EXPLORING NANOSCALE PHENOMENA BY UTILIZING RESONANCE IN SEMICONDUCTOR MATERIALS WITH PLASMONIC AND NANOMECHANICAL DEVICES

DURGESH BANSWAR

DEPARTMENT OF MATERIALS SCIENCE AND ENGINEERING
INDIAN INSTITUTE OF TECHNOLOGY DELHI
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by

Durgesh Banswar

DEPARTMENT OF MATERIALS SCIENCE AND ENGINEERING

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Dedicated to my beloved Parents and Lord Shiva

Durgesh Banswar

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CERTIFICATE

This is to certify that the thesis entitled **Exploring Nanoscale Phenomena by utilizing Resonance in Semiconductor Materials with Plasmonic and Nanomechanical Devices**, submitted by **Mr. Durgesh Banswar**, to the Indian Institute of Technology Delhi, for the award of the degree of **Doctor of Philosophy** (PhD), is a bonafide record of the research work done by him under my supervision. The contents of this thesis work, in full or in parts, have not been submitted to any other Institute or University for the award of any degree or diploma.

Dr. Ankur Goswami
Associate Professor
Department of Materials Science and Engineering (DMSE)
Indian Institute of Technology Delhi, Delhi, India

New Delhi
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(Durgesh Banswar)

ABSTRACT

The domain of nanoscale science and engineering presents how energy and matter behave at the atomic and molecular level, a scale where traditional physics often does not apply. At this scale, materials exhibit unique physical properties that deviate significantly from their bulk counterparts, opening pathways for revolutionary advancements in areas ranging from ultra-sensitive sensing and energy harvesting to quantum computing and advanced optoelectronic. To Unlock these potentials, One need to have a profound understanding of how various physical phenomena manifest and interect when dimensions shrink to the nanometer regime.

This thesis delves into this fascinating world by focusing on the important role of resonance. Resonance, in its fundamental forms: optical and mechanical provides a way for probing, enhancing, and controlling interactions within nanoscale systems. By integrating semiconductor materials with meticulously designed plasmonics and nanomechanical structures, this research aims to reveal complex physical behaviors and develop novel methodologies for characterization and device applications. One critical aspect of understanding materials at the nanoscale is the accurate determination of their mechanical properties. Traditional methods often prove inadequate for structures just a few nanometers thick or wide. Therefore, in the first work, thesis explores the measurement of Youngs modulus by utilizing resonance frequency via optical methods. This noncontact and highly sensitive approach leverages the precise relationship between a nanostructure's resonant frequency and its intrinsic mechanical stiffness. By optically exciting and detecting these minute oscillations, we can calculate fundamental mechanical parameters, providing crucial insights for designing robust and functional nanodevices.

Further, As the optical properties of the semiconductor materials are mainly influenced by quasi-particles such as excitons or trions, the bound states of electron-hole pairs (excitons) or charged electron-hole complexes (trions) which dictate light absorp-

tion and emission characteristics. This work investigates exciton-trion dynamics in semiconductor materials ($MoSe_2$), particularly in the presence of plasmonic nanostructures. Plasmonic resonance in these metallic components creates a highly concentrated local electromagnetic fields that can strongly interact with charge carriers such as excitons and trions, modifying their generation, recombination, and energy transfer pathways. Understanding these light-matter interactions with plasmonics can assist in the development of high-performance nanoscale devices.

Finally, a significant challenge in the efficiency of optoelectronic devices based on perovskite nanocrystals exists known as the phonon bottleneck, which describes the hindered relaxation of energetic charge carriers or excitons due to a mismatch between their discrete electronic energy levels and the continuous phonon spectrum available for energy dissipation. The thesis will explore the implications of this bottleneck and further investigate the potential way to overcome or to minimise its effects by plasmonic interactions, thereby improving the overall efficiency of nanoscale systems.

Through interdisciplinary investigation, this thesis work provides a profound understanding of nanoscale phenomena governed by resonance. By developing and applying advanced optical characterization techniques for mechanical properties, demonstrating complex exciton-trion-plasmon interactions, and addressing the challenges of phonon -mediated energy relaxation, this research delivers a pathway for the implementation of next-generation semiconductor-based plasmonic and nanomechanical applications.

Keywords: Resonance frequency, mechanical resonance, optical resonance, quality factor, plasmons, exciton, trion, phonon-bottolneck.

