SYNTHESIS AND CHARACTERIZATION OF ZINC OXIDE CO-DOPED WITH GADOLINIUM AND ALUMINIUM USING CO-SPUTTERING TECHNIQUE FOR P-N HETEROJUNCTION DIODE APPLICATION

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ELECTRICAL AND ELECTRONIC ENGINEERING

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ABSTRACT

Diluted magnetic semiconductors (DMS) are being extensively researched as a significant step toward the development of spintronic devices. The discovery of appropriate materials that exhibit ferromagnetic behavior at room temperature and high magnetism is critical for the realization of such devices. However, generating ferromagnetism in ZnO based DMS remains a major obstacle to the fabrication of spintronic devices operating above room temperature and the understanding on the origin of its ferromagnetism is still lacking. In this study, shallow donors of Aluminium (Al) was incorporated into Gadolinium (Gd) doped Zinc Oxide (ZnO) films to explore the possibility of developing a new DMS through co-sputtering technique deposited at room temperature. This study investigated the effect of sputtering parameters and the effect of dopant amount (Gd and Al) on the film characteristics. The findings reveal that 3 at% of (Gd, Al) co-doped ZnO exhibited good physical properties with enhanced magnetic behavior at room temperature as compared to the Gd-doped ZnO and undoped ZnO. X-ray diffraction (XRD) study confirmed the films are well crystalline indexed to the hexagonal wurtzite structure of ZnO with no secondary phases and further supported by energy-dispersive spectroscopy (EDS) analysis study that indicating the existence of Zn, O, Al and Gd elements in the prepared film. Homogeneous nanostructure with well-aligned structure as well as small grains observed field-emission scanning electron microscope (FESEM) and atomic force microscopy (AFM) correlates with the magnetic properties, which contributes to the improvement in saturation magnetization (M_s) and high coercivity (H_c). The optical transmittance obtained above 90% in the visible region with band gap was found red-shifted by Al co-doping. The incorporation of Al into Gd-doped ZnO demonstrated a free electron carrier concentration dependence, which increases considerably when the carrier concentration surpasses ~$5.3 x 10^{26}$ m^{-3}. The magnetic force microscopy (MFM) measurement proved the existence of room temperature ferromagnetism and spin polarization in 3 at% (Gd, Al) co-doped ZnO film as it exhibited smaller domain size with shorter magnetic correlation length *L*, larger phase shift Φ_{rms} and highest value of δf_{rms}. These findings were further supported by the room temperature M-H curves of the 3 at% (Gd, Al) co-doped ZnO film with improvement of M_s and H_c by 37.9 % and 60.7 %, respectively from 3 at% Gd-doped ZnO film, which the film were induced by carrier-mediated ferromagnetism. Potential n-ZnO based DMS/p-Si heterojunction diode was also demonstrated with the use of (Gd, Al) co-doped ZnO film indicating lowest leakage current of 1.28×10^{-8} A and the ideality factor, n of 1.11 almost to ideal diode behavior of n=1 as compared to the p-Si/n-Gd-doped ZnO and p-Si/n-undoped ZnO heterojunction diodes. The obtained results conclude that (Gd, Al) co-doped ZnO films synthesized by co-sputtering technique have improved electrical and magnetic properties where the films indicate room temperature ferromagnetism with the origin of its magnetism were induced by carrier-mediated ferromagnetism, thus proving that this type of DMS is a promising material for potential spin-based electronic application.

