of FM is due to double exchange interactions occur via oxygen vacancies [11, 43]. The electronic or lattice defects of the materials associated with the intrinsic nature of the materials can be responsible for the high temperature FM of TM doped SnO_2 [39]. Despite much experimental success, the idea behind FM in most O-DMS is controversial. Here, we present a brief review of the Fe, Ni and Mn Doped SnO_2 system experimental work.

2.1 Ferromagnetism in Mn-doped SnO_2

Mn-doped SnO_2 is an excellent candidate and promising materials for RTFM study, but only very little work has been reported so far compared to others. Among other preparation methods sol-gel preparation technique is best method for preparation of TM doped SnO_2 thin film and nano structures [44, 45].

2.1.1 Experimental

$SnMnO_2$ thin film is prepared by sol-gel method according to the literature reported [44]. The solution was prepared by dissolving a certain amount of tin tetrachloride $SnCl_4$ and manganese nitrate hydrate [Mn $(NO_3)_2 6H_2O$] in distilled water and

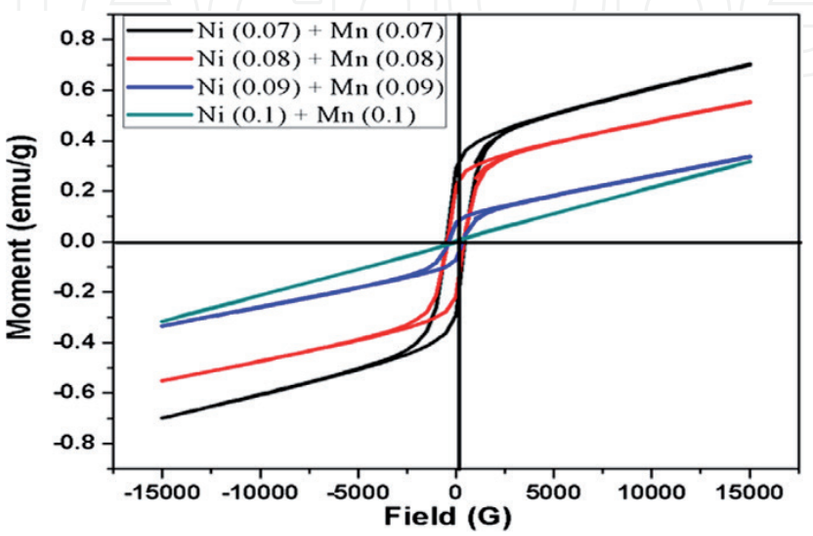

Figure 2.
M-H curves of (Ni-Mn) co-doped with SnO_2 [11].

ethanol respectively and stirring for 5 hour and aging for at least a week, the prepared solution was spin-coated on silicon substrate and heated at 120°C for 25 min. The film precursors were obtained after multilayer coating. Finally, to obtain $SnMnO_2$ thin films, the precursors of films were calcinated in atmospheric air at 5000°C for an hour.

As reported by Tian et al., the chemical co-precipitation method was used to synthesize Mn doped SnO_2 nanoparticles [46]. First, appropriate quantities of $SnCl_2$ and manganese acetic acid were dissolved in ethanol solution, then a few drops of HCl solution were applied to ensure dissolution. Then a 10 M ammonium bicarbonate solution with continuous stirring at 60°C was applied to the mixture solution until a pH of 9 was reached. After being distributed by ultrasonic for 15 min to get nano-crystalline powders. The resulting precipitation washed to clean the impurities and dried in air at 150°C. Finally, the nano crystalline powders were sintered in the air for 3 hours. X-ray diffraction (XRD) recorded the crystal structure of the synthesized $SnMnO_2$ thin film as shown in **Figures 3** and **4**. The study magnetization property and RTFM were performed using a superconductive quantum interference device (SQUID) and vibrating sample magnetometer (VSM) respectively.