सारांश:

नैनोस्केल विज्ञान और इंजीनियरिंग का क्षेत्र अपार संभावनाओं की एक सीमा प्रस्तुत करता है, जो पदार्थ और ऊर्जा को उसकी मूलभूत सीमाओं पर हेरफेर करने के अभूतपूर्व अवसर प्रदान करता है। इस छोटे पैमाने पर, सामग्री अद्वितीय भौतिक गुण प्रदर्शित करती है जो अपने बड़े समकक्षों से काफी अलग होती है, जो अति-संवेदनशील संवेदन और ऊर्जा संचयन से लेकर क्वांटम कंप्यूटिंग और उन्नत ऑप्टोइलेक्ट्रॉनिक्स तक के क्षेत्रों में क्रांतिकारी प्रगति के लिए मार्ग खोलती है। इन संभावनाओं को अनलॉक करने के लिए इस बात की गहन समझ की आवश्यकता होती है कि जब आयाम नैनोमीटर शासन में सिकुड़ते हैं तो विभिन्न भौतिक घटनाएँ कैसे प्रकट होती हैं और परस्पर क्रिया करती हैं। यह थीसिस अनुनाद की महत्वपूर्ण भूमिका पर ध्यान केंद्रित करके इस आकर्षक दुनिया में प्रवेश करती है। अनुनाद, अपने असंख्य रूपों में - ऑप्टिकल, मैकेनिकल और इलेक्ट्रिकल नैनोस्केल सिस्टम के भीतर अंतःक्रियाओं की जांच, वृद्धि और नियंत्रण के लिए एक शक्तिशाली उपकरण प्रदान करता है। सावधानीपूर्वक डिज़ाइन किए गए प्लास्मोनिक्स और नैनोमैकेनिकल संरचनाओं के साथ अर्धचालक सामग्रियों को रणनीतिक रूप से एकीकृत करके, इस शोध का उद्देश्य जटिल भौतिक व्यवहारों को उजागर करना और लक्षण वर्णन और उपकरण अनुप्रयोगों के लिए नई पद्धतियाँ विकसित करना है। नैनोस्केल पर सामग्रियों को समझने का एक महत्वपूर्ण पहलू उनके यांत्रिक गुणों का सटीक निर्धारण है। पारंपरिक तरीके अक्सर कुछ नैनोमीटर मोटी या चौड़ी संरचनाओं के लिए अपर्याप्त साबित होते हैं। इसलिए, पहले कार्य में थीसिस ऑप्टिकल विधियों के माध्यम से अनुनाद आवृत्ति विश्लेषण का उपयोग करके यंग्स मापांक के मापन की खोज करती है। यह गैर-संपर्क और अत्यधिक संवेदनशील दृष्टिकोण नैनोस्ट्रक्चर की अनुनाद कंपन आवृत्ति और इसकी आंतरिक यांत्रिक कठोरता के बीच सटीक संबंध का लाभ उठाता है। ऑप्टिकल रूप से उत्तेजित करके और इन सूक्ष्म दोलनों का पता लगाकर, हम मूलभूत यांत्रिक मापदंडों का अनुमान लगा सकते हैं, जो मजबूत और कार्यात्मक नैनोडिवाइस डिजाइन करने के लिए महत्वपूर्ण अंतर्दृष्टि प्रदान करते हैं। इसके अलावा, नैनोस्केल पर अर्धचालक पदार्थों के ऑप्टिकल और इलेक्ट्रॉनिक गुण एक्साइटन और ट्रिऑन जैसे क्वासिपार्टिकल्स से गहराई से प्रभावित होते हैं। इलेक्ट्रॉन-होल युग्मों (एक्साइटन) या आवेशित इलेक्ट्रॉन-होल कॉम्प्लेक्स (ट्रिऑन) की ये बंधी हुई अवस्थाएँ प्रकाश अवशोषण और उत्सर्जन विशेषताओं को निर्धारित करती हैं। यह कार्य अर्धचालक पदार्थों में एक्साइटन-ट्रिऑन गतिशीलता की जांच करता है, विशेष रूप से प्लास्मोनिक नैनोस्ट्रक्चर की उपस्थिति में। इन धातु घटकों में स्थानीयकृत सतह प्लास्मोन अनुनाद अत्यधिक केंद्रित विद्युत चुम्बकीय क्षेत्र बनाते हैं जो एक्साइटन और ट्रिऑन के साथ दृढ़ता से बातचीत कर सकते हैं, उनकी पीढ़ी, पुनर्संयोजन और ऊर्जा हस्तांतरण मार्गों को संशोधित कर सकते हैं। इन जटिल प्रकाश-पदार्थ इंटरैक्शन को समझना और उनका उपयोग करना उच्च-प्रदर्शन नैनोस्केल ऑप्टोइलेक्ट्रॉनिक उपकरणों को विकसित करने की कुंजी है।

अंत में, नैनोस्केल ऊर्जा प्रबंधन और डिवाइस दक्षता में एक महत्वपूर्ण चुनौती, विशेष रूप से नैनोक्रिस्टल और