ABSTRAK

Semikonduktor magnetik cair (DMS) sedang dikaji secara meluas sebagai langkah penting ke arah pembangunan peranti spintronik. Penemuan bahan yang mempamerkan tingkah laku feromagnetik pada suhu bilik dan kemagnetan yang tinggi adalah penting untuk merealisasikan peranti tersebut. Walau bagaimanapun, penjanaan feromagnetik dalam DMS berasaskan Zink Oksida (ZnO) kekal sebagai penghalang utama kepada fabrikasi peranti spintronik yang beroperasi di atas suhu bilik dan sifat feromagnetik masih kurang dikaji. Dalam kajian ini, penderma cetek Aluminium (Al) telah dimasukkan ke dalam filem ZnO berdop Gadolinium (Gd) untuk meneroka DMS baharu. Filem-filem tersebut dihasilkan dengan menggunakan kaedah percikan bersama yang dijalankan pada suhu bilik. Penyelidikan ini meneroka kesan daripada percikan parameter dan kesan jumlah dopan (Gd dan Al). Penemuan mendedahkan bahawa 3 at% daripada (Gd, Al) didop bersama ZnO menunjukkan peningkatan dalam sifat dan mempamerkan tingkah laku magnet pada suhu bilik. Kajian pembelauan sinar-X (XRD) mengesahkan filem itu diindekas kepada struktur wurzite heksagon ZnO tanpa fasa sekunder dan disokong oleh kajian analisis spektroskopi penyebaran tenaga (EDS) menunjukkan kewujudan unsur Zn, O, Al dan Gd dalam filem yang disediakan. Struktur nano yang sejajar serta butiran kecil daripada mikroskop elektron pengimbasan pancaran medan (FESEM) dan mikroskopi daya atom (AFM) berkorelasi dengan sifat magnetik, yang menyumbang kepada peningkatan dalam kemagnetan tepu (M_s) dan paksaan (H_c). Transmisi optik yang diperoleh melebihi 90% di kawasan UV oleh doping bersama Al.

Penggabungan Al ke dalam ZnO berdop Gd menunjukkan pergantungan kepekatan pembawa elektron bebas, yang meningkat dengan ketara apabila kepekatan pembawa melebihi ~$5.3x10^{26}$ m^{-3}. Pengukuran mikroskop daya magnet (MFM) membuktikan kewujudan feromagnetik pada suhu bilik dan polarisasi putaran dalam filem ZnO yang didop bersama 3 at% (Gd, Al) kerana ia mempamerkan saiz domain yang lebih kecil dengan panjang korelasi magnet yang lebih pendek *L*, anjakan fasa Φ_{rms} yang lebih besar dan nilai tertinggi bagi δf_{rms}. Penemuan ini disokong lagi oleh lengkung M-H suhu bilik filem ZnO yang didop bersama 3 at% (Gd, Al) dengan peningkatan M_s dan nilai H_c iaitu 37.9 %, dan 60.7%, masing-masing daripada filen ZnO yang didop pada 3 at% Gd, yang mana filem itu didorong oleh pembawa feromagnetik pengantara. Kesan filem ZnO terdop feromagnetik (Gd, Al) telah ditunjukkan pada prestasi diod heterojunction ZnO/Si. Keputusan menunjukkan arus bocor yang rendah iaitu 1.28 x 10^{-8} A dan faktor idealiti, *n* sebanyak 1.11 hampir kepada kelakuan diod ideal *n*=1. Keputusan yang diperolehi menyimpulkan bahawa filem ZnO yang didopkan bersama (Gd, Al) dihasilkan dengan menggunakan kaedah percikan bersama telah bertambah baik dari segi sifat elektrik dan sifat magnet di mana filem itu menunjukkan ferromagnetik pada suhu bilik dengan asal kemagnetannya didorong oleh pembawa feromagnetik pengantara, oleh itu membuktikan bahawa filem jenis DMS ini berpotensi untuk aplikasi spintronik.

