

**TRANSITION METAL DICHALCOGENIDE  
BASED NANOHETEROSTRUCTURES FOR  
ENERGY STORAGE AND OPTOELECTRONIC  
APPLICATIONS**

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INDIAN INSTITUTE OF TECHNOLOGY DELHI  
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AND OPTOELECTRONIC APPLICATIONS**

by

**SAHIL SINGH**

**DEPARTMENT OF CHEMISTRY**

Submitted

in fulfilment of the requirements of the degree of Doctor of Philosophy  
to the



**INDIAN INSTITUTE OF TECHNOLOGY DELHI**

**JULY 2025**

*Dedicated to  
my parents*

# CERTIFICATE

This is to certify that the thesis entitled, “**TRANSITION METAL DICHALCOGENIDE BASED NANOHETEROSTRUCTURES FOR ENERGY STORAGE AND OPTOELECTRONIC APPLICATIONS**” being submitted by **Mr. Sahil Singh** to the **Indian Institute of Technology Delhi** for the award of the degree of **Doctor of Philosophy** in Chemistry, represents authentic research conducted by him. Mr. Sahil Singh has conducted this research under my guidance and supervision. He has successfully met the submission requirements for this thesis, which, to the best of my knowledge, meets the necessary standards.

Date

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**Sahil Singh**

# ABSTRACT

Two-dimensional (2D) transition metal dichalcogenides (TMDs) have garnered significant attention due to their distinctive and tunable properties, including quantum-well with indirect-to-direct band transitions, intrinsic catalytic activity, thickness-dependent band gaps, high in-plane charge carrier mobility, large specific surface area, and pronounced spin-orbit coupling. The functionality of these materials is strongly dictated by their surface characteristics- with edge sites playing a key role in catalytic applications, while basal planes are crucial for optoelectronic devices. This thesis aims to harness these distinct surface features for electrochemical and optoelectronic devices, enabled by developing various synthetic methodologies. Defect-rich MoSe<sub>2</sub> nanosheets were synthesized using a colloidal route, and these intrinsic defects were effectively exploited to study the two distinct transformative processes and construct nanoheterostructures (NHSs) with different functional materials which tunes charge transfer resistance, increases the specific surface area and widens the absorption window. These NHSs were systematically studied for their performance in supercapacitors and photodiodes, highlighting their multifunctionality and promising potential for next-generation device applications.

The first two chapters (**Chapter 1 and 2**) are dedicated to the extensive literature survey of research work related to TMDs, metal chalcogenides (binary: CdS, CdSe, Co<sub>9</sub>S<sub>8</sub>; ternary: CuFeS<sub>2</sub>), their structure, applications, challenges, and possible solutions, along with summary of various techniques used for the characterization of as-synthesized materials. In **Chapter 3**, we demonstrated that the chemical reactivity of atomically thin two-dimensional transition metal dichalcogenides (TMDs) is strongly influenced by structural defects. We focused on the role of defects in MoSe<sub>2</sub> nanosheets (NSs) in facilitating two distinct transformations: the formation of

