OPTIMIZATION OF GADOLINIUM-DOPED ZINC OXIDE FILMS FOR DILUTED MAGNETIC SEMICONDUCTOR AND ITS EFFECTS ON ORGANIC SOLAR CELL PERFORMANCE

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ABSTRACT

Diluted Magnetic Semiconductors (DMS) are being actively studied as a major step towards the realization of spintronic devices. However, to obtain the DMS with ferromagnetic behaviour at room temperature is the main obstacle in the fabrication of spintronic devices. This obstacle has prompted significant research efforts aimed at exploring new DMS. In this research, Gadolinium (Gd) was doped with Zinc Oxide (ZnO) to explore the feasibility of making new DMS. The Gd-doped ZnO ($Zn_{1-x}Gd_xO$) was synthesized using the sol-gel spin coating technique. Parameters, such as the type of solvent, annealing environment and Gd contents, were varied in order to optimize the structural and magnetic properties. The optimum properties were obtained from a prepared solution using ethanol solvents, annealed in an Ar-environment with 8 % Gd contents. The structural, surface topology, optical, electrical and magnetic properties of the film were studied in detail. Structural studies revealed that the film had a hexagonal wurtzite structure. Surface topologies showed that the film had a smooth and uniform surface. In addition, the film was also highly transparent within visible range with optical transmittance of approximately ~ 90 %. The electrical analysis revealed that the resistivity and carrier concentration was about 3.17×10^{-3} Ωcm and 8.27×10^{14} cm^{-3}, respectively. The magnetic studies showed that the film was ferromagnetic at room temperature with a magnetization of 0.0423 emu/g. The effect of ferromagnetic Gd-doped ZnO film on device performance was demonstrated by fabricating the Spin-Polarized Organic Solar Cell (Spin-OSC). It was found that the ferromagnetic $Zn_{0.92}Gd_{0.08}O$ film contributed immensely to the performance of OSC. The result showed that the efficiency of Spin-OSC (0.16 %) was higher than conventional-OSC (0.04 %). Thus, this proved the possibility of spintronics in optoelectronic applications, especially in solar cells.

ABSTRAK

Semikonduktor Magnet Cair (DMS) sedang dikaji secara aktif sebagai langkah utama ke arah merealisasikan peranti spintronik. Walau bagaimanapun, untuk mendapatkan DMS dengan tingkah laku feromagnet pada suhu bilik adalah halangan utama dalam fabrikasi peranti spintronik tersebut. Halangan ini telah mendorong usaha penyelidikan yang signifikan untuk meneroka DMS baru. Dalam kajian ini, Gadolinium (Gd) telah didopkan dengan Zink Oksida (ZnO) untuk meneroka kebolehlaksanaan membuat DMS baru. ZnO didop Gd ($Zn_{1-x}Gd_xO$) telah disintesis menggunakan teknik sol-gel salutan putaran. Parameter seperti jenis pelarut, persekitaran penyepuhlindapan dan kandungan Gd diubah untuk mengoptimumkan sifat-sifat struktur dan magnetik. Sifat-sifat optimum diperoleh daripada larutan yang disediakan menggunakan pelarut etanol, disepuh lindap dalam persekitaran Ar dengan 8% kandungan Gd. Sifat-sifar struktur, permukaan topologi, optik, elektrik dan magnetik filem telah dikaji secara terperinci. Kajian struktur menunjukkan bahawa filem itu mempunyai struktur wurtzit heksagonal. Permukaan topologi menunjukkan bahawa filem itu mempunyai permukaan licin dan seragam. Di samping itu, filem tersebut juga lutsinar tinggi dalam julat nampak dengan kehantaran optik kira-kira ~ 90%. Analisis elektrik mendedahkan bahawa kerintangan dan kepekatan pembawa adalah kira-kira 3.17 x 10^{-3} cm dan 8.27 x 10^{14} cm^{-3}, masing-masing. Kajian magnet menunjukkan bahawa filem itu adalah feromagnet pada suhu bilik dengan kemagnetan 0.0423 emu / g. Kesan filem ferromagnet $Zn_{1-x}Gd_xO$ keatas prestasi peranti telah ditunjukkan dengan menfabrikasi Spin- terkutub Sel Suria Organik (Spin-OSC). Didapati bahawa filem ferromagnet $Zn_{0.92}Gd_{0.08}O$ sangat menyumbang kepada prestasi OSC. Keputusan menunjukkan bahawa kecekapan Spin-OSC (0.16 %) adalah lebih tinggi daripada konvensional-OSC (0.04 %). Oleh itu, ini membuktikan kemungkinan spintronik dalam applikasi optoelektronik terutamanya dalam sel solar.

