

MICROCAVITY COUPLED INTERLAYER EXCITONS IN MOSe_2 - WSe_2 HETEROSTRUCTURES

A THESIS SUBMITTED TO
THE GRADUATE SCHOOL OF ENGINEERING AND SCIENCE
OF BILKENT UNIVERSITY
IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR
THE DEGREE OF
MASTER OF SCIENCE
IN
MATERIALS SCIENCE AND NANOTECHNOLOGY

By
Şeyma Esra Atalay
August 2024

Microcavity Coupled Interlayer Excitons in MoSe₂ - WSe₂ Heterostructures

By Şeyma Esra Atalay

August 2024

We certify that we have read this thesis and that in our opinion it is fully adequate, in scope and in quality, as a thesis for the degree of Master of Science.

İbrahim Sarpkaya(Advisor)

Serkan Ateş

Wonmi Ahn

Approved for the Graduate School of Engineering and Science:

Orhan Arıkan
Director of the Graduate School

ABSTRACT

MICROCAVITY COUPLED INTERLAYER EXCITONS IN MoSe_2 - WSe_2 HETEROSTRUCTURES

Şeyma Esra Atalay

M.S. in Materials Science and Nanotechnology

Advisor: İbrahim Sarpkaya

August 2024

After the discovery of graphene, two-dimensional (2D) materials gained immense attention due to their exceptional mechanical, optical, and electronic properties. One of the well-known family of 2D materials is transition metal dichalcogenides (TMDs). Their electronic bandgap makes a transition from indirect to direct when the monolayer limit is reached, making them an excellent medium for studying many-body interactions in condensed matter and light-matter interactions. Also, stacking monolayer TMDs on top of each other enables the creation of heterostructures (HS). The vertical van der Waals heterostructures made from monolayer TMDs can host two types of excitons: one is the intralayer excitons consisting of the strongly Coulomb-bound electron-hole pair within the same material, and the other one is the interlayer exciton made up by spatially separated electrons and holes located in different layers. Intralayer excitons in TMDs exhibit higher oscillator strengths due to spatial confinement. Furthermore, coupling intra- and interlayer excitons with optical cavities allows for observing anticrossing phenomena between excitons and cavity photons. Consequently, TMDs and their HSs provide a versatile platform for investigating exciton-polariton formation and their associated photophysical properties. This thesis presents a detailed study of the room- and low-temperature photoluminescence (PL) spectroscopy of interlayer excitons (IEX) in near-hexagonal MoSe_2 – WSe_2 HSs. The studied hexagonal boron nitride (hBN) encapsulated heterostructures were fabricated using a combination of three methods: (i) mechanical exfoliation for cleaving the monolayers from bulk material, (ii) dry transfer technique to stack them vertically to achieve the heterostructure and (iii) edge identification method to adjust the twist angle between monolayers during stacking. Fabry-Pérot planar microcavities were fabricated for both room and low-temperature studies with different cavity modes using plasma-enhanced chemical vapor deposition with the aim of

studying the interlayer exciton-polaritons. Fabricated cavities were further characterized by transmission electron microscopy and focused ion beam lithography for imaging the alternating layers of distributed Bragg mirrors (DBRs) and their thicknesses. The PL of IEXs in $\text{MoSe}_2 - \text{WSe}_2$ heterostructures were measured both at room and low temperatures. Low-temperature PL measurements were conducted using a closed-cycle cryostat integrated into a home-built micro-PL setup. The results indicate that our heterostructures exhibit a near-hexagonal structure. The intensities of the spin-triplet and the spin-singlet states of the IEXs are prominent in the energy range of approximately 1.35-1.42 eV. The observed splitting between the spin-triplet and spin-singlet IEXs was in the range of 27-34 meV. Further investigations into the temperature and pump power dependence of the IEXs revealed that the intensity of the spin-triplet IEX is highly sensitive to temperature variations between 3.5 K and 50 K, becoming less intense at higher temperatures. Although the intensity of the spin-singlet IEX also decreased, it remained detectable at 50 K. With increasing pump power, a blueshift of 15 meV was observed for the spin-triplet IEX, while the spin-singlet IEX exhibited a blueshift of only 3 meV in the same pump power range. This indicates that the density of the spin-triplet state IEX increases more significantly with higher pump power than the spin-singlet state IEX. Additionally, the lifetime of the spin-singlet IEX was measured using Time-Resolved Photoluminescence (TRPL) spectroscopy. The fast and slow decay components were in the range of a few nanoseconds and a few tens of nanoseconds, respectively. Moreover, an approximately 9 meV splitting on the spin-singlet IEX PL emission was observed in one of the studied emitters, which has the same resonance wavelength as the FP cavity. This splitting might be attributed to the interlayer exciton-polariton formation at $k_{\parallel}$ equals zero. However, the whole angle-resolved PL spectrum should be measured to ensure this emission belongs to the polariton formation.

Keywords: Transition metal dichalcogenides, interlayer excitons, exciton-polaritons, photoluminescence spectroscopy, Fabry-Pérot cavity.

