RESEARCH REVIEW JOURNALS
Volume-04
Issue-04
April -2019

ISSN: 2455-3085 (Online)
RESEARCH REVIEW International Journal of Multidisciplinary
www.rrjournals.com [UGC Listed Journal]

Multifunctional Materials Science: An Experimental Study

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ARTICLE DETAILS

Article History
Published Online: 15 April 2019

Keywords
Multifunctional Materials, Difficulties, mutually exclusive, parameters, ferromagnetic, microscopic mechanism

ABSTRACT

One particular obstacle to the development of these materials is that the coexistence of ferromagnetic and ferroelectric properties is normally contra-indicated. Further studies are required to understand the microscopic mechanism producing this coexistence and coupling between typically mutually exclusive ferroic order parameters.Magneto electric multiferroics provide an important advantage in designing new devices, due to the control of magnetic order using electric field. This may lead to the development of a new class of spin based electronics, magnetic sensors, transducers, and attenuators. These technological benefits have made magneto electric multiferroics a very active area of research and have spurred the search and design of new multiferroic materials. The recently revived interest in a different class of multifunctional materials, in which two or more ferroic properties, being ferro or antiferroelectricity, ferro or ant ferromagnetism and ferro or antiferroelectricity coexist in what are known as multiferroic materials is driven both by the potential applications of these systems as well as a desire to more deeply understand the physics underlying this behavior. Among these multiferroic materials, those displaying anti/ferromagnetic and anti/ferroelectric properties are known as magneto electric multiferroics.

1. Introduction

The magnetic interaction among local spins on these magnetic ions together with the charge carriers gives rise to ferromagnetism in these systems. These DMSs are promising materials for spintronic device applications provided their Curie temperature Tc is above room temperature. A number of DMS systems have been identified and are widely studied including Mn doped InAsand GaAs which have Curie temperatures of 35 K and 170 K respectively. This low magnetic transition temperature makes these materials impractical for device applications but these materials have helped to guide the search for new DMS materials with ferromagnetic order above room temperature.This flexibility in tuning the carrier concentrations and type of charge carriers in semiconductors make them useful for numerous microelectronic applications. In many cases, it is difficult to avoid introducing various defects during and subsequent to the fabrication process leading to highly degenerate systems. These accidental charge carriers cloak the intrinsic materials properties of the semiconductor and may make them unsuitable for the desired application. We will discuss the effect/influence of numerous defects introduced during and and after the growth process for highly degenerate n-type InN and In_2O_3 compound semiconductors and will explore some possible application for these highly degenerate semiconductors.

2. Orign of magnetism in dilute magnetic semiconductors

The DMSs can be considered as magnetically inactive host semiconductor with localized spins, arising from dopants or defects. The host semiconductor may also be doped with either electrons or holes acting as mobile charge carriers, which can promote carrier mediated magnetism in these systems. The magnetic interactions in these systems are usually determined by the speed electronic exchange, where the s and p orbitals belong to the host and carrier donors and d orbitals to the transition metal ions. Electronic exchange interactions through carriers in the conduction or valence bands in the system may give rise to the magnetic ordering.

3. Multiferroics and microscopic origin of multiferroicity

There was renewed interest in multiferroics after N. Hill papers highlighting that magnetism and ferroelectricity are contraindicated due to the chemical incompatibility of the physical origin of these ordering phenomena. In general magnetism arises from the presence of localized electrons in partially filled d or f shells of transition metal or rare earth ions. The exchange interaction between these local moments gives rise to magnetic ordering. Conversely cations having empty d-shells is required for ferroelectricity. For example, the ferroelectricity in BaTiO3 is due (in large part) to the off-center displacement of Ti ion. This off-center displacement is only possible since the Ti4+ ion has an empty d-shell. This competition is referred as d 0 vs d n problem. Therefore, different microscopic mechanisms are necessary to explain the onset of ferrelectricity in multiferroic materials. D. Khomskii subdivided multiferroics into type-I and type-II multiferroics, based on their magnetic and ferroelectric ordering temperatures. Those multiferroics for which the ferroelectric and magnetic ordering temperatures are well separated are classified as type-I multiferroics. In these multiferroics the origins of magnetic and ferroelectric ordering are independent anddistinct. Two important and widely studied examples of type I multiferroics are $BiFeO_3$ and $YMnO_3$. Multiferroics where the magnetic order produces the ferroelectricity, so that the ferroelectric ordering temperature coincides with a magnetic transition, are called type-II multiferroics. Some examples of type-II multiferroics are $Ni_3V_2O_8$, $FeVO_4$, $TbMnO_3$.

