A SIMULATION STUDY OF PHOTOLUMINESCENCE SILICON NANOWIRES

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This dissertation is dedicated to my parents and wife for their endless support and encouragement

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ABSTRACT

The need for the integration of silicon (Si) nanostructures (NS) in optical devices has led to the search for silicon based materials and structures that emit lights with high quantum efficiency. Since after the observation of room temperature visible photoluminescence (PL) from silicon nanostructures (Quantum dot or wire) and the feasibility of turning the optical response of Si NS by modifying their size, many experimental and theoretical works have been done to explain the origin of the visible photoluminescence mechanism. However, the mechanism of visible luminescence remains unclear despite the efforts. Lately, Si nanostructure with hydrogen and oxygen passivated surface become attractive due to enhanced light emission. the purpose of this research is to develop a hybrid model of quantum confinement effect (QCE), surface state effects (SSE) and exciton energy state (EES) to study PL spectra of Si nanowire using data obtained from experimental and simulation (pseudo-potential approximation and tight binding method) findings. A simple, linear simulation method (matlab codes) is used to examine the optical responses of Si NWs with diameter between 1.5 and 5.8nm. An integrated photoluminescence intensity model is adopted to generate PL spectra. The results of this work indicate that both QCE and surface passivation together with exciton effects determine the optoelectronic properties of Si NWs. We demonstrate that by controlling a set of parameters extracted by fitting the model with experiment and simulation, it is possible to interpret the PL spectral features. The band gap is found to decrease with the increase of NWs diameter and increases more with decreasing oxygenated surface than hydrogenated surface. The finding results are compared with other model calculations and experimental works. The admirable features of the results suggest that the present model is significant for understanding the mechanism of visible PL from Si NWs. The model can be extended for other nanostructures of different shapes and size.

ABSTRAK

Keperluan integrasi terhadap struktur nano (NS) silicon (Si) di dalam bidang alat optik telah membawa kepada pencarian berasaskan bahan silicon dan struktur yang mengeluarkan cahaya dengan kecekapan kuantum yang tinggi. Setelah pemerhatian terhadap suhu bilik fotoluminesen(PL) dilihat dari pandangan struktur nano silikon (Titik atau kawat quantum) dan kemungkinan untuk merubah tindak balas Si NS dengan perubahan saiz, justeru terdapat banyak kerja ujikaji dan teori telah dijalankan bagi menjelaskan asal usul mekanisma fotoluminesen. Walaubagaimanapun mekanisma luminesen dilihat masih tidak jelas walaupun dengan usaha tersebut. Kebelakangan ini, Si struktur nano dengan hydrogen dan oksigen yang dipasivasi menjadi perhatian disebabkan oleh peningkatan emisi cahaya. Tujuan kajian ini adalah untuk membangunkan satu model hibrid kesan penurungan kuantum (QCE), kesan keadaan permukaan (SSE) dan keadaan tenaga ‘exciton’ (EES) untuk mengkaji Si kawat nano spektra PL menggunakan data yang deperoleh daripada ujikaji dan simulasi (pseudo-potensi pendekatan dan kaedah mengikat) penemuan. Satu kaedah simulasi linear mudah (kod Matlab) digunakan untuk mengkaji tindak balas optik Si kawat nano dengan diameter antara 1.5 dan 5.8 nm. Sebuah model intensity integrasi photoluminesen telah diguna pakai untuk menghasilkan spektra PL. Hasil kerja menunjukkan bahawa kedua-dua QCE dan keadaan passivitasi bersama dengan kesan ‘exciton’ menentukan sifat optoelektronik Si kawat nano. Kami menunjukkan bahawa dengan mengawal satu set parameter diekstrak dengan memasang model ujikaji dan simulasi, ia adalah mungkin untuk mentafsir ciri-ciri spectrum PL. Jurang jalur didapati menurun dengan peningkatan diameter NWs dan kenaikan yang lebih dengan penurunan permukaan oksigen daripada permukaan terhidrogenasi. Keputusan pencarian dibandingkan dengan pengiraan model lain dan kerja-kerja ujikaji. Hasil ciri-ciri yang mengagumkan dapat menunjukkan bahawa model ini adalah penting untuk memahami mekanisma terlihat PL daripada Si kawat nano. Model ini boleh dilanjutkan untuk struktur nano yang lain dengan saiz dan bentuk yang berbeza-beza.