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LIST OF SYMBOLS AND ABBREVIATIONS

α	-	Absorption coefficient
β	-	Full width at half maximum
δ	-	Dislocation density
η	-	Efficiency
θ	-	Angle between incidents light
λ	-	Wavelength
μ_B	-	Bohr magneton
χ	-	Magnetic susceptibility
$\hbar$	-	Plank constant
$-1 \leq x \leq 1$	-	x is greater than or equal to -1 and x is less than or equal to 1
d_{hkl}	-	Spacing between the planes
e	-	Electron charge
hv	-	Absorbed photon
m	-	Magnetic moment
m_e	-	Mass of electron
n	-	Integer 1, 2, 3……
$n_\uparrow$	-	Number of spin up.
$n_\downarrow$	-	Number of spin down
t	-	Thickness
A	-	Energy-independent constant
Ar	-	Argon
AFM	-	Atomic force microscope
BMP	-	Bound magnetic polaron
C	-	Material specific Curie constant
CoO	-	Cobalt oxide

CB	-	Conduction band
Cr	-	Chromium
D	-	Crystallite size
Dy	-	Dysprosium
DMS	-	Diluted magnetic semiconductor
Er	-	Erbium
Eu	-	Europium
E_g	-	Energy band gap
EDX	-	Energy dispersive x-ray
FF	-	Fill factor
Gd	-	Gadolinium
GaN	-	Gallium nitrate
GMR	-	Giant magneto-resistance
GaAs	-	Gallium arsenide
GaMnAs	-	Gallium-manganese-arsenide
H	-	Applied magnetic field.
HOMO	-	Highest occupied molecular orbital
I	-	Transmitted light intensity
I_o	-	Original light intensity
ITO	-	Indium tin oxide
InAs	-	Indium arsenide
InMnAs	-	Indium-manganese-arsenide
J_{SC}	-	Short circuit current
J_{mpp}	-	Current at the maximum power point.
LUMO	-	Lowest occupied molecular orbital
M	-	Magnetization
Mn	-	Manganese
MBE	-	Molecular beam epitaxy
MFM	-	Magnetic force microscope
MTJ	-	Magnetic tunnel junction
MRAM	-	Magnetic random access memory
Ni	-	Nickel
NiO	-	Nickle oxide

O	-	Oxygen
OSC	-	Organic solar cell
P	-	Polarization
P3HT	-	Poly-3-hexylthiophene
PCBM	-	[6, 6] - phenyl-C61-butyric acid methyl ester
RE	-	Rare earth
RMS	-	Root mean square
SQUID	-	Super-conducting quantum interference device
T	-	Absolute temperature
T	-	Transmittance
Tc	-	Curie temperature
T_N	-	Neel temperature
Tb	-	Terbium
TM	-	Transition Metal
TMR	-	Tunnel magneto-resistance
UV	-	Ultra violet
UV- Vis	-	Ultra violet visible
VB	-	Valence band
V_H	-	Hall voltage
VSM	-	Vibrating sample magnetometer
XRD	-	X-ray diffractometer
Zn	-	Zinc
ZnO	-	Zinc oxide

LIST OF PUBLICATIONS

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(i) Sahdan, M.Z., Malek, M.F., Alias, M.S., Kamaruddin, S.A., **Norhidayah, C.A.**, Sarip, N., Nafarizal, N. and Rusop, M.(2016). Metamorphosis of the ZnO buffer layer thicknesses on the performance of inverted organic solar cells. Journal of Materials Science: Materials in Electronics, pp.1-12.