4. Multiferroics with collinear magnetic structure

In this class of multiferroics, the Ferro electricity is driven by a collinear magnetic phase. In this collinear magnetic phase all magnetic moments are aligned along a particular direction without involving the spin orbit interaction. The magnetically induced polarization in these systems is due to the exchange striction because the magnetic coupling varies with atomic positions. The recently discovered Ca3CoMnO6 multiferroic exhibit this behaviour. This consists of 1D chain of alternating Co2+ and Mn4+ ions. Above the magnetic ordering temperature, the distance between ions along chain is constant, which does not favour polarization. However, below the magnetic ordering temperature, a combination of alternating charge ordering of transition metal ions of different valence, a similar situation as explained in"Electronic Ferro electricity" section, and an Ising-like magnetic structure ↑↑↓↓ can give rise to Ferro electricity via exchange striction. This exchange striction is due to competition between nearest neighbour (NN) ferromagnetic and next nearest neighbour (NNN) antiferromagnetic super exchange interactions.

5. Multifunctional magnetic semiconductors

Magnetic semiconductors exhibit both magnetic and semiconducting properties. One general approach to make a magnetic semiconductor is to replace a small fraction of cations in the non-magnetic parent semiconductor by magnetic ions, which produces a dilute magneticsemiconductor (DMS). A schematic representation of this process is shown in Fig. 1

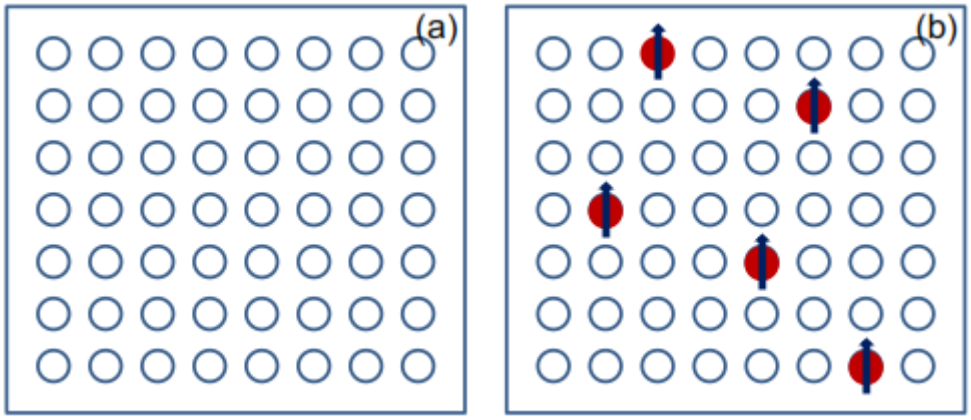

Figure 1: A schematic representation of (a) a non-magnetic and (b) a dilute magnetic semiconductor

Theoretical work by Dietl et al showed that Mn doped GaN and ZnO could be ferromagnetic above room temperature, given certain materials assumptions. Ab initio electronic structure calculations by Sato et al have also predicted ferromagnetic ordering in transition metal doped ZnO. These theoretical results propelled additional research into DMS materials, leading to the discovery of numerous transition metal doped semiconductors with ferromagnetism above room temperature. Extensive work on various transition metal doped oxides including TiO_2, ZnO, In_2O_3, SnO_2 has been carried out to search for ferromagnetism, which has been observed at or above room temperature in many of these systems. Re-markably, even undoped In_2O_3 has shown high temperature ferromagnetism together with 55% spin polarization at low temperatures. Whereas these DMSO systems have opened the possibility of developing room temperature spintronic devices, the origin of this ferromagnetism remains highly controversial due the lack of reproducibility and large variations in the measured magnetic properties in these materials. The main puzzle is whether the magnetism in these systems is due to the uniformly distributed transition metal ions in the host matrix or due to the presence of secondary phases, including magnetic impurity phase clusters. For possible technological applications of DMSs, it is important to understand the origin of ferromagnetism in these systems. A number of different mechanism have been propesed to explain the onset of ferromagnetism in DMSs.