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LIST OF ABBREVIATIONS

OD	-	Zero Dimensional
1D	-	One Dimensional
2D	-	Two Dimensional
3D	-	Three Dimensional
AFM	-	Atomic Force Microscopy
CB	-	Conduction Band
CNT	-	Carbon Nano-Tube
C-Si	-	Crystalline Silicon
CVD	-	Chemical Vapor Deposition
DFT	-	Density Function Theory
DFTB	-	Density Functional Tight Binding
DOS	-	Density of State
DVDs	-	Digital Versatile Disks
EBE	-	Excitons Binding Energy
e-h	-	Electron-Hole
Eg	-	Energy Gap
EL	-	Electroluminescence
ETB	-	Empirical Tight Binding
FFT	-	Fast Fourier Transformer

GGA - Generalised Gradient Approximation
ICs - Integrated Circuits
IR - Infra Red
K - Space momentum
LCBB - Linear Combination of Bulk Bands
LDA - Local Density Approximation
LED - Light Emitting Diode
MBE - Molecular beam epitaxy
NC - Nano-Crystal
NC-Si - Nano-Crystalline Silico
NEGF - Non-Equilbrium Green Function
NWs - Nanowires
PECVD - Plasma Enhanced Chemical Vapor Deposition
PBE - Perdew-Burke-Ernzernhof
PL - Photo Luminescence
PLE - Photo Luminescence Excitation
PP - Pseudo-Potential
PPA - Pseudo Potential Approximation
P-Si - Porous Silicon
PMM - Potential Morphing Method
QC - Quantum Confinement
QCE - Quantum Confinement Effect
QCLCM - Quantum Confinement Luminescence Center Model
QCM - Quantum Confinement Model
QD - Quantum Dot

ULSI	-	Ultra Large Scale Integration
UV	-	Ultraviolet
Si	-	Silicon
Si NWs	-	Silicon Nanowires
Si NS	-	Silicon Nanostructure
SC	-	Semi-Conductor
SiO_2	-	Silicon Dioxide
SSM	-	Surface State Model
SSE	-	Surface State Effect
STM	-	Scanning Tunneling Microscopy
TB	-	Tight Binding
TOFMS	-	Time of Flight Mass Spectroscopy
VB	-	Valence Band
VBM	-	Valence Band Minimum
VLS	-	Vapour-Liquid-Solid

LIST OF SYMBOLS

β - Quantum confinement parameter in eV

γ - Quantum confinement parameter

a_o - Exciton Bohr radius

n - Quantum number of different energy level

μ - Reduced mass

E_{RY}^{*} - Exciton Rydberg energy

E_{bulk} - Band gap of bulk material

E_{ex} - Excitonic binding energy

δ - Delta function

g(E) - Density of state as a function of energy

M_e - Mass of electron

M_e^{*} - Effective mass of electron

M_h^{*} - Effective mass of electron

r - Radius

$\Phi_j(r)$ - Atomic wave function

Δq - Wave vector (Momentor)

G_F - Number of degree of freedom

G_C - Number of direction of confinement

O_2 - Oxygen gas

OH	-	Hydroxide
L	-	Crystallite of diameter size
L_o	-	Mean crystallites size
A	-	Surface area
I(L)	-	PL Intensity from crystallite of size
f	-	Oscillator strength
α	-	Size dependence of oscillator strength
σ	-	Standard deviation
N_s	-	Toatal number of surface of state
N_v	-	Number of photo excited carriers in a crystallites proportional to volume
Vc	-	Confinement potential
V_B	-	Volume associated with bulk
V_S	-	Volume associated with surface

CHAPTER ONE

INTRODUCTION

1.1 Introduction

Nanosilicon research has become predominantly significant over last two decades. A nanocrystal made of semiconductor material is now an interesting means of new electronics and optoelectronics devices (Ghoshal et al., 2012). The huge efforts made toward matter manipulation at the nanometer scale have been motivated by the fact that desirable properties can be generated by just changing the material dimension, morphology and shape (Kanemitsu, 1995). In particular, silicon nanowires (NWs) are attracting attention from the electronics industries due to their demand for small devices, from cell phones to computers etc. The discovery of room temperature visible PL from porous silicon and Si nanostructure and possibility of tuning the optical response of silicon nanosized material by modifying their size has invited several attentions in these special kinds of nanoclusters and in small semiconductor structures (Ghoshal et al., 2012). It is found that unlike bulk silicon which has indirect bandgap, the nanostructure Si have direct bandgap and emit light from violet to red depending upon the size of nanostructure (Ghoshal et al., 2014). Silicon NWs constitute an example of one-dimensional (two dimensional confinement) structure with remarkable size and orientation dependent absorption and PL properties (Harrison, 2005).

The nanoscale diameter makes the radial dimension of NWs to be at or below the characteristic length scale such as the exciton Bohr radius, wavelength of light, phonon mean free path, critical size of magnetic domains, exciton diffusion length, etc. As a result, many physical properties of semiconductors are significantly altered within the confined NWs surfaces. In addition, their large surface-to-volume ratio allows for distinct structural and chemical behaviour as well as greater chemical reactivity. This two-dimensional confinement endows NWs with unique properties which stray from those of their corresponding bulk material (O' Mara et al., 1990). Secondly, the one unconstrained dimension can direct the conduction of quantum particles such as electrons, phonons and photons. This control over various forms of energy transport which recommends NWs as ideal materials from which to manufacture advanced solid-state devices (Proot, 1992).

Due to their novel properties silicon nano-wires (Si NWs) have grabbed such great attentions. It is their electrical, optical and mechanical properties that make them to receive such interest (Hassan et al., 2013). Semiconductor material has been widely studied in recent years for their potential use in non-linear optics devices. Silicon is dominant material in the present day microelectronics technology (Shi et al., 2009). However, bulk crystalline silicon is known to be non-linear material of choice due to the long life of its carriers and indirect bandgap in the near infrared (IR) spectral region with very low emission efficiency (one photon emitted for every 10^7 photon-generated electron-hole pair) (Kanemitsu, 1995).