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Proceeding Paper:

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CHAPTER 1

INTRODUCTION

1.1 Overview on Electronics

Today, almost all technological application such as data processing storage, integrated circuits, high-frequency devices and etc. are comprised of electronic devices (e.g. diode, transistor, etc.,). The electronic devices are very important in our daily. Our kitchens from water coolers to the microwave ovens are equipped with electronic equipment. Even the doctors at the hospital rely on electronics in order to diagnose and treat the various diseases. It began with the invention of the vacuum tubes [1]. The vacuum tubes are also known as electron tube device widely used to amplify the electronic signals. It consists of two or more electrodes inside a glass tube. The invention of the vacuum tube has played an important role in our modern electronic technology.

Until the advent of the transistor in the 1950s, the vacuum tubes have an enormous number of applications. They have been widely used in radio receiver as well as in early digital computers. Unfortunately, the vacuum tubes have many drawbacks. It is bulky, therefore; it is less suitable for portable products. It operates generally at a higher voltage and consumes high power. Furthermore, the glass tubes are fragile, limited life and low reliability therefore not suitable for many industrial applications [2].

The invention of the transistor in the 20th century is an alternative to the vacuum tube [1]. In contrast to the vacuum tube, the transistors are smaller and more compact; e.g., one transistor can replace the equivalent of 40 vacuum tubes. Furthermore, the transistors are usually made of solid material, such as silicon; therefore, it is inexpensive, able to conduct electricity faster and give off virtually no heat compared to vacuum tubes [3]. After the invention of the transistor, the microelectronics industries have achieved tremendous growth in integrated circuit chips that have some application today. The integrated circuit is a pack containing a large number of transistors in a single chip that able to increase the efficiency of the computer in addition to reduce the costs [4].

Along with the rapid development of microelectronic technology, Moore's law is running out of momentum for better device performance. According to Moore's law, the numbers of the transistor in an integrated circuit will double approximately every two years [5]. In general, the process of the miniaturization is good for the transistor and other electronic devices because it can enhance the efficiency of the devices while reducing cost. However, the continued miniaturization of the devices will face with the problems related to the electric current leakage, power consumption and heat dissipation [6, 7]. Therefore, an alternative solution must be taken to face this problem.

Most of the electronic devices are based on the principle to generate or control the electric current by exploiting the charge of electrons, i.e. by steering the motion of the charge through their interaction with external electrical or electromagnetic fields. In fact, the electron actually has another property i.e. spin. The movement of the Earth circulating the Sun in the solar system (Figure 1.1) can represent the analogy of the spin. As the Earth circulates around the Sun, it also spins on its axis. In a similar fashion, the electron circulating around the nucleus and at the same time it also spins around its axis.

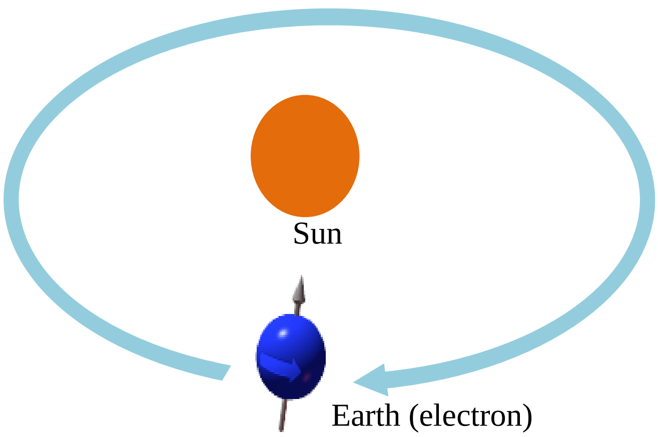

Figure 1.1: Schematic representing the analogy of the electron spins circulating the nucleus (the sun is representative the nucleus while the earth representing the electron**)**

Indeed, the spins are the root of magnetism and it cannot be gained or lost. It also can exist in one of two states i.e. “up” or “down,” rather than “on” “or” off. The spins can be controlled electrically with the application of few volts. Furthermore, the spin is a quantum nature, so it can exist in infinitely many intermediate states depending on the energy of the system. Hence, the researcher and physicists believe that these advantages offer the possibility to develop a new generation of devices that based on the principle of the electron spin or also called spintronics.