6. Multifunctional Multiferroics

The integration of new functionalities in a material generates additional degrees of freedom to interact with various physical properties. For example, the spin of electrons in DMS provides an additional degree of freedom couple to electronic charge. Multiferroics represent another such class of multifunctional systems, which show at least two ferroic orders simultaneously in a single phase material. These ferroic orders are anti/ferromagnetism, anti/ferroelectricity, ferroelasticity, and ferrotoroidicity. The simultaneous co-existence ofboth magnetic and ferroelectric order is often associated with magnetoelectric (ME) coupling, which can cross control of magnetic and electric properties in these materials. Typically, magnetoelectric coupling produces a polarization in an applied magnetic field, as a linear effect, or by applying both electric and magnetic fields simultaneously, as anon-linear effect. This also includes the converse coupling, the induction of a magnetization by an electric field.

The phenomenon of intrinsic ME effect was predicted by P. Curie in 1894 based on crystal symmetry arguments. I. E. Dzyaloshinskii explained the ME effect in antiferromagnetic Cr_2O_3 by explaining the violation of time-reversal symmetry, which was confirmed experimentally by Astrov showing electric field induced magnetization and by Rado et al showing magnetic field induced polarization. After these experiments various linear magneto electric compounds were discovered. A comprehensive list of linear magneto electric compounds was compiled by H. Schmid. These linear magnetoelectric materials, which are generally referred to simply as magnetoelectrics, often show long range magnetic ordering but do not typically develop any spontaneous polarization. Yet an electric polarization can be induced by applying a magnetic

field. In contrast, multiferroics exhibit both spontaneous magnetic order and polarization in a single phase material even in the absence of applied electric or magnetic fields. Linear magnetoelectric materials can be explored by considering magnetic point group symmetry. In a single-phase crystals, the ME effect can be described in Landau theory using the expansion of the total free energy Fin terms of the applied magnetic field H and electric field E, i.e.

$$F(E,H) = F_0 - \frac{1}{2}\epsilon_0\epsilon_{ij}E_iE_j - \frac{1}{2}\chi_0\chi_{ij}H_iH_j - \alpha_{ij}E_iH_j - \frac{1}{2}\beta_{ijk}E_iH_jH_k - \frac{1}{2}\gamma_{ijk}H_jE_iE_k + ..$$

where ϱ0 and χ0 are the free space permittivity and permeability, ϱij and ϱij are the relative permittivity and permeability, αij is the linear magnetoelectric tensor and βijk andγijk are higher order magnetoelectric tensors. The polarization and magnetization can be obtained by differentiating above equation with respect to electric field and magnetic field respectively and are given by:

$$P_i = -\frac{\partial F}{\partial E_i} = \frac{1}{2}\epsilon_0\epsilon_{ij}E_j + \alpha_{ij}H_j + \frac{1}{2}\beta_{ijk}H_jH_k + ..$$

And,

$$M_j = -\frac{\partial F}{\partial H_j} = \frac{1}{2}\chi_0\chi_{ij}E_j + \alpha_{ij}H_j + \frac{1}{2}\beta_{ijk}H_jH_k + ..$$

Thus, a necessary condition for all linear ME material is that they must contain the linear term αijEiHj , where the tensor αij corresponds to the induced polarization or magnetization by an applied magnetics or electric field respectively. This is not the necessary condition for being a multiferroic. The classic example is Cr2O3, which is a linear ME material but not a multiferroic as it is not an intrinsic ferroelectric. Thus all linear ME materials are not multiferroics and vice-versa; all multiferroics are not linear ME materials. For example, YMnO3, which a multiferroic (exhibits antiferromagnetic and ferroelectric ferroic orders simultaneously) but not a ME as symmetry of this compound does not allow linear ME effect. The detailed taxonomy of magnetoelectric and multiferroic materials is given by Eerenstein et al. The schematic relationship between multiferroic and magnetoelectric materials motivated by Eerenstein et al and D. Khomskii is shown in Fig. 2.