ÖZET

MİKROKAVİTE İÇERİSİNDE ENTEGRE MOSE₂ - WSE₂ HETEROYAPILARINDA KATMANLAR ARASI EKSİTONLAR

Şeyma Esra Atalay

Malzeme Bilimi ve Nanoteknoloji, Yüksek Lisans

Tez Danışmanı: İbrahim Sarpkaya

Ağustos 2024

Grafenin keşfinden sonra, iki boyutlu (2D) malzemeler olağanüstü mekanik, optik ve elektronik özellikleri nedeniyle büyük ilgi gördü. İyi bilinen 2D malzeme ailelerinden biri geçiş metali dikalkogenidlerdir (TMD'ler). Elektronik bant aralıkları, tek tabaka sınırına ulaşıldığında dolaylıdan doğrudana geçer ve bu da onları yoğun madde ve ışık-madde etkileşimlerinde çok cisim etkileşimlerini incelemek için mükemmel bir ortam haline getirir. Ayrıca, tek tabaka TMD'lerini üst üste istiflemek, heteroyapıların (HS) oluşturulmasını sağlar. Tek tabaka TMD'lerinden yapılan dikey van der Waals heteroyapılarında iki tür eksiton oluşumu gözlenir: biri, aynı malzeme içinde güçlü bir şekilde Coulomb etkileşimli elektron-deşik çiftinden oluşan katman içi eksitonlar ve diğeri, farklı katmanlardaki uzamsal olarak ayrılmış elektronlar ve deşiklerden oluşan katmanlar arası eksitondur. TMD'lerdeki katman içi eksitonlar, uzamsal sınırlama nedeniyle daha yüksek osilatör kuvvetleri sergiler. Ayrıca, katman içi ve katmanlar arası eksitonların optik kaviteler ile birleştirilmesi, eksitonlar ve fotonlar arasındaki çapraz geçiş olaylarının gözlenmesine olanak tanır. Sonuç olarak, TMD'ler ve onların heteroyapıları, eksiton-polariton oluşumunu ve ilişkili fotofiziksel özelliklerini araştırmak için çok yönlü bir platform sağlar. Bu tez, neredeyse hegzagonal MoSe₂ – WSe₂ HS'larda katmanlar arası eksitonların (IEX) oda ve düşük sıcaklık fotoluminesans (PL) spektroskopisinin ayrıntılı bir çalışmasını sunar. İncelenen hegzagonal bor nitrür (hBN) kapsüllenmiş heteroyapılar, aşağıdaki yöntemlerin bir kombinasyonu ile üretilmiştir: (i) yığın malzemeden monokatmanları ayırmak için mekanik eksfoliasyon, (ii) heteroyapıya ulaşmak için bunları dikey olarak

istiflemek üzere kuru transfer tekniği ve (iii) istifleme sırasında monokatmanlar arasındaki dönme açısını ayarlamak için kenar tanımlama yöntemi. Fabry-Pérot (FP) düzlemsel mikro kavite, katmanlar arası eksiton-polaritonları incelemek amacıyla plazma destekli kimyasal buhar biriktirme yöntemi kullanılarak farklı kavite modlarıyla hem oda hem de düşük sıcaklık fotolüminesans spektroskopisi çalışmaları için üretildi. Üretilen boşluklar, dağıtılmış Bragg aynalarının (DBR'ler) dönüşümlü katmanlarını ve kalınlıklarını görüntülemek için transmisyon elektron mikroskobu ve odaklanmış iyon demeti litografisi ile daha detaylı olarak karakterize edildi. $\text{MoSe}_2 - \text{WSe}_2$ heteroyapılarındaki IEX'lerin PL'ı hem oda hem de düşük sıcaklıklarda ölçüldü. Düşük sıcaklık ölçümleri, bir mikro-PL kurulumuna entegre edilmiş kapalı çevrimli bir kriyostat kullanılarak gerçekleştirildi. Sonuçlar, heteroyapılarımızın neredeyse altıgen bir yapı sergilediğini göstermektedir. IEX'lerin spin-üçlü ve spin-tekli durumlarının yoğunlukları yaklaşık 1,35-1,42 eV enerji aralığında belirgindir. Spin-üçlü ve spin-tekli IEX'lerin arasında gözlemlenen bölünme 27-34 meV aralığındaydı. IEX'lerin sıcaklık ve pompa gücü bağımlılığına ilişkin daha ileri araştırmalar, spin-üçlü IEX'nun yoğunluğunun 3,5 K ile 50 K arasındaki sıcaklık değişimlerine karşı oldukça hassas olduğunu ve daha yüksek sıcaklıklarda daha az yoğun hale geldiğini ortaya koydu. Spin-tekli IEX'nun yoğunluğu da azalmasına rağmen, 50 K'de tespit edilebilir düzeyde kaldı. Pompa gücünün benzer bir uyarım aralığı içinde artmasıyla birlikte, spin-üçlü IEX için 15 meV'luk bir maviye kayma gözlemlenirken, spin-tekli IEX yalnızca 3 meV'luk bir maviye kayma sergiledi. Bu, spin-üçlü durum IEX'lerinin yoğunluğunun, spin-tekli durum IEX'una göre daha yüksek pompa gücüyle daha önemli ölçüde arttığını göstermektedir. Ek olarak, spin-tekli IEX'un ömrü zaman çözünürlüklü fotolüminesans (TRPL) spektroskopisi kullanılarak ölçüldü. Hızlı ve yavaş bozunma bileşenleri sırasıyla birkaç nanosaniye ve birkaç on nanosaniye aralığındaydı. Dahası, FP kavite ile aynı rezonans dalga boyuna sahip olan spin-tekli IEX emisyonunda enerjide yaklaşık 9 meV bölünme gözlemlendi. Bu bölünme, $k_{\parallel}$ sıfıra eşit olduğunda katmanlar arası eksiton-polariton oluşumuna atfedilebilir. Ancak, bu emisyonun polariton oluşumuna ait olduğundan emin olmak için tüm açı çözünürlüklü PL spektrumu ölçülmelidir.

Anahtar sözcükler: Geçiş metali dikalkojenitler, ara katman eksitonları, eksiton-polaritonlar, fotolüminesans spektroskopisi, Fabry-Pérot kavite.

Acknowledgement

It has been a long journey to earn my master's degree. There were tears but also moments of joy and achievement. To readers, I hope you enjoy reading; hopefully, I will be helpful to someone with this study. Most importantly, thank you for your interest.

I want to thank my group leader, Asst. Prof. Dr. İbrahim Sarpkaya, for his invaluable support and guidance throughout my academic journey. I am sincerely appreciative of his confidence in me, as well as for welcoming me into his group. His unwavering support has been instrumental in my studies, and I am grateful for his willingness always to address my questions and concerns.

I also want to thank my thesis committee, Assoc. Prof. Dr. Serkan Ateş and Asst. Prof. Dr. Wonmi Ahn for their precious time and comments.

Big thanks to my fantastic colleague, Hilal Korkut, from the "Quantum Photonics Lab," for always having my back and offering invaluable assistance whenever I needed it. I genuinely believe she will be an exceptional group leader in the future. I thank Mehmet Atıf Durmuş for being a good friend and his technical expertise and support, particularly in the context of lifetime measurements of interlayer excitons. I also thank Ahmet Osman Ölçer for his assistance in constructing the room-temperature photoluminescence setup in our lab.