1.2 Introduction to Spintronics

Spin-based electronics or spintronics is a term used to describe the new field that manipulates the spin of the electron. The spintronic become an important subject of the science from both its fundamental and application because it has a strong potential to bring novel and promising concepts.

Spintronics emerged from the discovery of Giant Magnetoresistance (GMR) effect by Albert Fert and Peter Grünberg [8]. Applications of this GMR have revolutionized techniques for retrieving data from hard disks. The GMR’s application contributed to the fast rise in the density of stored information. For instance, since the discovery of GMR in around 1997, storage capacities have increased approximately 100 times [9]–[11]. However, so far most of the successfully commercialized devices are made of magnetic materials. Because the spin in magnetic material is easy to control since they are polarized. In a semiconductor, the spin is difficult to manipulate because the direction of spin is randomly oriented since the material is non-magnetic.

Since a few years ago, the search for materials combining both the semiconducting and ferromagnetic properties has evolved into an important field of materials science [12]. The combinations of these properties are expected to realize a new generation of spintronic device. In conjunction with this, Diluted Magnetic Semiconductor (DMS) that can be formed by doping magnetic ions into semiconductor has become a major focus. In DMS both the charge and spin of electrons can be used since it has both semiconducting and magnetic properties. The discovery of DMS opens possibility to develop semiconductor-based spintronic devices such as spin-solar cell, spin-transistors, spins Light emitting diodes, magnetic sensors, non-volatile memory, logic devices, optical isolators and ultra-fast optical switches.