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APPROVAL

The Examination Committee has met on **12 April 2023** to conduct the final examination of **Nur Amaliyana Binti Raship** on his degree thesis entitled **Synthesis and characterization of ZnO co-doped with Gd and Al using co-sputtering technique for p-n heterojunction diode application.**

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Academic session : 2019/2020

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AFM	-	Atomic force microscopy
BMP	-	Bound magnetic polaron
CBD	-	Chemical bath deposition
CVD	-	Chemical vapor deposition
DMS	-	Diluted magnetic semiconductor
DC	-	Direct current
EDS	-	Energy dispersive X-ray spectroscopy
FESEM	-	Field emission scanning electron microscope
FWHM	-	Full width at half maximum
GMR	-	Giant magneto resistance
hcp	-	Hexagonal close packed
HIPPIMS	-	High power impulse magnetron sputtering
ICSD	-	Inorganic crystal structure database
LED	-	Light emitting diode
MBE	-	Molecular beam epitaxy
MFM	-	Magnetic force microscopy
PL	-	Photoluminescence
PLD	-	Pulse laser deposition
PVD	-	Physical vapour deposition
RF	-	Radio frequency
RE	-	Rare-earth
RKYY	-	Ruderman, Kittel, Kasuya and Yoshida
RT	-	Room temperature
TM	-	Transition Metal
TMP	-	Turbo molecular pump
SQUID	-	Super-conducting quantum interference device
UV-Vis	-	Ultraviolet-visible spectroscopy
VSM	-	Vibrating sample magnetometer
W-H	-	Williamson hall
XAS	-	X-ray absorption spectroscopy
XRD	-	X-ray diffraction

LIST OF SYMBOLS

Al	-	Aluminum
Ar	-	Argon
Å	-	Armstrong
At%	-	Atomic percentage
f_o	-	Average MFM signal
μ_B	-	Bohr magneton
Φ_B	-	Barrier height
k	-	Boltzman constant
CdSe	-	Cadmium selenide
CdTe	-	Cadmium telluride
Ce	-	Cerium
q	-	Charge
$CdCr_2S_4$	-	Chromium spinels
Co	-	Cobalt
H_c	-	Coercivity
J	-	Coulombic
D	-	Crystallite size
C	-	Curie constant
T_c	-	Curie temperature
I	-	Current
^oC	-	Degree Celsius
τ	-	Distance of electron travels around a nucleus
Dy	-	Dysprosium
A	-	Effective contact area
A*	-	Effective Richardson constant
e	-	Electron charge
m_e	-	Electron mass
eV	-	Energy
E_g	-	Energy band gap
Eu	-	Europium

Er	-	Erbium
EuO	-	Europium chalcogenides
GaAs	-	Gallium arsenide
GaMnAs	-	Gallium manganese arsenide
GaN	-	Gallium nitrate
Gd	-	Gadolinium
Ge	-	Germanium
Ho	-	Holmium
n	-	Ideality factor
M_i	-	Incident atom mass
In	-	Indium
InAs	-	Indium arsenide
Fe	-	Iron
K	-	Kelvin
La	-	Lanthanum
Lu	-	Lutetium
M	-	Magnetization
L	-	Magnetic correlation length
H	-	Magnetic field
m	-	Magnetic moment
χ	-	Magnetic susceptibility
Mn	-	Manganese
MnAs	-	Manganese arsenide
Nm	-	Nanometer
T_N	-	Néel temperature
Nd	-	Neodymium
Ni	-	Nickel
N_2	-	Nitrogen
V_o	-	Oxygen vacancies
Pr	-	Praseodymium
Pm	-	Promethium
%	-	Percentage
μ_o	-	Permeability

f_i	-	Pixel of MFM signal
p	-	Pressure
h	-	Planck constant
M_r	-	Remanent magnetization
Sm	-	Samarium
I_o	-	Saturation current
M_s	-	Saturation magnetization
Si	-	Silicon
Ag	-	Silver
M_t	-	Target atom mass
T	-	Temperature
Tb	-	Terbium
Sn	-	Tin
N	-	Total pixels number of MFM image
Tm	-	Thulium
V	-	Vanadium
V	-	Velocity
v	-	Volume
λ	-	Wavelength
Yb	-	Ytterbium
Zn_i	-	Zinc interstitials
ZnSe	-	Zinc selenide
ZnO	-	Zinc oxide
V_{Zn}	-	Zinc vacancies