I thank Assoc. Prof. Dr. Talip Serkan Kasırga and the "Strongly Correlated Materials Lab" members for granting me access to their optical microscopy and dry transfer setup during my first year of studies. I want to thank Emre Ünal from "Demir Group" for conducting the reflectivity measurement of the planar cavities I fabricated. I thank all the technical and administrative staff from UNAM who helped me whenever I needed it and the training for the tools I used during my studies. I also want to thank Tübitak for funding my project through the TUBITAK-ARDEB 1001 and TUBITAK-BİDEB 2232 International Fellowship for Outstanding Researchers Program.

I want to thank my parents for instilling a deep appreciation for education. They have always supported my pursuit of higher education and encouraged me to strive toward becoming an independent and self-reliant woman. I express my gratitude to my mother, Serpil Cengiz. She is my best friend. She cried with me when I cried and laughed with me when I laughed. I genuinely love her. I am grateful to my father, Selim Atalay, who did his best to support me in every way. His encouragement and belief in me enabled me to pursue a career as an engineer. Little brother, Yusuf Ziya Atalay, I also love you. You are my favorite brother. Merve Mıntaş Küçük, I am thanking you in the family section; you know why, "my favorite person" and sister for life.

I am grateful to Assoc. Prof. Dr. Aylin Zıylan from Dokuz Eylül University for her unwavering support and inspiration. I vividly recall the day I nervously entered the Metallurgical and Materials Engineering department, hoping everything would fall into place. Her words, "It is never too late to start over," meant everything to me.

I am grateful that I am now an alumnus of İhsan Doğramacı Bilkent University. The university and its beautiful campus have been my home for the last three years, and I am grateful for the opportunity that the professors at UNAM have given me by accepting me into the program.

Per aspera ad astra.



Nomenclature

$0D$ Zero-dimensional

$1D$ One-dimensional

$2D$ Two-dimensional

$3D$ Three-dimensional

ϵ_0 Dielectric constant of the Vacuum

$\hbar$ Reduced Planck Constant

ARPS Angle-resolved Photoluminescence Spectrometer

c Speed of Light in a Vacuum

DBR Distributed Bragg Mirror

EIM Edge Identification Method

FIB Focused Ion Beam

FP Fabry-Pérot

H – stacking Near-hexagonal Stacking

hBN hexagonal Boron Nitride

HS Heterostructure

IEX Interlayer Exciton

$k_{\parallel}$	In-plane Momentum
M	Metal Atom
ML	Monolayer
MSL	Moire Superlattices
$PDMS$	Polydimethylsiloxane
$PECVD$	Plasma-enhanced Chemical Vapor Deposition
PL	Photoluminescence
$R - stacking$	Near-rhombohedral Stacking
SHG	Second Harmonic Generation
TEM	Transmission Electron Microscopy
TMD	Transition Metal Dichalcogenides
X	Chalcogen Atom

Contents

1	Introduction	1
2	Theoretical Background	4
2.1	Transition Metal Dichalcogenides	5
2.2	Intra- and Interlayer Excitons	14
2.3	Fabry-Pérot Microcavities	18
2.4	Exciton Polaritons	22
2.4.1	The Quantum Model of the Exciton-Polariton	27
3	Experimental Methods	30
3.1	Mechanical Exfoliation	31
3.2	Raman Spectroscopy and PL Measurement	33
3.2.1	WSe_2 Monolayer	33
3.2.2	$MoSe_2$ Monolayer	34
3.3	Stacking MLs with Dry Transfer Method	36

3.3.1	The Edge Identification Method	39
3.4	Fabrication and Characterization of The Fabry-Pérot Cavities . .	44
3.5	Optical Setups	54
3.5.1	Room Temperature Angle Resolved Photoluminescence Spectrometer	54
3.5.2	Micro PL Optical Setup	55
3.5.3	Time resolved Photoluminescence Spectroscopy	57
4	Results and Discussion	58
4.1	Results	59
4.1.1	Room Temperature PL Measurements	59
4.1.2	Low-Temperature PL Measurements	62
4.2	Discussion	73
A	Two Oscillator Model	85

List of Figures

2.1	(a) Image of a bulk MoS_2 crystal that is roughly 1 cm in length [1] and (b) Presented below is a three-dimensional schematic representation of a standard MX ₂ structure, where the chalcogen atoms (X) are depicted in yellow, and the metal atoms (M) are portrayed in grey [1].	5
2.2	Schematic diagrams depicting atomic structures for different phases of TMDs. (a) Shows bonding and stacking modes for five different phase structures. (b) Compares d-orbital splitting caused by spin-orbit coupling in 1H and 1T phases [2].	6
2.3	(a) Calculated band structures of (1) bulk MoS_2 , (2) quadrilayer MoS_2 , (3) bilayer MoS_2 , and (4) monolayer MoS_2 . The solid arrows indicate the lowest energy transitions Reprinted with permission from [3]. Copyright 2010 American Chemical Society., (b) PL spectra of MoS_2 ML and bilayer in the photon energy range from 1.3 to 2.2 eV. Reprinted with permission from [4] as follows: Kin Fai Mak, Changgu Lee, James Hone, Jie Shan, and Tony F. Heinz, Atomically Thin MoS_2 : A New Direct-Gap Semiconductor, 105, 136805, 2010. Copyright (2010) by the American Physical Society.	7
2.4	(a) Band structure of monolayer MoS_2 , $MoSe_2$, WS_2 and WSe_2 obtained from DFT calculations [5] and (b) DFT calculated bandgap energies of the monolayer TMDs [5]	8

2.5	Two $MoSe_2$ MLs, dark green triangle-shaped flakes, on silicon substrate pointed with arrows	9
2.6	PL of the (a) $MoSe_2$ and the (b) WSe_2 [6]	10
2.7	Raman Shift of the (a) WSe_2 and (b) $MoSe_2$ [6]	10
2.8	Step by step mechanical exfoliation of $MoSe_2$ with scotch tape . .	11
2.9	(a) Finding the MoS_2 flake's lattice direction by identifying the edge that has been cleaved at a 60° angle. The white scale bar is $2 \mu m$ (top), Finding the same MoS_2 flake's (top) lattice direction with the SHG method [7] (b)(1) MoS_2 monolayer flake is fracturing along armchair direction under the tension along zigzag direction. (2) MoS_2 monolayer flake is fracturing along a zigzag direction under the tension along the armchair direction. (3) MoS_2 monolayer flake is peeling off along the armchair direction. (4) MoS_2 monolayer flake is peeling off along zigzag direction [7].	12
2.10	Diagram showing the band structure at the K points' band edges. Reprinted with permission from [8] as follows: Di Xiao, Gui-Bin Liu, Wanxiang Feng, Xiaodong Xu, and Wang Yao, Coupled Spin and Valley Physics in Monolayers of MoS2 and Other Group-VI Dichalcogenides, 108, 196802, 2012. Copyright 2012 by the American Physical Society.	13
2.11	C_3 symmetry rotation centers as metal site (M), chalcogen cite (X) and hollow center created by M and X (h) of TMDs	13