MoSe<sub>2</sub>-CdS nano-heterostructures (NHSs) and CdSe quantum dots (QDs). Passivating these defects with thiol groups enabled controlled CdS growth, resulting in MoSe<sub>2</sub>-CdS NHSs. In the absence of thiols, the defects accelerated cation displacement reactions (CDR) with cadmium (Cd) precursors, leading to the formation of CdSe QDs. This transformation was confirmed through Raman and optical spectroscopy. Additionally, density functional theory (DFT) calculations showed that CDR occurs preferentially on defect-rich surfaces, while defect-free or thiol-passivated MoSe<sub>2</sub> remains unreactive. These findings underscore the crucial role of defects in controlling the reactivity and chemical transformations of TMDs. In **Chapter 4**, we report a facile hot-injection colloidal strategy for synthesizing MoSe<sub>2</sub>-Co<sub>9</sub>S<sub>8</sub> nanoheterostructures (NHSs) by passivating defects in MoSe<sub>2</sub> nanosheets (NSs), enabling the epitaxial growth of Co<sub>9</sub>S<sub>8</sub> nanoparticles (NPs) on their basal planes. X-ray photoelectron spectroscopy confirmed intimate interfacial contact, while interfacial engineering reduced nanoparticle agglomeration, enhanced specific surface area, and improved charge transfer due to the formation of interfacial defects. Benefiting from the pseudocapacitive nature of cobalt and improved conductivity, MoSe<sub>2</sub>-Co<sub>9</sub>S<sub>8</sub> NHSs delivered a high specific capacitance of 910.5 F g<sup>-1</sup> at 1 A g<sup>-1</sup>, with excellent cyclic stability, retaining ~90% capacitance and 93.3% coulombic efficiency after 10,000 cycles at 15 A g<sup>-1</sup>. As a proof-of-concept, coin cell supercapacitors fabricated with MoSe<sub>2</sub>-Co<sub>9</sub>S<sub>8</sub> NHSs and [EMIM][BF<sub>4</sub>]:PVDF electrolyte demonstrated impressive electrochemical performance, highlighting their potential for next-generation high-performance energy storage devices. In **Chapter 5**, we established an efficient colloidal synthesis strategy for MoSe<sub>2</sub>-CuFeS<sub>2</sub> (MCFS) nanoheterostructures (NHSs), utilizing defect-passivated MoSe<sub>2</sub> nanosheets (NSs) as templates for the growth of CuFeS<sub>2</sub> (CFS) nanoparticles (NPs). Sulfur passivation of selenium vacancies facilitated uniform CFS nucleation, as confirmed by transmission electron microscopy (TEM) and

X-ray photoelectron spectroscopy (XPS), which revealed strong atomic-level interfacial contact. The MCFS NHSs exhibited broad optical absorption spanning the UV-visible-NIR regions, enabling superior light–matter interactions. To assess their optoelectronic properties, photodiodes were fabricated using pure MoSe<sub>2</sub> NSs, pure CFS NPs, and MCFS NHSs with ITO as the substrate and MoO<sub>3</sub> as a hole transport layer. Among these, devices based on MCFS NHSs demonstrated enhanced ON/OFF ratios and improved charge transfer, attributed to the intimate interface and wide spectral response. **Chapter 6** summarizes the key findings of this thesis, highlighting how the supercapacitor and optoelectronic properties of TMDs have been tuned for diverse applications. It also offers a brief outlook on future directions and potential advancements based on the work presented.

## संक्षेपण

द्वि-आयामी (2D) संक्रमण धातु डाइचेल्कोजेनाइड्स (TMDs) ने अपने विशिष्ट और ट्यूनेबल गुणों के कारण महत्वपूर्ण ध्यान आकर्षित किया है, जिसमें अप्रत्यक्ष-से-प्रत्यक्ष बैंड संक्रमण के साथ क्वांटम-वेल, आंतरिक उत्प्रेरक गतिविधि, मोटाई पर निर्भर बैंड अंतराल, उच्च इन-प्लेन चार्ज वाहक गतिशीलता, बड़े विशिष्ट सतह क्षेत्र और स्पष्ट स्पिन-ऑर्बिट युग्मन शामिल हैं। इन सामग्रियों की कार्यक्षमता उनकी सतह विशेषताओं द्वारा दृढ़ता से निर्धारित होती है- किनारे वाले स्थान उत्प्रेरक अनुप्रयोगों में महत्वपूर्ण भूमिका निभाते हैं, जबकि बेसल प्लेन ऑप्टोइलेक्ट्रॉनिक उपकरणों के लिए महत्वपूर्ण होते हैं। इस थीसिस का उद्देश्य विभिन्न सिंथेटिक तरीकों को विकसित करके इलेक्ट्रोकेमिकल और ऑप्टोइलेक्ट्रॉनिक उपकरणों के लिए इन विशिष्ट सतह विशेषताओं का उपयोग करना है। दोष-समृद्ध  $\text{MoSe}_2$  नैनोशीट को कोलाइडल मार्ग का उपयोग करके संश्लेषित किया गया इन एनएचएस का सुपरकैपेसिटर और फोटोडियोड में उनके प्रदर्शन के लिए व्यवस्थित रूप से अध्ययन किया गया, जिससे उनकी बहुक्रियाशीलता और अगली पीढ़ी के उपकरण अनुप्रयोगों के लिए आशाजनक क्षमता पर प्रकाश डाला गया।