अन्य सीमित अर्धचालक प्रणालियों में, फोनन अड़चन है। यह घटना उनके असतत इलेक्ट्रॉनिक ऊर्जा स्तरों और ऊर्जा अपव्यय के लिए उपलब्ध निरंतर फोनन स्पेक्ट्रम के बीच बेमेल के कारण ऊर्जावान चार्ज वाहकों या एक्साइटन के बाधित विश्राम का वर्णन करती है। थीसिस इस अड़चन के निहितार्थों का पता लगाएगी और इसके प्रभावों को कम करने के लिए प्लास्मोनिक और नैनोमैकेनिकल इंटरैक्शन को शामिल करने वाली संभावित रणनीतियों की जांच करेगी, जिससे नैनोस्केल सिस्टम की समग्र दक्षता में सुधार होगा। इस अंतःविषय जांच के माध्यम से, इस थीसिस का उद्देश्य अनुनाद द्वारा शासित नैनोस्केल घटनाओं की मौलिक समझ में योगदान करना है। यांत्रिक गुणों के लिए उन्नत ऑप्टिकल लक्षण वर्णन तकनीकों को विकसित और लागू करके, जटिल एक्साइटन-ट्रियन-प्लास्मोन इंटरैक्शन को स्पष्ट करके, और फोनन-मध्यस्थ ऊर्जा विश्राम की चुनौतियों का समाधान करके, यह शोध अगली पीढ़ी के उच्च-प्रदर्शन अर्धचालक आधारित प्लास्मोनिक और नैनोमैकेनिकल उपकरणों के डिजाइन और कार्यान्वयन का मार्ग प्रशस्त करना चाहता है।

मुख्य शब्द: अनुनाद आवृत्ति, यांत्रिक अनुनाद, ऑप्टिकल अनुनाद, गुणवत्ता कारक, प्लास्मोन्स, एक्सिटोन, ट्रियोन, फोनोन-बॉटलनेक।।

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AFM	Atomic Force Microscopy
Ag	Silver
Au	Gold
C.B	Conduction Band
DMF	Dimethylformamide
EDS	Energy Dispersive X-ray Spectroscopy
ETR	Exciton Trion Intensity Ratio
$FAPbBr_3$	Formamidinium Lead Bromide
FEM	Finite Element Method
FESEM	Field Enhanced Scanning Electron Microscopy
FWHM	Full Width Half Maximum
Ga	Gallium
GGA	Generalized Gradient Approximation
GPNS	Gallium Based Plasmonic Nanosubstrates
HEA	High Entropy Alloy
LDV	Laser Doppler Vibrometer
LEDs	Light Emitting Diodes
LO	Lorentz Oscillator
LPF	Low Pass Filter
LSPR	Local Surface Plasmon Resonance
MEMS	Micro Electromechanical Systems

$MoSe_2$	Molybdenum Diselenide
NCs	Nanocrystals
NEMS	Nano Electromechanical Systems
NTP	Normal Temperature Pressure
PL	Photoluminescence Spectroscopy
PNC	Photonic Crystal Cavity
RFMU	Resonance Freqency Measurement Unit
RN	Exciton Radiative Recombination Rate
SEM	Scanning Electron Microscopy
SERS	Surface Enhanced Raman Spectroscopy
SHM	Simple Harmonic Oscillator Model
SPs	Surface Plasmons
TA	Transient Absorbance
TCSPC	Time Correlated Single Photon Counting
TMD	Transition Metal Dichalcogenide
UV-vis	Ultraviolet Visible
VO_2	Vanadium Di Oxide
WLC	White Light Continuum

List of Symbols

k_{eff}	stiffness
ϕ	Phase
Q	Quality factor
ω	frequency
I	moment of inertia
ρ	density
T	Temperature
γ	damping coefficient
$H(\omega_{id,\eta})$	hydrodynamic load function
A_{ex}	A exciton
B_{ex}	B exciton
B_{tr}	A trion
$E_g(eV)$	Bandgap energy
χ_B	exciton-trion energy difference
E_B^-	intrinsic B trion binding energy
$E_F(eV)$	Fermi energy
N	exciton density
R_{N}	exciton radiative recombination rate
R_{TF}	exciton to trion scattering rate
P	vacant phonon density of states
R_{NRT}	trion non-radiative recombination rate
D	total available phonon density of states
c_e	ionized electron density in conduction band
T_r	Trion density
X_{BE}	population of carriers at bandedge

X_E	excited carriers
D_{L0}	phonon density of states
E_{c}	conduction band edge energy
E_{th}	thermal energy
L_o	occupied optical phonon states
T_{NR}	intraband recombination rate
G	generation rate
R	radiative recombination rate
NR	non radiative recombination rate
KC	Klemen's channe