- 2.12 (a) Schematic of electrons and holes forming excitons in real space in three-dimensional bulk and quasi-two-dimensional monolayers. Changes in the dielectric environment are indicated by varying dielectric constants (ϵ_{3D} and ϵ_{2D}) and vacuum permittivity (ϵ_0). (b) Illustrates the impact of dimensionality on electronic and excitonic properties. The dashed red line shows that transitioning from 3D to 2D is expected to increase both the band gap and exciton binding energy. Reprinted with permission from [9] as follows: Alexey Chernikov, Timothy C. Berkelbach, Heather M. Hill, Albert Rigosi, Yilei Li, Burak Aslan, David R. Reichman, Mark S. Hybertsen, and Tony F. Heinz, Exciton binding energy and non-hydrogenic Rydberg series in monolayer WS_2 , 113, 076802, 2014. Copyright 2014 by the American Physical Society. 14
- 2.13 Diagrammatic representation of a TMD heterostructure's type-II band alignment (bottom) and side-view structure (top) [10] . . . 15
- 2.14 The image shows near-R-stacked and near-H-stacked MSLs, consisting of electron-layer MX_2 and hole-layer $M'X'_2$. The M, X, M', and X' atoms are represented by green, magenta, blue, and gold colors. Purple and orange dashed diamond shapes indicate one supercell of the R- and H-stacked MSL [11]. Reproduced with permission from Springer Nature. 16
- 2.15 PL spectra of the MSL $MoSe_2/WSe_2$ heterostructure with a nonzero twist angle. (a) H-stacking $MoSe_2/WSe_2$ heterostructure with an optically bright spin-triplet state and spin-singlet interlayer exciton [12], (b) R-stacking $MoSe_2/WSe_2$ heterostructure with only optically bright spin-singlet state interlayer exciton [12]. Reprinted with permission from [12]. Copyright 2021 American Chemical Society. 17

2.16	Schematic representation of the Moiré trapping and de-trapping process of the IEX. Reprinted with permission from [12]. Copyright 2021 American Chemical Society.	18
2.17	(a)An example of FP Cavity with $SiO_2 - Si_3N_4$ alternating layers. Reproduced with permission from Springer Nature. (b)The reflectivity spectra depict the absence of a MoS_2 monolayer in a passive location (indicated by the black line) and the presence of a MoS_2 monolayer in an active location (indicated by the blue line). The vertical red dashed line denotes the energy associated with the MoS_2 exciton (exA).[13].Reproduced with permission from Springer Nature.	19
2.18	(a) The real image of the Plasma-Therm PECVD and (b)The schematic representation of the PECVD.	21
2.19	Full cavity with embedded quantum well [14]	23
2.20	Exciton-Polariton Energy vs In-Plane momentum graph representation	23
2.21	The schematic of the cavity photon coupled with TMD materials .	24
3.1	The scotch tape used for mechanical exfoliation	31
3.2	The $MoSe_2$ and the WSe_2 exfoliated crystals on scotch tape . . .	31
3.3	(a) Cutting a PDMS sheet with a scissor and (b) A piece of PDMS sheet on a slide.	32
3.4	WSe_2 ML on silicon substrate pointed by an arrow.	33

3.5	(a) Raman shift of the WSe_2 ML in figure 3.4, the laser power was 150 μ W, laser wavelength was 532 nm, and (b) Raman shift of the WSe_2 ML in figure 3.4, the laser power was 150 μ W, laser wavelength was 532 nm	34
3.6	$MoSe_2$ ML on PDMS substrate	34
3.7	(a) Raman shift of the $MoSe_2$ ML in figure 3.7, the laser power was 150 μ W, laser wavelength was 532 nm, and (b) PL spectrum of the $MoSe_2$ ML in figure 3.7, Lorentz fitted, the laser power was 150 μ W, laser wavelength was 532 nm	35
3.8	(a) Real image of the Dry Transfer Setup, and (b) The schematic representation of the Dry Transfer Setup	36
3.9	WSe_2 MLs on PDMS sheet	37
3.10	The meniscus of the PDMS during transfer	37
3.11	After transfer, WSe_2 flakes on silicon substrate	38
3.12	ML $MoSe_2$ on PDMS sheet	38
3.13	(a) HS labeled region is the heterostructure area, and (b) PL results of the heterostructure in (a)	39
3.14	$MoSe_2$ ML flake with identified zigzag edge	40
3.15	WSe_2 ML flake identified with zigzag edges	41
3.16	The area shown with dashed line is $MoSe_2$ flake on top of the few-layers hBN flake	41
3.17	HS closed with a thin layer of hBN flake shown with the red dashed line	42

3.18 HS closed with a thin layer of hBN flake	42
3.19 (a)HS image with dashed lines on bottom DBR, and (b) HS fabricated by utilizing the edge identification method on bottom DBR	43
3.20 (a, b, c, d) Other examples of HSs by utilizing Dry Transfer method	44
3.21 Bottom DBR	45
3.22 (a) The refractive index of the Si_3N_4 obtained with Ellipsometer, and (b) The refractive index of the SiO_2 obtained Ellipsometer.	45
3.23 The bottom DBR	46
3.24 Cross-section image of the bottom DBR obtained with FIB	46
3.25 TEM image of the DBR with scaled layers of thin films	47
3.26 The color difference between bottom DBRs fabricated with two different PECVD recipes	48
3.27 The TEM results of the second DBR, target thicknesses which were 150 nm for SiO_2 and 123 nm for Si_3N_4	49
3.28 The stopband of the full cavity, the DBR of this cavity is shown in figure 3.27.	49
3.29 (a) The DBR stop band, the resonating wavelength at 890 nm for low-temperature interlayer exciton measurements, and (b) The DBR stop band, the resonating wavelength at 958 nm for room-temperature interlayer exciton measurements	50
3.30 The bare DBR PL response	51