पहले दो अध्याय (अध्याय 1 और 2) टीएमडी, धातु चालकोजेनाइड्स (बाइनरी: सीडीएस, सीडीएसई,  $\text{Co}_9\text{S}_8$ ; टर्नरी:  $\text{CuFeS}_2$ ), उनकी संरचना, अनुप्रयोगों, चुनौतियों और संभावित समाधानों से संबंधित शोध कार्यों के व्यापक साहित्य सर्वेक्षण के साथ-साथ संश्लेषित सामग्रियों के लक्षण वर्णन के लिए उपयोग की जाने वाली विभिन्न तकनीकों के सारांश के लिए समर्पित हैं। अध्याय 3 में, हमने दिखाया कि परमाणु रूप से पतले द्वि-आयामी संक्रमण धातु डाइचेल्कोजेनाइड्स (टीएमडी) की रासायनिक प्रतिक्रियाशीलता संरचनात्मक दोषों से काफी प्रभावित होती है। हमने दो अलग-अलग परिवर्तनों को सुविधाजनक बनाने में  $\text{MoSe}_2$  नैनोशीट्स (NSs) में दोषों की भूमिका पर ध्यान केंद्रित किया:  $\text{MoSe}_2$ -CdS नैनो-हेटेरोस्ट्रक्चर (NHS) और CdSe क्वांटम डॉट्स (QDs) का निर्माण थियोल्स की अनुपस्थिति में, दोषों ने कैडमियम (Cd) अग्रदूतों के साथ धनायन