2.1.2 Result and discussion

According to various reports, the origin of the observed FM in Mn-doped SnO_2 depends on a number of factors; some reported that Mn- SnO_2 prepared by PLD method exhibits the only paramagnetic behavior [47]. Similarly, others reported that the dopant Mn does not contribute any role for the observed FM behavior of Mn-doped SnO_2 films; it is assumed that oxygen vacancies and defects are the main factors contributing to the FM order in the system as shown in **Figure 5** below [48]. Others report on Mn-SnO_2 powders confirmed that the observed FM property is likely the results from oxygen vacancies, and Mn doping has only a significant role of the observed source of RTFM in the materials [49]. High level of Mn dopant can degrade the FM behavior where as small doping concentration intrinsic defects can act as a source of the FM in Mn-doped SnO_2 due to a very large magnetic moment [39]. On the other hand, a study reported on Mn doped SnO_2 nanoparticles shows sintering temperature and doping concentration can affects the magnetism of the materials system [46]. As shown in **Figure 7** that Mn-doped SnO_2 powder that concentration of Mn ion contributes to a decrease in the average magnetic moment of magnetic ions, this is due to the competition between the super-exchange antiferromagnetic coupling and the F-center coupling mechanism [50]**.** Similarly, other research on Mn-doped SnO_2 thin films

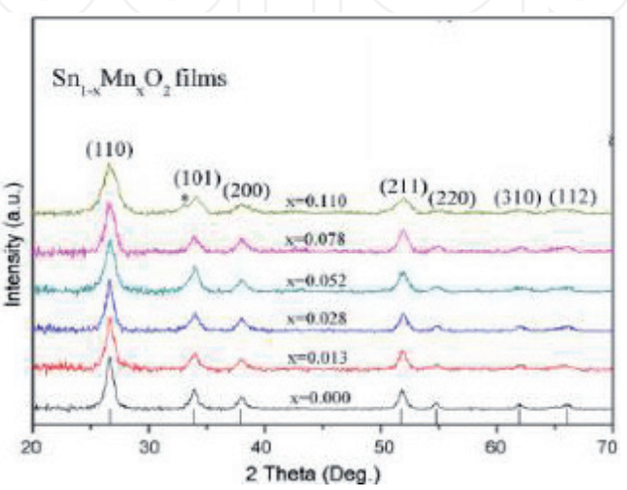

Figure 3.
XRD patterns of $SnMnO_2$ films [44].

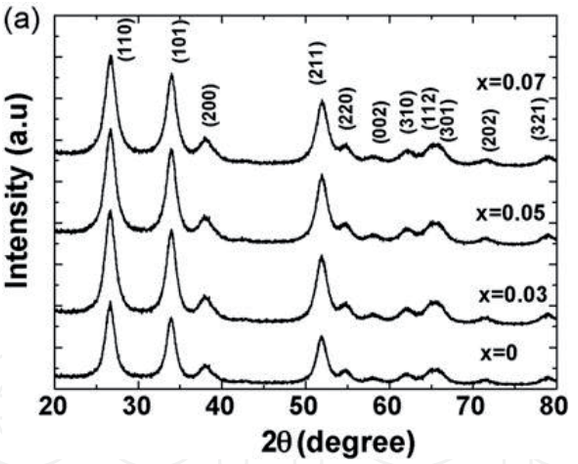

Figure 4.
XRD patterns of $SnMnO_2$ nano particle [46].

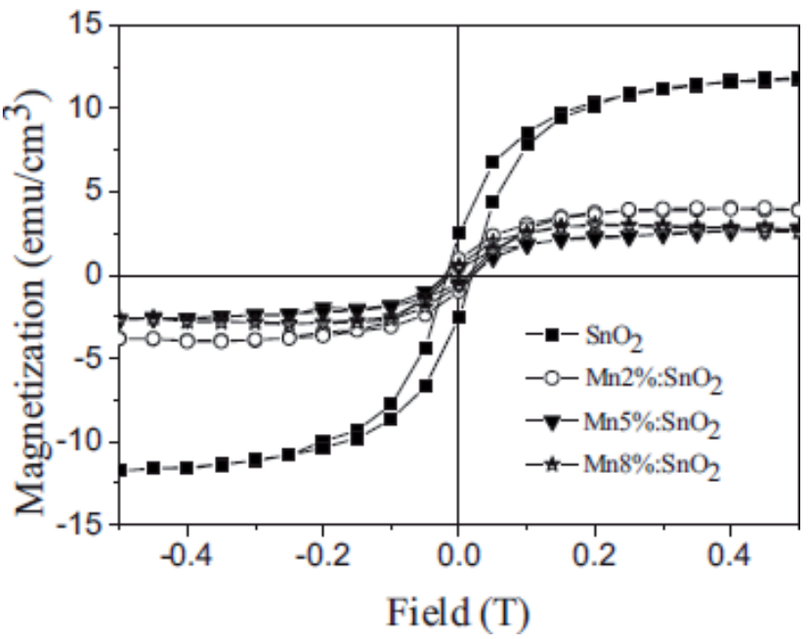

Figure 5.
RTFM for undoped and Mn doped tin oxide thin films, room temperature ferromagnetism.