3.31	(a) The stopband of the full cavity fitted with Gauss function resonating at 890 nm approximately with FWHM at 23 nm, and (b) The stopband of the full cavity fitted with Gauss function resonating at 958 nm approximately with FWHM 20 nm	52
3.32	(a) SiO_2 thin film color change according to the thickness [15], and (b) Si_3N_4 thin film color change according to the thickness [15] .	53
3.33	The schematic of full cavity embedded with HS	53
3.34	(a) Room temperature optic PL setup ARPS, and (b) Schematic of ARPS optical setup	54
3.35	(a) Micro PL Optic Setup: i) The optical breadboard setup includes the white light source, laser source, and camera, ii) the spectrograph, iii) the cryostat (attoDRY 1000), iv) the vacuum tube, v) the window allowing light transmission, along with upper and lower cage plates and a microscope holder, vi) the objective lens and the sample holder with a nano-positioner [16] and (b) The schematic presentation of the micro PL setup	56
3.36	The time-resolved photoluminescence setup schematic.	57
4.1	(a) The $WSe_2 - MoSe_2$ HS with nearly 0° twist angle on bottom DBR, (b) The PL spectrum of the intralayer exciton region of the heterostructure in (a) at room temperature, and (c) The PL spectrum of the interlayer exciton region of the heterostructure in (a) at room temperature	59

4.2	(a) The $WSe_2 - MoSe_2$ HS with nearly 57° twist angle on bottom DBR, (b) The PL spectrum of the intralayer exciton region of the heterostructure in (a) at room temperature measured with home-built ARPS setup, (c) The PL spectrum of the interlayer exciton region of the heterostructure in (a) at room temperature measured with home-built ARPS setup	60
4.3	The ARPS measurements of the HS in figure 4.2 (a).	61
4.4	(a) The optical image of HS1 for low-temperature measurements and (b) The PL result of the interlayer exciton region of the HS1 in (a) at room temperature.	62
4.5	Intra- and interlayer region of the PL emission shown in 4.4 (a)	63
4.6	(a, b, c, d) The interlayer exciton peaks of the HS1 in figure 4.4 (a) obtained from different regions.	64
4.7	The temperature dependency of the PL emission of the HS1 in figure 4.6 (a) with the non-normalized PL intensity.	65
4.8	Temperature dependence of the PL emission of the HS1 in figure 4.6 (a)	66
4.9	(a) The PL emission of the HS1 in figure 4.6 (a) at 3.5 K, fitted with Lorentz function and (b) The splitting in the spin-singlet IEX peak in (a) as 3.5 K to 50 K.	67
4.10	The IEX measured in HS1 with different laser powers, the lines added as a reference to follow the blueshift.	68
4.11	The IEX peaks positions measured in HS1 vs. Laser power graph to observe the evolution of blueshift.	68
4.12	The optical microscope image of the second heterostructure.	69

4.13 (a) IEX PL Peak and (b,c,d) The Lifetime measurements of the spin-singlet IEX peak resonates around 1.39 eV.	70
4.14 (a) Lifetimes of IEX's slow and fast components in figure 4.13 (a) vs. pump power, (b) Slow time component of the lifetimes of the IEX vs. pump power and (c) Fast time component of the lifetimes of the IEX vs. pump power	71
4.15 (a, b) IEX region PL results with changing laser power obtained from different regions of the HS2 with visible Moiré trapped IEX.	72
A.1 Illustration of strong coupling, mechanical oscillators with masses m_i and spring constants k_i demonstrate frequency shifts and characteristic frequency splitting as a result of their coupling (κ) [17].	85
A.2 Eigenfrequencies of two uncoupled oscillators $\kappa=0$, masses are equal and spring constants are k_0 and $k_0+\Delta k$. (b) When coupling strength $\kappa=0.08k_0$ the anticrossing occurs [17]	87

Chapter 1

Introduction

Since Konstantin Novoselov and Andre Geim discovered obtaining graphene from graphite in 2004, [18], there has been considerable interest in 2D nanomaterials. But what is a nanomaterial? Nanomaterials can be categorized based on their sizes: zero-dimensional (0D), one-dimensional (1D), two-dimensional (2D), and three-dimensional (3D) nanomaterials [19]. For a material to be classified as a nanomaterial, at least one of its dimensions should fall within the nanoscale range of 1-100 nm. In this thesis, the 2D nanomaterials are used as the quantum well.

An essential scientific database search is enough to reveal the widespread research interest in 2D nanomaterials. The Google Scholar database for academic publishings shows around 2.3 million results for graphene alone and 5.4 million for two-dimensional materials. Moreover, there are various 2D materials besides graphene, such as black phosphorus, hexagonal boron nitride (hBN), and transition metal dichalcogenides (TMD). Because of the lack of bandgap in graphene, TMDs gain attention, which possess direct band gaps in their monolayer form [3, 20].

Transition-metal dichalcogenides (TMDs) are a fascinating class of two-dimensional materials that have attracted the attention of scientists and researchers because of their superior mechanical, optical, and electronic properties. TMDs exhibit intriguing characteristics, as their indirect to direct band gap transition from bulk to monolayer (ML) form, making them ideal for studying multiple physical phenomena, especially the photophysics of intra- and interlayer excitons and light-matter interactions [1, 3, 4, 13, 21, 22].

Due to the spatial in-plane confinement, the transition from indirect bandgap to direct bandgap occurs in TMD monolayers. The spatial confinement changes the dielectric environment, resulting in reduced Coulombic screening. Consequently, the binding energy of excitons and the bandgap of the TMD monolayers increase [9]. This phenomenon causes the formation of intralayer excitons in TMD monolayers. Moreover, by stacking the monolayer of TMDs vertically, it is possible to achieve type-II band alignment; hence, the interlayer excitons form between the conduction band minimum and valence band maximum of the two monolayers in heterobilayers. As a result, TMDs serve as valuable tools for studying the photophysics of intra- and interlayer excitons.

