Zinc oxide nanophotonics - toward quantum photonic technologies

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Declaration of Authorship

I certify that the work in this thesis has not previously been submitted for a degree nor has it been submitted as part of requirements for a degree except as fully acknowledged within the text.

I also certify the thesis has been written by me. Any help that I have received in my research work and the preparation of the thesis itself has been acknowledged. In addition, I certify that all information sources and literature used are indicated in the thesis.

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Date: 13/10/2016

Abstract

Zinc oxide (ZnO) is a large bandgap (3.37 eV at room temperature) semiconductor and is a good candidate for short wavelength photonic devices such as laser diodes. A large exciton binding energy (60 meV) at room temperature in addition to the advantages of being able to grow various nanostructure forms have made ZnO suitable for a wide range of applications in optoelectronic devices.

Driven by the rapid advance of nanophotonics, it is necessary to develop single photon sources (SPSs) and optical resonators in new class of materials. In particular, SPSs are required for a wide range of applications in quantum information science, quantum cryptography, and quantum communications. ZnO has been investigated for classical light emitting applications such as energy efficient light emitting diodes (LEDs) and ultraviolet (UV) lasers. Significantly ZnO has recently been identified as a promising candidate for quantum photonic technologies. Thus in this thesis the optical properties of ZnO micro- and nano-structures were investigated for ZnO nanophotonic technologies, specifically their applications in single photon emission and optical resonators.

Firstly, the formation of radiative point defects in ZnO nanoparticles and their photophysical properties were investigated. In particular, using correlative photoluminescence (PL), cathodoluminescence (CL), electron paramagnetic resonance (EPR), and x-ray absorption near edge spectroscopy (XANES) it is shown that green luminescence (GL) at 2.48 eV and an EPR line at $g = 2.00$ belong to a surface oxygen vacancy ($V_{O,s}^{+}$) center, while a second green emission at 2.28 eV is associated with zinc vacancy (V_{Zn}) centers. It is established that these point defects exhibit nanosecond lifetimes when excited by above bandgap or sub-bandgap (405 nm and 532 nm excitation wavelength) excitation. These results demonstrate that point defects in ZnO nanostructures can be engineered for nanophotonic technologies.

ZnO nanoparticles were consequently studied for the investigation of room temperature single photon emission from defect centers in ZnO nanoparticles. Under

the optical excitation with 532 nm green laser, the emitters exhibit bright broadband fluorescence in the red spectral range centered at 640 nm. The red fluorescence from SPSs in ZnO defect center is almost fully linearly polarized with high signal-to-noise ratio. The studied emitters showed continuous blinking; however, it was confirmed that bleaching can be suppressed using a polymethyl methacrylate (PMMA) coating. Furthermore, passivation by hydrogen treatment increase the density of single photon emitters by a factor of three.

ZnO/Si heterojunctions were fabricated and used to investigate electrically driven light emission from localized defects in ZnO nanostructures at room temperature. It is shown that excellent rectifying behaviors were observed with the threshold voltages at $\sim$ 18 V and $\sim$ 7 V for ZnO nanoparticles and thin film-based devices, respectively. Both devices exhibit electroluminescence (EL) in the red spectral region ranging from $\sim$ 500 nm to 800 nm when 40 V and 15 V were applied to ZnO nanoparticles/Si and ZnO thin film/Si, respectively. The emission is bright and stable for more than 30 minutes, providing an important prerequisite for practical devices.

Finally, ZnO optical resonators were fabricated and investigated to enhance the visible light emission. Hexagonal ZnO microdisks with diameter ranging from 3 μm to 15 μm were grown by a carbothermal reduction method. Optical characterization of ZnO microdisks was performed using low temperature (80 K) CL imaging and spectroscopy. The green emission is found to be locally distributed near the hexagonal boundary of the ZnO microdisks. High resolution CL spectra of the ZnO microdisks reveal whispering gallery modes (WGMs) emission. Two different sizes (5 μm and 9 μm) of the ZnO microdisks were simulated to analyze the nature of light confinement in terms of geometrical optics. Respective analysis of the mode spacing and the mode resonances are used to show that the ZnO microdisks support the propagation of WGMs. The results show that the experimentally observed WGMs are in excellent agreement with the predicted theoretical positions calculated using a plane wave model. This work could provide the means for ZnO microdisk devices operating in the green spectral range.

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Abbreviations

APD – avalanche photo diode

ASE – amplified spontaneous emission

CB – conduction band

CCD – charge coupled device

CL – cathodoluminescence

CW – continuous wavelength

DAP – donor-acceptor pair

DFT – density functional theory

DL – deep-level

EL – electroluminescence

EPR – electron paramagnetic resonance

ESR – electron spin resonance

FPMs – Fabry-Pérot modes

FWHM – full width at half maximum

GL – green luminescence

HBT – Hanbury Brown and Twiss

LDA – local density approximation

LED – light emitting diode

LPB – lower polariton branch

NBE – near-band edge

PL – photoluminescence

PMMA – polymethyl methacrylate

RL – red luminescence

SEM – scanning electron microscope

SPSs – single photon sources

sccm – standard cubic centimeters per minute

TEM – transmission electron microscopy

TCSPC – time correlated single photon counting

UPB – upper polariton branch

UV – ultraviolet

VB – valence band

W-H – Williamson-Hall

WGMs – whispering gallery modes

XANES – x-ray absorption near edge spectroscopy

XRD – x-ray diffraction

YL – yellow luminescence

ZPL – zero phonon line