synthesized by sol-gel method show that dopants and electronic cloud interactions play a significant role in establishing FM [44]. The ferromagnetic property of Mn doped SnO_2 confirmed that BMP's overlapping, oxygen vacancies and F-center exchange interaction are the cause for the existence of ferromagnetic behavior in in pure and doped Mn doped SnO_2 materials [51]. The increment of Mn concentration lead to the decline of magnetic moment of the origin of ferromagnetism behavior in Mn-doped SnO_2 films is explained BMP and the average magnetic moment per Mn concentration decreases with increasing Mn content [44]. Overall, the origin of FM in Mn doped SnO_2 system is still controversial and there is no such exact cause FM in this material.

2.2 Ferromagnetism in Fe-doped SnO_2

2.2.1 Experimental

Rodrı et al. reported that Fe-SnO_2 thin films on the $LaAlO_3$ subtract were synthesized with PLD techniques. The doped SnO_2 target was synthesized with metallic Fe powders and SnO_2. The powders were mixed with a ball-mill for 2 minutes,

then pressed uniaxially (200 MPa) into a disk and finally sintered at 1000°C [52]. The crystallographic structures of the prepared Fe doped SnO_2 were determined by X-ray diffraction (XRD) and the magnetic measurements were performed with superconducting quantum interference device (SQUID).

2.2.2 Result and discussion

There have been reports of Fe doped SnO_2 in which the ferromagnetic interactions between magnetic impurities mediated oxygen or free carriers in the Fe doped SnO_2 system responsible for forming FM. Similarly, defects in undoped SnO_2 semiconductors may contribute to the observed ferromagnetism [33, 53]. Similarly both undoped and Fe-doped SnO_2 thin films shows the observed FM property is due to oxygen vacancies near Fe increased the magnetic moment, the RTFM behavior observed in the SnO_2 film must be associated with the sample shape or to defects incorporated during film growth and, part of the magnetism observed in SnO_2 as shown in figure [52]. The results from Fe-doped SnO_2 powders prepared by polymerized complex method confirms that the annealing temperature contributes to decline of magnetic saturation which is related to the defects rather than from dopant iron sites shown in **Figure 6a** below [54, 55] the existence of vacancies and defects in the grain boundaries and interfaces in Fe doped SnO_2 nanoparticles leads to decline the ferromagnetic behavior of system [56]. The decrease in Fe ion's magnetic moments, with their doping concentrations increasing, The super-exchange interaction may result in the interaction between neighboring TM-ions of the anti-ferromagnetic form, resulting in the observed decrease in the magnetic moment with increased concentration of TM as shown in **Figure 10** [57]. The work of the other group reporting shows that the lattice distortion induced by co-doping Fe-SnO_2 enhance ferromagnetic saturation magnetization of the compared with not co-doped one [26]**.** As shown in **Figure 7** oxygen vacancies has a great impact on the FM property of Fe doped SnO_2 [58]. The ferromagnetic behavior of Fe-doped SnO_2 thin films is caused by the coupling of ferric ions through an electron trapped in an oxygen vacancy [59]**.** In some cases the introduction of iron in semiconducting nano particles SnO_2 is responsible for appearance of paramagnetic behavior of the system that is due to weak antiferromagnetic interaction [60]. The decline of antiferromagnetic interaction in Fe-SnO_2 nano particles was reduced by the increment of Fe concentration [61]. Some study reported that the magnetic properties Fe-doped SnO_2 nano powders shows that the an increased Fe concentration leading to the reduction of oxygen-related vacancy changes magnetic property to paramagnetic