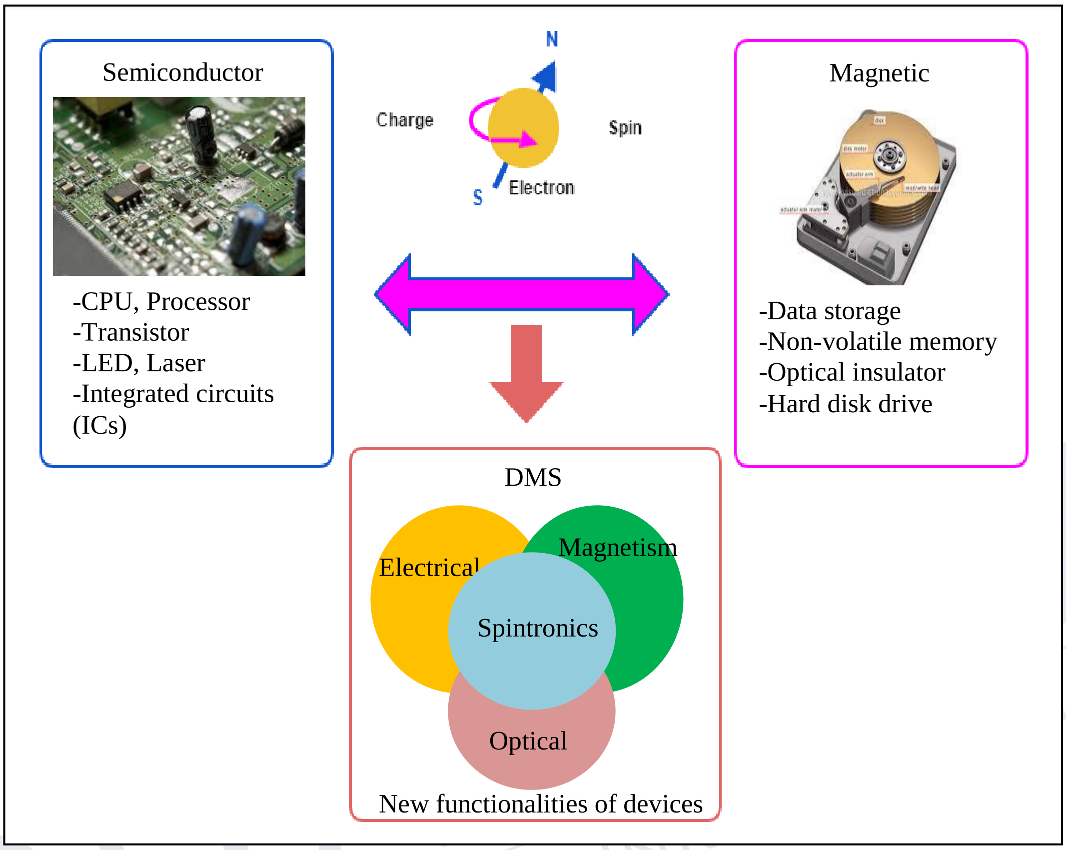

Figure 1.2: Concept chart of semiconductor-based spintronics

1.3 Background of Study

Ferromagnetism and semiconducting behaviour indeed coexist in a magnetic semiconductors such as europium (Eu) chalcogenides (e.g., EuSe, EuS, EuO) and semiconducting spinels (e.g., $CdCr_2S_4$, $CdCr_2Se_4$). The materials were extensively studied from the late 1960s to early 1970s [13]–[15]. However, the Curie temperature (Tc) is lower than room temperatures, which make the ferromagnetic not retain at room temperature [16]–[18].

DMS is an alternative to the magnetic semiconductor. The first discovery DMS is transition metal (TM) doped II-VI compound semiconductors, typical examples are ZnSe, CdSe and CdTe [19], [20]. However, these materials exhibited weak ferromagnetic behaviour with *Tc* only a few Kelvin (K).

Later on, Munekata *et al.* successfully synthesize Mn-doped indium arsenide (InAs) using molecular beam epitaxy (MBE) [21]. The films that have been grown at 200 ^{o}C showed homogeneous which exhibit paramagnetic behaviour, while the films grown at 300 ^{o}C was showed ferromagnetic behaviour. However, the behaviour of ferromagnetism is due to the presence of MnAs nanoclusters. If the temperature is low, the atoms of the magnetic element have not enough thermic energy to diffuse up to the surface and form the so-called second phase (the nanoparticles). Seven years later, Ohno *et al.* [22] have successfully grown the first intrinsic ferromagnetic gallium-manganese-arsenide (GaMnAs) DMS thin films using low-temperature MBE (LT-MBE) and proved the ferromagnetism by super-conducting quantum interference device (SQUID) measurements. Nevertheless, the highest reported *Tc* achieved in this system is about ~185 K. Although the *Tc* of these compounds has greatly improved compared to previous group compounds, it is still far below room temperature [23].

In year 2000s, Dielt *et al*. simulated *Tc* for various host semiconductor doped with 5 % Mn using Zener model (see Figure 1.4) [24]. Interestingly, among the host semiconductors that simulated, wide band gap semiconductor, zinc oxide (ZnO) and gallium nitrate (GaN) were predicted as a potential host material to exhibit *Tc* higher than room temperature. Furthermore, both materials have smaller lattice constants (Figure 1.4), large p–d hybridization, and small spin-orbit interaction, resulting in a large spin coherence length which is a prerequisite to exhibit higher *Tc* [25], [26]. Although the GaN was predicted as one of a promising candidate as a host semiconductor, however, the development of GaN require complex processing steps and difficult to synthesize without vacuum based heavy instrumentation [27]. Hence, to obtain ferromagnetic films with *Tc* higher than room temperature are challenging. On the other hand, the ability of the ZnO to synthesize high quality films at the lower processing temperature and simple deposition technique without vacuum make it suitable as a host semiconductor. This is particularly important for the design and fabrication of devices.

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