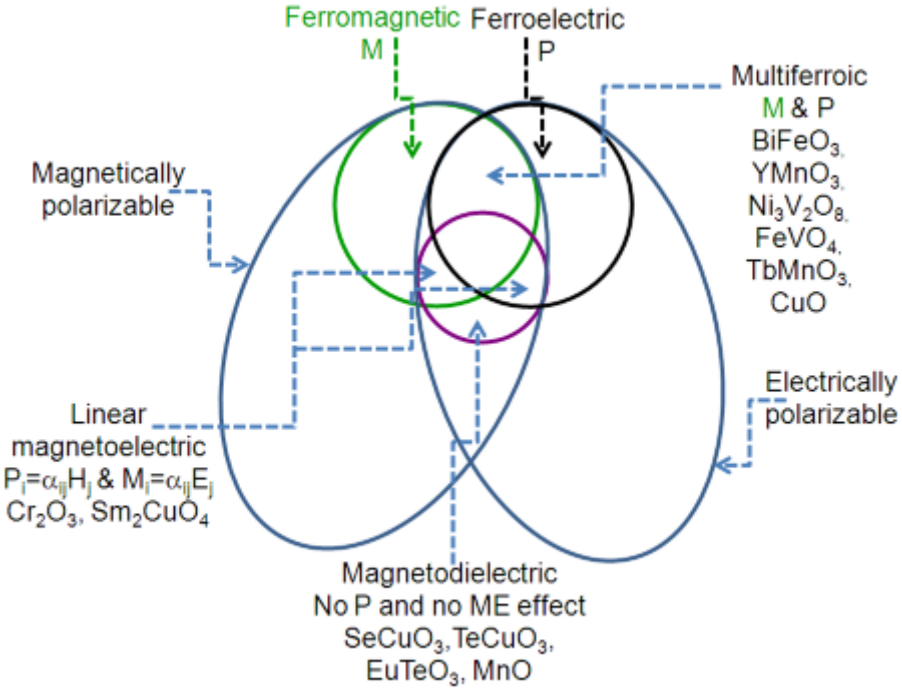

Figure 2: Schematic representation of magnetic, ferroelectric, linear magneto electric, magneto-dielectric, and multiferroic systems

There are materials which are neither multiferroics nor linear ME, including $SeCuO_3$, $TeCuO_3$. These systems show dielectric anomalies at magnetic ordering temperature associated with higher order magneto electric coupling. Lawes et al observed a change in the dielectric constant by applying magnetic field,coupling between dielectric constant and magnetization. This is marked in Fig 2 as a region without spontaneous polarization.

7. Conclusion:

The multifunctional semiconducting and magnetic properties in Cr:InN make this system particularly attractive for devices.Another important multifunctional system studied is

In_2O_3, a semiconducting material that can exhibit additional properties by introducing oxygen vacancies or by using other techniques to modify the structure.The ferromagnetic oxide semiconductors considered in this thesis offer the potential to yield a mechanism for injecting spin polarized electrons into silicon at room temperature, which would be an important step in the long development of viable spintronics devices. The tunable plasmon-mediated infrared absorption in degenerate InN films could, potentially, be used to develop new optoelectronic components, which could be crucial for furthering optical computers. The ability to combine and couple multiple distinct materials properties in a single system offers incredible opportunities to design and engineer materials to develop an almost limitless bounty of new devices.The work conducted for this thesis has been integral in identifying additional multifunctional properties in degenerate InN films, namely the development of magnetism on introduction a small amount of Cr in these samples. We found that adding 2% and 5% Cr to these InN thin films produces a high n-type carrier concentration ∼ 1020cm−3 , attributed to oxygen defects introduced during fabrication, but also to room temperature ferromagnetic order. These systems were confirmed to have a finite spin polarization of the conduction electrons at low temperature using Point Contact Andreev Reflection, establishing their suitability for some spintronic applications. Although the III-V GaMnAs p-type system, where magnetotransport measurements have confirmed carrier-mediated ferromagnetism, has been studied in depth, the critical temperature for this system falls below ∼ 200K, making it unsuitable for room temperature applications.

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