Studying the photo-physics of excitons within TMDs became especially significant due to their superior optoelectronic properties, mainly because of the high oscillator strength of the TMD excitons. These materials provide an excellent platform for investigating the behavior of intra- and interlayer exciton-polaritons, which are hybrid particles arising from the coupling of electromagnetic waves, the cavity photon, with electric dipoles, the excitons in a material.

Exciton-polaritons, quasi-particles arising from coupling excitons and photons in a microcavity, have collected significant interest in the scientific community due to their potential applications and unique behavior. The research on exciton-polaritons contributes to our understanding of Bose-Einstein condensation and superfluidity and holds promise for practical applications such as polariton switches and polariton lasers. These developments can potentially revolutionize various fields, from fundamental physics to advanced optoelectronic devices.

Our research primarily investigates the photophysical characteristics of interlayer excitons in $MoSe_2 - WSe_2$ heterostructures confined within planar microcavities and interlayer exciton-polaritons. This study predominantly relies on experimental methodologies, utilizing advanced microfabrication techniques to facilitate the investigation. In addition, we are employing essential techniques for the fabrication of TMD heterostructures, including edge identification and the dry transfer method, which play critical roles in ensuring the quality of the heterostructures under examination.

Chapter 2 contains brief information about the properties of TMDs, including the intra- and interlayer excitons form in TMDs by photo-excitation and a short description of the exciton-polaritons. Chapter 3 heavily focuses on the experimental part of our study; the methodology and the characterization of the devices are explained comprehensively. Finally, chapter 4 covers the results at both cryogenic and room temperatures, mainly focusing on the photoluminescence of interlayer excitons, time-resolved photoluminescence measurements of the interlayer excitons, and analyzing the findings to look for the signs of the formation of interlayer exciton-polaritons.

Chapter 2

Theoretical Background

In this chapter, the essential information about the theoretical background of TMDs and their heterostructures, the intra- and interlayer excitons forms in TMD monolayers and their heterostructures, and the exciton-polaritons of the TMDs coupled with optical microcavities are presented. Specifically, the TMDs chapter briefly discusses the molecular composition, phases, the direct band transition from bulk to monolayer, the mechanical exfoliation of the monolayers, the edge identification method, and the symmetry groups of TMDs. In the intra- and interlayer chapter, the band gap transition from monolayer to bulk is explained in detail, and the formation of type-II band alignment in the heterostructures of TMDs and their crystal structure, depending on the twist angle is also described in detail. The FP cavities are briefly explained, and exciton-polaritons in TMDs are elaborated with the quantum model of exciton-polaritons.

2.1 Transition Metal Dichalcogenides

Since the discovery of the exfoliation of graphene by Konstantin Novoselov and Andre Geim [18], two-dimensional (2D) materials have gained attention because of their exotic properties associated with their electronic properties and their potential applications in electronics, optoelectronics, sensors, and so on. The pristine graphene is unsuitable for electronic applications due to its extraordinary band gap. As a result, other two-dimensional materials, such as transition metal dichalcogenides (TMDs), have received much attention. You can see the bulk form of MoS_2 crystal in figure 2.1 (a).

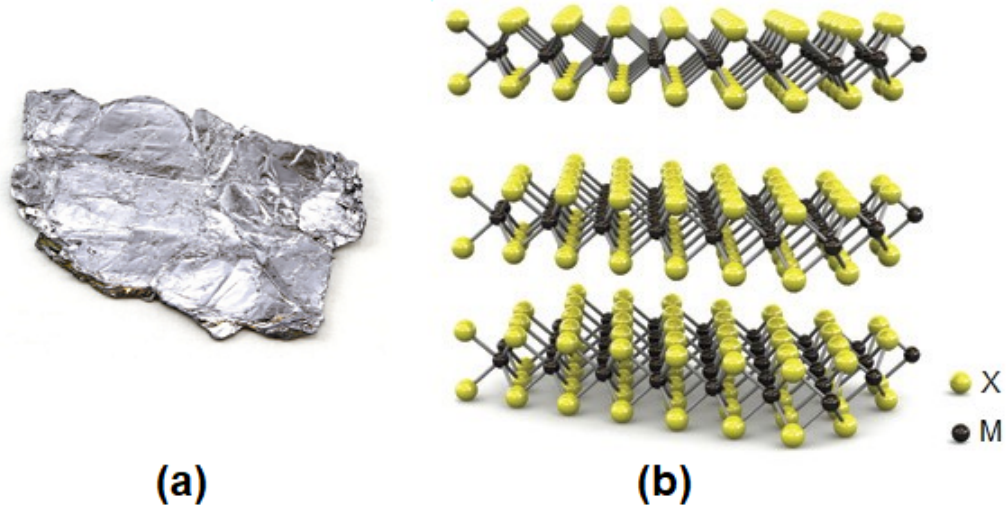


Figure 2.1: (a) Image of a bulk MoS_2 crystal that is roughly 1 cm in length [1] and (b) Presented below is a three-dimensional schematic representation of a standard MX_2 structure, where the chalcogen atoms (X) are depicted in yellow, and the metal atoms (M) are portrayed in grey [1].

TMDs are also van der Waals 2D materials with MX_2 stoichiometry where M is the transition metal atom, such as tungsten and molybdenum, and X is the chalcogen atom, such as selenium and sulfur. TMDs have hexagonal lattices, and the bond between the transition metal and chalcogen atoms is covalent and in-plane. There are van der Waals bonds, which are weak out-of-plane interactions between layers [1], see figure 2.1 (b). Moreover, TMD monolayers do not have inversion symmetry, which causes spin-orbit coupling to arise in TMD monolayers. The spin-orbit coupling removes spin degeneracy that causes spin-splitting, making them suitable for spintronic applications as well [23, 5].