विस्थापन अभिक्रिया (CDR) को त्वरित किया, जिससे CdSe QDs का निर्माण हुआ। इस परिवर्तन की पुष्टि रमन और ऑप्टिकल स्पेक्ट्रोस्कोपी के माध्यम से की गई। इसके अतिरिक्त, घनत्व कार्यात्मक सिद्धांत (DFT) गणनाओं से पता चला कि CDR दोष-समृद्ध सतहों पर अधिमानतः होता है, जबकि दोष-मुक्त या थियोल-निष्क्रिय MoSe<sub>2</sub> अक्रियाशील रहता है। ये निष्कर्ष TMDs की प्रतिक्रियाशीलता और रासायनिक परिवर्तनों को नियंत्रित करने में दोषों की महत्वपूर्ण भूमिका को रेखांकित करते हैं। अध्याय 4 में, हम MoSe<sub>2</sub> नैनोशीट (NS) में दोषों को निष्क्रिय करके MoSe<sub>2</sub>-Co<sub>9</sub>S<sub>8</sub> नैनोहेटेरोस्ट्रक्चर (NHS) को संश्लेषित करने के लिए एक आसान हॉट-इंजेक्शन कोलाइडल रणनीति की रिपोर्ट करते हैं, जिससे उनके बेसल प्लेन पर Co<sub>9</sub>S<sub>8</sub> नैनोपार्टिकल्स (NPs) की एपिटैक्सियल वृद्धि संभव हो पाती है। एक्स-रे फोटोइलेक्ट्रॉन स्पेक्ट्रोस्कोपी ने अंतरंग इंटरफेशियल संपर्क की पुष्टि की, जबकि इंटरफेशियल इंजीनियरिंग ने नैनोपार्टिकल एग्लोमरेशन को कम किया, विशिष्ट सतह क्षेत्र को बढ़ाया और इंटरफेशियल दोषों के गठन के कारण चार्ज ट्रांसफर में सुधार किया। कोबाल्ट की छद्म कैपेसिटिव प्रकृति और बेहतर चालकता से लाभ उठाते हुए, MoSe<sub>2</sub>-Co<sub>9</sub>S<sub>8</sub> NHS ने 1 A g<sup>-1</sup> पर 910.5 F g<sup>-1</sup> की उच्च विशिष्ट धारिता प्रदान की, जिसमें उत्कृष्ट चक्रीय स्थिरता थी, 15 A g<sup>-1</sup> पर 10,000 चक्रों के बाद ~90% धारिता और 93.3% कोलोम्बिक दक्षता बनाए रखी। अवधारणा के प्रमाण के रूप में, MoSe<sub>2</sub>-Co<sub>9</sub>S<sub>8</sub> NHS और [EMIM][BF<sub>4</sub>]:PVDF इलेक्ट्रोलाइट के साथ निर्मित कॉइन सेल सुपरकैपेसिटर ने प्रभावशाली इलेक्ट्रोकेमिकल प्रदर्शन का प्रदर्शन किया, जो अगली पीढ़ी के उच्च-प्रदर्शन ऊर्जा भंडारण उपकरणों के लिए उनकी क्षमता को उजागर करता है। अध्याय 5 में, हमने MoSe<sub>2</sub>-CuFeS<sub>2</sub> (MCFS) नैनोहेटेरोस्ट्रक्चर (NHS) के लिए एक कुशल कोलाइडल संश्लेषण रणनीति स्थापित की, जिसमें CuFeS<sub>2</sub> (CFS) नैनोकणों (NPs) की वृद्धि के लिए टेम्पलेट के रूप में दोष-निष्क्रिय MoSe<sub>2</sub> नैनोशीट (NSs) का उपयोग किया गया। सेलेनियम रिक्तियों के सल्फर निष्क्रियता ने एक समान CFS न्यूक्लियेशन की सुविधा प्रदान की, जैसा कि ट्रांसमिशन इलेक्ट्रॉन माइक्रोस्कोपी (TEM) और एक्स-रे फोटोइलेक्ट्रॉन

स्पेक्ट्रोस्कोपी (XPS) द्वारा पुष्टि की गई, जिसने मजबूत परमाणु-स्तरीय इंटरफेसियल संपर्क का खुलासा किया। MCFS NHS ने UV-दृश्य-NIR क्षेत्रों में फैले व्यापक ऑप्टिकल अवशोषण का प्रदर्शन किया, जिससे बेहतर प्रकाश-पदार्थ इंटरैक्शन संभव हुआ। उनके ऑप्टोइलेक्ट्रॉनिक गुणों का आकलन करने के लिए, सब्सट्रेट के रूप में ITO और होल ट्रांसपोर्ट लेयर के रूप में  $\text{MoO}_3$  के साथ शुद्ध  $\text{MoSe}_2$  NSs, शुद्ध CFS NPs और MCFS NHS का उपयोग करके फोटोडियोड का निर्माण किया गया। इनमें से, MCFS NHS पर आधारित उपकरणों ने बेहतर ON/OFF अनुपात और बेहतर चार्ज ट्रांसफर का प्रदर्शन किया, जिसका श्रेय अंतरंग इंटरफेस और विस्तृत स्पेक्ट्रल प्रतिक्रिया को जाता है। अध्याय 6 इस थीसिस के प्रमुख निष्कर्षों का सारांश देता है, जिसमें बताया गया है कि TMD के सुपरकैपेसिटर और ऑप्टोइलेक्ट्रॉनिक गुणों को विभिन्न अनुप्रयोगों के लिए कैसे ट्यून किया गया है। यह प्रस्तुत कार्य के आधार पर भविष्य की दिशाओं और संभावित प्रगति पर एक संक्षिप्त दृष्टिकोण भी प्रस्तुत करता है।