Quantum confinement in NWs result in optical band gap widening which also modify the energy band gap such that visible luminescence is produced as observed experimentally (Ghoshal et al., 2014). The exciton energy states in the forbidden region have an amazing effect on optical properties and the blue shift in PL peak is observed (Kanemitsu, 1995). Our present work will provide more comprehensive account of optical bandgap of varying diameter of nano-wire and also made an effort to develop a phenomenological model to which the luminescence mechanism is investigated.

1.2 Problem Background

Si nanowires being a direct gap, can emit light in the visible range depending upon the size of the nanostructures, and is ideal for optoelectronics applications, unlike bulk silicon which cannot be used in optoelectronics. The search for alternative Si based visible light emitter started, since the discovery of intense red photoluminescence from nanosilicon in 1990 (Canham, 1990; Ledoux et al., 2002a), much work has been carried out to investigate the optical properties of this material. Silicon nanowire is considered as an effective way to turn silicon into a photonic material (Ghoshal et al., 2014). There exist two classes of explanation for the origin of the visible PL, the quantum confinement of e-h pairs and defect states in the oxide layer, still there have been many arguments about the mechanism of the PL in these materials and the question is not yet satisfactory answered (John and Singh, 1995).

1.3 Problem Statement

Recently, the discovery of room temperature visible photoluminescence (PL) from silicon nanostructures has attracted an attentions toward the quantum mechanical nature of this phenomenon (Canham, 1990). The mechanism of visible PL of the Nanoscale Si is far from being understood, despite of several proposed model to explain the luminescence, including quantum confinement, surface states, defects in the chemical and oxides complexes.

The possibility of turning the optical response of silicon nanosized material by modifying their size have invited an intense theoretical and experimental investigations in recent years. The importance of such an investigation stems from the fact that the modeling of such novel materials requires a fundamental understanding of the electronic structure including the role played by surface having different geometry, disorder, and inhomogeneity and so on. In spite of intensive studies, no conclusive argument has been given on the mechanisms of efficient light emission from porous silicon, silicon NWs and related semiconductor materials.

1.4 Research Objectives

i) To derive an expression for NWs diameter dependent PL intensity and band gap.
ii) To determine the effects of confinement and surface on the luminescence properties of NWs by simulating the model using Matlab code.
iii) To compare and validate the model results with other simulations (tight binding and pseudo potential) and experimental data.
iv) To determine the influence of surface state and confinement dependent visible PL and band gap variation.

1.5 Scope of the study

Silicon NWs exhibit room temperature visible PL, but it has a series of disadvantages. The main striking ones are: phenomena issues like size-dependent oscillator strength, radiative recombination rate, the absorption coefficient, weak PL and EL intensity and the influences on surface passivation on optical properties which are not clearly explained. Thus, Scope of this study includes:

i) Combine three models (QC, SS and exciton states) to develop a hybrid model.
ii) Gather experimental data from literatures and fit with the hybrid model to get the empirical fitting parameters.
iii) Derive an expression for NWs size dependent PL intensity and band gap from the model.
iv) Simulate the analytical expression by Matlab program to get size dependent band gap and PL intensity.
v) Compare the hybrid model results with existing experimental and other simulation data and check the validity.

vi) Analyze the data obtained to understand the mechanism of PL emission.

1.6 Significance of the Study

The search for new semiconductors materials and improvement of existing material is an important field of study in material science. Since the discovery of room-temperature visible luminescence from silicon nanostructure by Canham in 1990, many efforts have been made toward the understanding of the PL emission mechanism. Many experimental and theoretical works have been carried out to explain the origin of the visible luminescence (Bruno et al., 2007). A theoretical approach will be explored to verify the experimental and other simulation (tight binding and pseudo potential) work done on the luminescence exhibited by silicon nanostructure. It is hoped to generate fundamental knowledge on room temperature visible emission from Si NWs and widening of the band gap. This model for PL from silicon NWs will set up a systematic theoretical framework to which experimental data can be fit to examine the validity of the model. Regular improvement of knowledge of the fundamental PL mechanism due to quantum confinement, surface defects and exciton energy state of Si NWs will enhance its usage in optoelectronic devices.

1.7 Summary

To summarize, it has been observed that reduction in dimensionality of material from a shank that might be recognized to the nanometer scale changes virtually all of its most basic properties in a fundamental way. Its shape and crystalline structure change, as do its melting and boiling temperatures. Its magnetic properties also change. Material properties change dramatically because quantum effects arise from the confinement of electrons and holes in the materials. The dramatic changes in the properties of these materials due to miniaturization develop an interest in researchers in the field to dream about what to be done next. Silicon quantum wires is considered as unique way to turn silicon into a photonic material and have attracted strong theoretical and experimental investigations in recent years. It’s expected that breakthrough in medicine and computer science will be instrumental in realizing the full potential of nanoscience and technology.

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