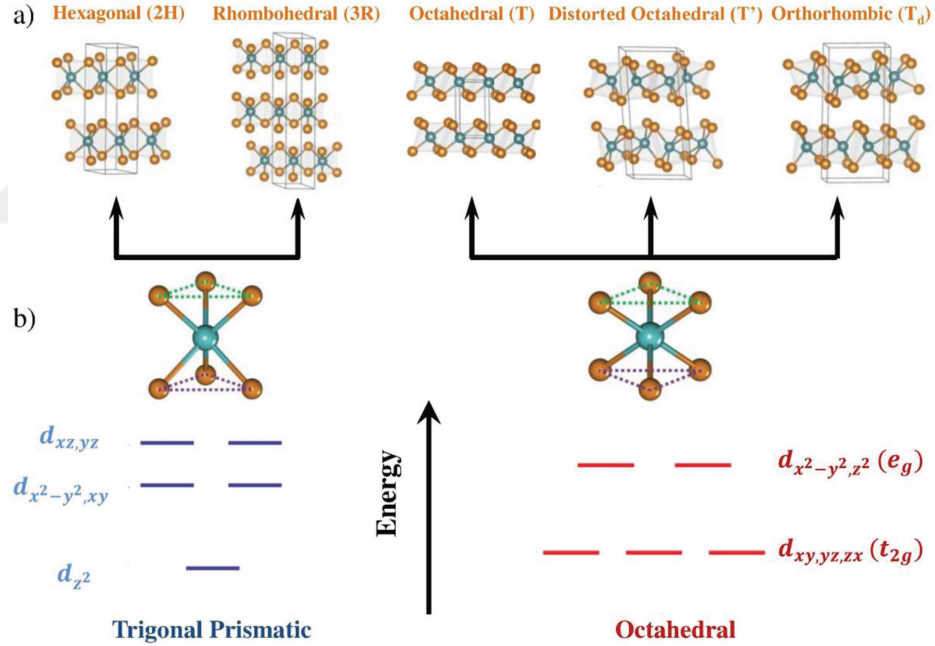


Figure 2.2: Schematic diagrams depicting atomic structures for different phases of TMDs. (a) Shows bonding and stacking modes for five different phase structures. (b) Compares d-orbital splitting caused by spin-orbit coupling in 1H and 1T phases [2].

In TMDs, there can be five different phases, which are 2H (1H when single layer), 3R, 1T, 1T', and T_d . [2] 2H and 3R phases are trigonal prismatic phases, meaning hexagonal symmetry, and they belong to the D_{3h} group [24]. To summarize, as we are working with single-layer TMDs, they have hexagonal symmetry (1H). You can see the atomic structures of the phases in figure 2.2.

In the 1H phase, which is a trigonal prismatic structure, d orbitals of the transition metal are split into three degenerated states which are two degenerated orbital of $d_{xy,yz}$, two degenerated orbital of $d_{x^2-y^2,xy}$ and d_{z^2} orbital. However, the T phase is an octahedral structure with two degenerate states as $d_{x^2-y^2,z^2}$ (e_g) and $d_{xy,yz,zx}$ (t_{2g}) [2, 24], see figure 2.2.

Bulk TMDs have indirect electronic band gaps but a direct band gap in their monolayer form; hence, the radiative recombination of electrons and holes is more efficient in direct bandgap TMD semiconductors [1]. In the figure 2.3 (a), the bandgap transformation of MoS_2 with the change in thickness [3] can be seen. Since the ML MoS_2 have direct bandgap, in figure 2.3 (b) the difference between monolayer MoS_2 and bilayer MoS_2 photoluminescence (PL) spectrum [4] can be seen.

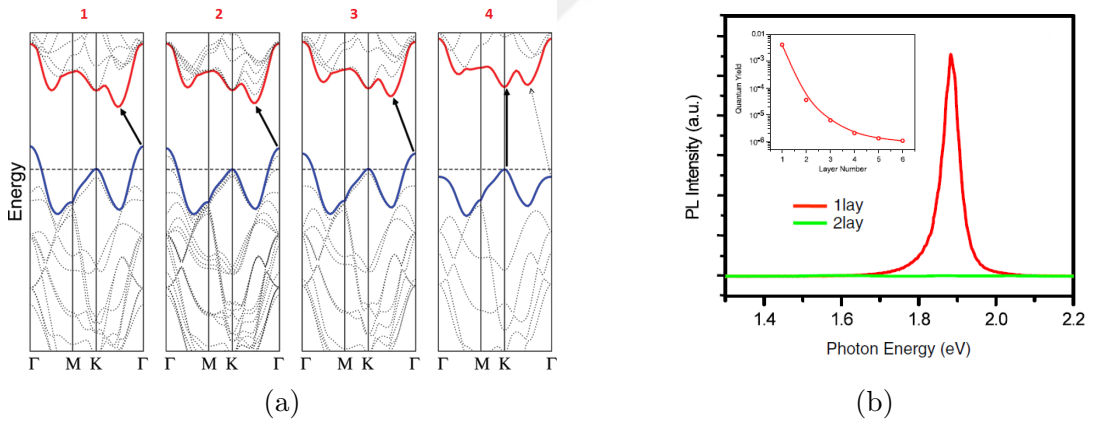


Figure 2.3: (a) Calculated band structures of (1) bulk MoS_2 , (2) quadrilayer MoS_2 , (3) bilayer MoS_2 , and (4) monolayer MoS_2 . The solid arrows indicate the lowest energy transitions Reprinted with permission from [3]. Copyright 2010 American Chemical Society., (b) PL spectra of MoS_2 ML and bilayer in the photon energy range from 1.3 to 2.2 eV. Reprinted with permission from [4] as follows: Kin Fai Mak, Changgu Lee, James Hone, Jie Shan, and Tony F. Heinz, Atomically Thin MoS_2 : A New Direct-Gap Semiconductor, 105, 136805, 2010. Copyright (2010) by the American Physical Society.

The DFT calculations of the bandgap of the MoS_2 , $MoSe_2$, WS_2 , and WSe_2 monolayers are shown in figure 2.4 (a); the table shows the calculated bandgap energies of the monolayer of the TMDs materials with the DFT method [5] in figure 2.4 (b).