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## GLOSSARY OF SYMBOLS AND ABBREVIATIONS

0D	Zero-dimensional
1D	One-dimensional
2D	Two-dimensional
3D	Three-dimensional
QDs	quantum dots
NPs	nanoparticles
NSs	nanosheets
hBN	Hexagonal boron nitride
RT	room temperature
TMDs	transition metal dichalcogenides
DOS	Density of states
PL	photoluminescence
NIR	near-infrared
LEDs	light-emitting diodes
BZ	Brillouin zone
SOC	Spin-orbit coupling
MoSe <sub>2</sub>	molybdenum diselenide
CVD	chemical vapor deposition
MBE	molecular beam epitaxy
STM	scanning tunneling microscope
NHSs	nanoheterostructures

CFS	copper iron sulfides
CB	conduction band
VB	valence band
SCs	supercapacitors
EDLCs	electrical double-layer capacitors
PCs	pseudocapacitors
AFM	atomic force microscopy
rGO	reduced graphene-oxide
MGF	mesoporous graphene foam
CdX	cadmium chalcogenide
CNTs	carbon nanotubes
ZB	zinc-blende
WZ	wurtzite
RS	rock salt
HER	hydrogen evolution reaction
OER	oxygen evolution reaction
ORR	oxygen reduction reactions
LIBs	lithium-ion batteries
XRD	X-ray diffraction
TEM	transmission electron microscopy
FESEM	field emission scanning electron microscopy
EIS	electrochemical impedance spectroscopy
TRPL	time resolved photoluminescence

EDX	Energy dispersive X-ray
FTIR	Fourier transform infrared
XPS	X-ray photoelectron spectroscopy
HRTEM	High-Resolution Transmission Electron Microscopy HRTEM
K.E.	kinetic energy
B.E.	binding energy
A	absorbance
T	transmittance
UV	ultraviolet
ps	picosecond
ns	nanoseconds
$\mu$ s	microseconds
ms	milliseconds
IC	internal conversion
ISC	intersystem crossing
$\Gamma$	fluorescence lifetime
$E_g$	band gap
eV	electron volts
CV	cyclic voltammetry
GCD	galvanostatic charge-discharge
DFT	density functional theory
TCSPC	time correlated single photon counting
PAW	projected augmented wave

FWHM	full width at half maximum
PBE	Perdew, Burke and Ernzerhof
$\Gamma_{avg}$	average PL lifetime
GGA	generalized gradient approximation
Vis	visible
$\Gamma_{mean}$	mean lifetime
$\Delta G$	Gibbs free energy
BET	Brunauer-Emmett-Teller
CDR	cation displacement reaction
DDT	dodecanethiol
OLAM	oleylamine
FFT	fast Fourier transform
PVDF	polyvinylidene fluoride
KOH	potassium hydroxide
$E$	energy density
$P$	power density
[EMIM][BF <sub>4</sub> ]	1-Ethyl-3-methylimidazolium tetrafluoroborate
$D$	diffusion coefficient
$R$	universal gas constant
$T$	absolute temperature
$n$	number of electrons
$\nu$	scan rate
$F$	Faraday's constant

ESR	equivalent series resistance
EDL	electric double layer
IL	ionic liquid
$C_{sp}$	specific capacitance
SLM	Single-layer $\text{MoSe}_2$
NCs	nanocrystals
DSSCs	dye-sensitized solar cells
IB	interband
SAED	selected area electron diffraction
SPR	surface plasmon resonance
$\mu\text{A}$	microampere
ITO	indium tin oxide
ODE	1-Octadecene
$\text{MoO}_3$	molybdenum trioxide
Ag	silver
$I_p$	photocurrent
$I_d$	dark current