ZnO and MgZnO Nanostructures and Heterostructures

by

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Certificate of Original Authorship

I, Muhammad Zakria declare that this thesis with title "ZnO and MgZnO Nanostructures and Heterostructures" is submitted in the fulfilment of the requirement for the award of Doctor of Philosophy (PhD), in the School of Mathematical and Physical Sciences, Faculty of Science, at the University of Technology Sydney.

This thesis is wholly my own work unless otherwise reference or acknowledged. In addition, I certify that all information sources and literature used are indicated in the thesis. This document has not been submitted for qualifications at any other academic institution.

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Publications and Presentation

1. **M. Zakria**, T. T. Huynh, F. C. C. Ling, S. C. Su, M. R. Phillips, C. Ton-That. "Highly Luminescent MgZnO/ZnO Multiple Quantum Wells for Photonics Devices" ACS Applied Nano Materials, (2019), 2, 2574 – 2579.
2. **M. Zakria,** P. Bove, D. J. Rogers, F. H. Teherani, E. V. Sandana, M. R. Phillips, and C. Ton-That. "Chemical Structure and Optical Signatures of Nitrogen Acceptors in MgZnO" J. Mater. Chem. C, (2020),8, 6435-6441.
3. **M. Zakria**, Matthew Phillips, Cuong Ton-That. "Luminescence Enhancement of MgZnO/ZnO Multiple Quantum Wells by Hydrogen Plasma Treatment" ICONN 2020.
4. T. T. Huynh, Chikoidze, E., Curtis P. Irvine, **Muhammad Zakria**, Dumont, Y., Teherani, F.H., Sandana, V.E., Bove, P., Rogers, D.J., MR Phillips, C Ton-That "Red Luminescence in H-doped β-Ga2O3" Phys. Rev. Materials 4, 085201 (2020).

Table of Contents

List of Figures

List of Tables

List of Abbreviations

AEs	Auger electrons
AFM	Atomic force microscopy
A^{o}X	Neutral acceptor bound exciton
BSE	Back scattered electrons
BSF	Basal stacking fault
CCD	Charge-coupled device
CL	Cathodoluminescence
DAP	Donor–acceptor pair
D^{o}X	Neutral donor-bound exciton
DX	Donor-bound exciton
EDX	Energy-dispersive X-ray
FX	Free exciton
FWHM	Full width at half maximum
GL	Green luminescence
h-BN	Hexagonal boron nitride
HEMT	High electron mobility transistor
LED	Light-emitting diode
LO	Longitudinal optical
MBE	Molecular beam epitaxy
MQW	Multiple quantum well
NBE	Near band edge
NEXAFS	Near-edge X-ray absorption fine structure
NIST	National Institute of Standard and Technology
PL	Photoluminescence
PLD	Pulsed laser deposition
RRPA	Rapid remote plasma annealing
RMS	Root mean square

RRS	Resonant Raman scattering
SE	Secondary electron
SEM	Scanning electron microscope
TEM	Transmission electron microscope
TEY	Total electron yield
TFY	Total fluorescence yield
TMD	Transition metal dichalcogenide
UV	Ultraviolet
VBM	Valence band maximum
XRD	X-ray diffraction

Abstract

ZnO-based heterostructures and nanostructures have attracted significant interest owing to their wide range of technological applications. The recent achievement of high electron mobility at the MgZnO/ZnO heterointerface has sparked great interest in a multitude of research fields. In order to exploit the extraordinary electron states at the MgZnO/ZnO interface, high quality films with bespoke optical and electronic properties must be achieved. Although the ZnO-based heterostructures have been widely explored for various applications, where the performance is often hindered by intrinsic and extrinsic defects. This work aims to elucidate the physics of defects and the properties of ZnO-based thin films, interfaces and 2D nanosheets.

Oxide-based multiple quantum wells (MQWs) were investigated using cathodoluminescence (CL) and high-resolution electron microscopy techniques. A rapid remote plasma annealing (RRPA) method was used to treat MgZnO/ZnO MQWs in order to modify their defect structure. Following the RRPA in hydrogen, the MQW optical emission increased by more than 10 times after a 40 seconds treatment, while the basal stacking faults (BSFs) and point defects emissions were completely quenched. Furthermore, the RRPA-treated MQWs were found to be highly stable up to a temperature of 400°C.

A major challenge in the development of ZnO-based devices is the lack of reliable p-type material. In this work, chemical and optical signatures of nitrogen in N-doped MgZnO were investigated using near-edge X-ray absorption fine structure (NEXAFS) and photoluminescence (PL). The MgZnO epilayer, grown under nitrogen ambient, exhibits higher resistivity compared with epilayers grown under oxygen or vacuum

atmospheres, and displays a dominant donor-acceptor-pair (DAP) peak located at 160 meV below its exciton emission. NEXAFS reveals that nitrogen in the N-doped MgZnO exists in multiple chemical states with molecular N_2 and substitutional N on O sites (N_O) being the dominant species. The PL emission peak at 3.45 eV in the N-doped MgZnO is attributed to a shallow donor to a deep acceptor recombination, where the compensating acceptor is most likely molecular N_2.

The last part of this thesis reports the luminescence and morphological properties of ZnO 2D nanosheets, fabricated by chemical exfoliation of ZnO microparticles. High-spatial-resolution CL was employed to acquire the optical properties of individual nanosheets. Combined CL and PL analysis shows strong thickness-dependent quantum confinement of excitons in few-atomic-layer thin nanosheets, which leads to substantial variations in the excitonic and phonon coupling properties. The superior excitonic properties of ZnO nanosheets could potentially lead to the development of efficient nano-optoelectronic devices.