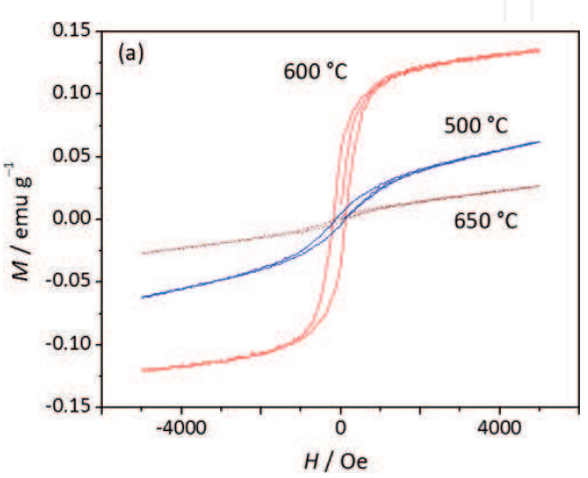

Figure 6.
Fe doped SnO_2 magnetic hysteresis annealed for certain hours at different temperatures [54].

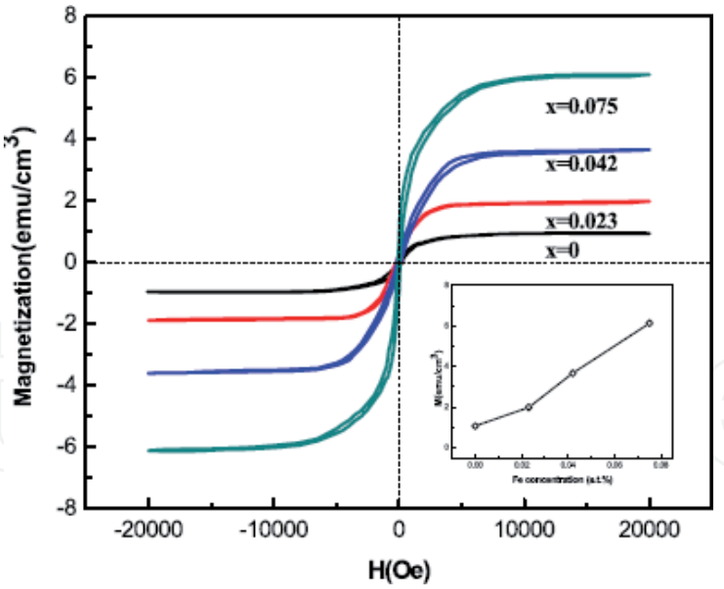

Figure 7.
Sn_{1-x} Fe_xO_2 film hysteresis loops with different concentration at 300 K. The inset is the difference in the concentration of Fe doping in saturation magnetization [58].

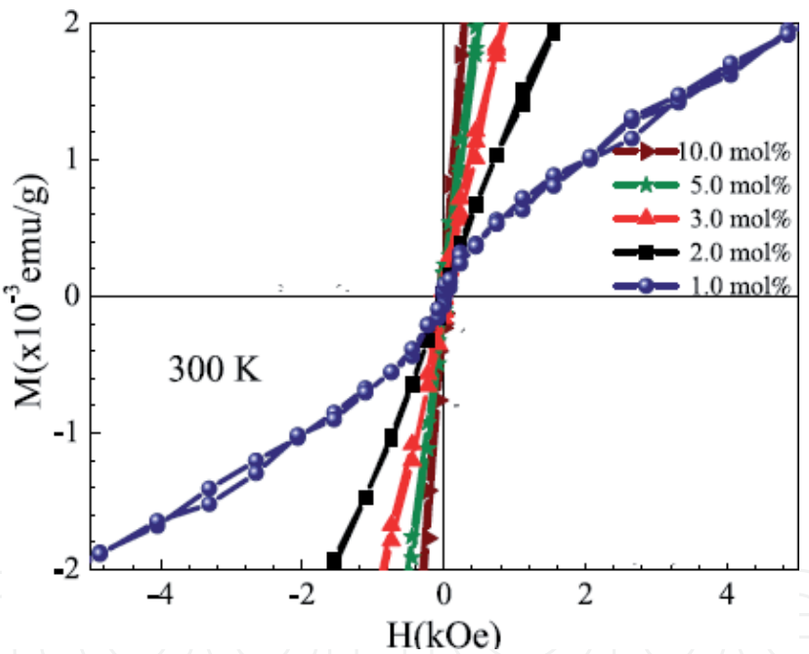

Figure 8.
RT magnetization versus magnetic field curves for the Fe-doped SnO_2 nano powders [62].

system as shown in **Figure 8** and the FM interactions is based on Bound Magnetic Polarons (BMPs) formation [62]. Sometimes the host systems SnO_2 and SnO doped with Fe during sample preparation can affect the observed magnetic properties of the system [63]. Another study confirms that changes in temperature play a role in the magnetic transition from paramagnetic to ferromagnetic behavior at ambient temperatures and low temperatures.

2.3 Ferromagnetism in Ni doped SnO_2

2.3.1 Experimental

As reported in the literature $SnNiO_2$ films were prepared by sol-gel method [64] the same procedure as [44]. The solution was prepared by dissolving $SnCl_4$ and

$NiCl_2{\cdot}6H_2O$ in distilled water and ethanol. For the film preparation, to get the thin films the solution was spin-coated on silicon substrate. As reported by [57] undoped and Ni doped SnO_2 prepared using a method of co-precipitation, the solution was prepared by dissolving $SnCl_4.5H_2O$ and $NiCl_2.5H_2O$ properly into de-ionized water. After the white precipitates were obtained, ammonium hydroxide (NH_4OH) was added with stirring to the solution. The resulting mixtures were washed with de-ionized water to remove unwanted ionic impurities that may develop during the process of synthesis. Such washed precipitates were dried in air and Ni doped SnO_2 powder products were eventually obtained.

Detail crystallographic structures of the prepared $SnNiO_2$ thin films and powders were carried out using XRD and the details of the magnetic properties were probed by vibrating sample magnetometer (VSM) measurements.

2.3.2 Result and discussion

According to recent experimental investigation of RTFM on Ni doped SnO_2 has made it important and promising materials for spintronics application [64–68]. The observed FM in these materials is linked to oxygen vacancy and structural defects of the materials [67]. In some cases, nano-crystalline Ni doped SnO_2 exhibits Paramagnetic character [68]. As shown in **Figure 9** the super-exchange interaction may result in an anti-ferromagnetic type interaction between neighboring TM-ions, resulting in a decline in magnetic moment with an increase in TM concentration [57]. In some studies the introduction of more Ni doping concentration leads for reduction of magnetic moment Ni ion of because the antiferromagnetic super-exchange interaction among closest neighbor in Ni^{2+} ions in Ni doped SnO_2 samples the BMP model can explain for RTFM on these systems on the other hand nickel (Ni) doped SnO_2 powder shows a substitution of Sn atom by Ni atom interstitially lead to the appearance of diamagnetic state [64]. Kuppan et al. shows that oxygen vacancy around magnetic impurity plays a major role in establishing ferromagnetism in Ni doped SnO_2. Nevertheless, saturation magnetization slowly decreases with a persistent rise in Ni doping concentration [68]. Thus we strongly feels that

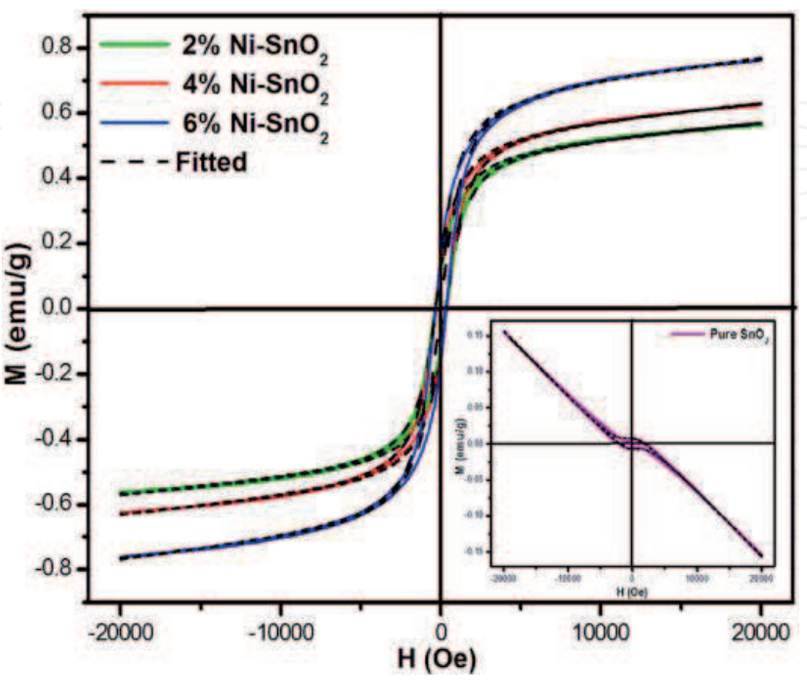

Figure 9.
The RTFM hysteresis of $SnFeO_2$ and $SnNiO_2$ at different doping concentration [57].

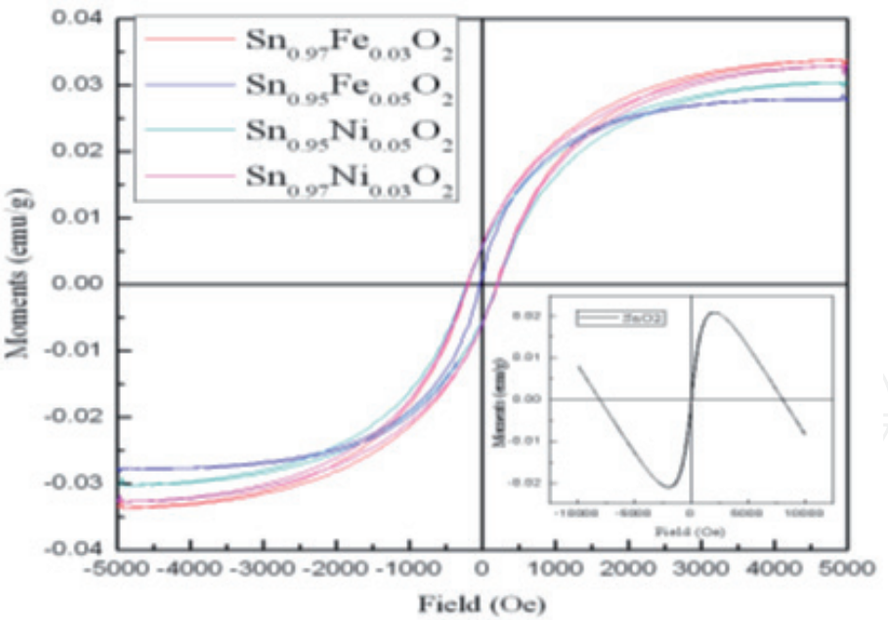

Figure 10.
Room temperature magnetization for pure and Ni doped SnO_2 NPs [69].

the oxygen vacancy and or defects in the Ni doped SnO_2 system. Similarly a report from Ni-doped SnO_2 nanoparticles synthesized by a polymer precursor method demonstrated that doping small amount of Ni doping concentration can push defect-related FM while introducing high Ni concentration favors the paramagnetic phase stabilization [70]. Similarly oxygen vacancy and defects on Ni doped SnO_2 thin film contribute for the formation RTFM [71]. As shown in **Figure 10** some studies have confirmed that Ni ions doping creates numerous defects or oxygen vacancies in SnO_2 nanoparticles in order to introduce RTFM in SnO_2 nanoparticles [69]. Some reported that substrates on thin film deposition have a strong impact on the magnetic moment of these material and the result confirmed that that FM in the films is as result of the doped matrix grown in different substrates [65]. In some cases the decrease of magnetic moment of per Ni ion is observed with the introduction of more dopants Ni ions that is associated with antiferromagnetic superexchange interaction between in Ni ions in the system [64, 72]. Some reported that the mechanism of the observed FM in nickel (Ni) doped tin oxide thin films can be explained in bound magnetic polaron (BMP) mode [73].

3. Conclusion

Most of the results reported in the review shows that the perfect mechanism of induced FM in Mn, Fe and Ni doped SnO_2 is related to the intrinsic nature of the material itself, especially oxygen vacancies and defects of the crystal formed during sample preparation and doping magnetic impurity influence the magnetism of the systems. Even though, a different type of FM is reported. However, the different reported results are contradictory with each other and further research is needed to bring new solution for the contradiction idea behind FM.

Author details

Yared Worku[1], Dipti Ranjan Sahu[2*] and Vijaya Vallabhapurapu Srinivasu[1]

1 Department of Physics, University of South Africa (UNISA), Johannesburg, South Africa

2 Department of Natural and Applied Sciences, Namibia University of Science and Technology, Windhoek, Namibia

*Address all correspondence to: diptirs@yahoo.com

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