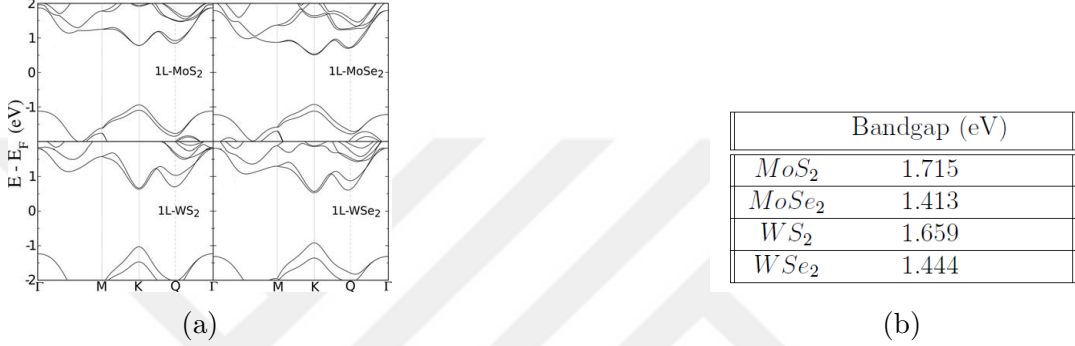


Figure 2.4: (a) Band structure of monolayer MoS_2 , $MoSe_2$, WS_2 and WSe_2 obtained from DFT calculations [5] and (b) DFT calculated bandgap energies of the monolayer TMDs [5]

For the characterization of MLs, optical microscopy, Raman spectroscopy, and PL spectroscopy are used to differentiate the ML TMD flakes from the few-layer TMD flakes. By optical microscopy, MLs can be distinguished by analyzing the contrast of the flakes. However, this method is not always accurate. Therefore, Raman and PL spectroscopy must be done for those potential MLs. Raman Spectroscopy is a method based on the materials' lattice vibrations. By analyzing the vibrational mode signals, the TMDs can be characterized whether they are single-layer, multi-layer, or bulk because these vibrational modes' intensity, width, and energy are influenced by the layer number of the TMDs [6]. For example, the B modes are inactive in the WSe_2 monolayer. Hence, the ML WSe_2 does not have a signal around 310 cm^{-1} . An example of ML $MoSe_2$ flake can be seen in figure 2.5.

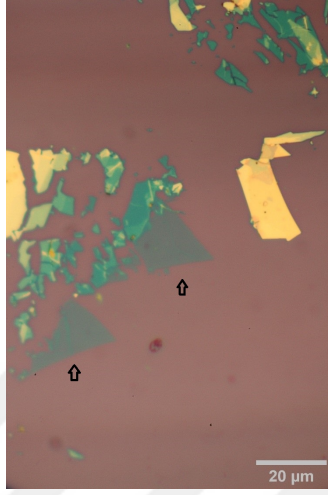


Figure 2.5: Two $MoSe_2$ MLs, dark green triangle-shaped flakes, on silicon substrate pointed with arrows

The vibrational modes of the flakes are investigated via Raman Spectroscopy to determine the layer number of the TMD flakes. Moreover, the photoluminescence of the TMD flakes is also measured for further analysis. As mentioned before, the monolayer TMDs have a direct band gap, and their PL peaks are a characteristic property for each. PL spectra and Raman shifts of WSe_2 and $MoSe_2$ MLs can be seen in figures 2.6 and 2.7. As seen in figure 2.6, ML $MoSe_2$ has a photoluminescence peak at around 790 nm, and WSe_2 ML has a peak at around 755 nm [6].

As you can see in the figure 2.7, A_{1g} and E_{2g}^1 Raman modes of the ML WSe_2 are overlapped at around 250 cm^{-1} . Also, the ML WSe_2 does not have any peaks as B_{2g}^1 mode around 310 cm^{-1} . A_{1g} and E_{2g}^1 Raman modes of the ML $MoSe_2$ are at around 240 cm^{-1} and 290 cm^{-1} , respectively. Also, ML $MoSe_2$ has a peak as B_{2g}^1 mode around 360 cm^{-1} [6].

There are several synthesis methods for TMD monolayers: mechanical exfoliation, liquid phase exfoliation, chemical vapor deposition, and molecular beam epitaxy [25]. Mechanical exfoliation is also known as the scotch-tape method; it aims to mechanically cleave the layers of MLs from bulk layered materials [26].

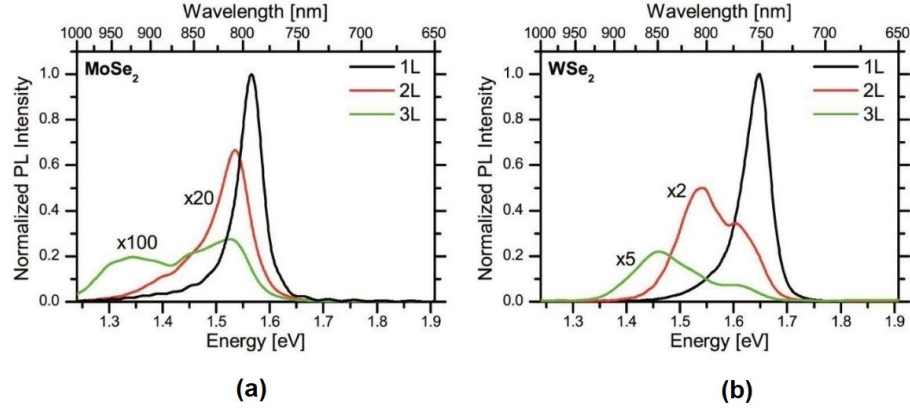


Figure 2.6: PL of the (a) MoSe_2 and the (b) WSe_2 [6]

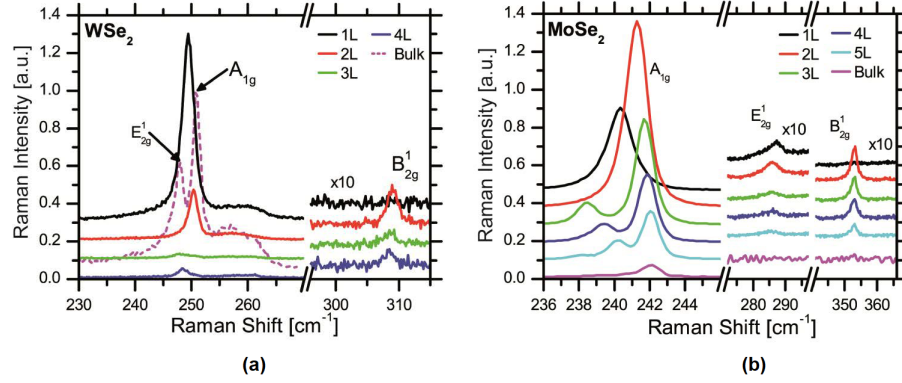


Figure 2.7: Raman Shift of the (a) WSe_2 and (b) MoSe_2 [6]

The demonstration of the mechanical exfoliation of the MoSe_2 can be seen in figure 2.8.

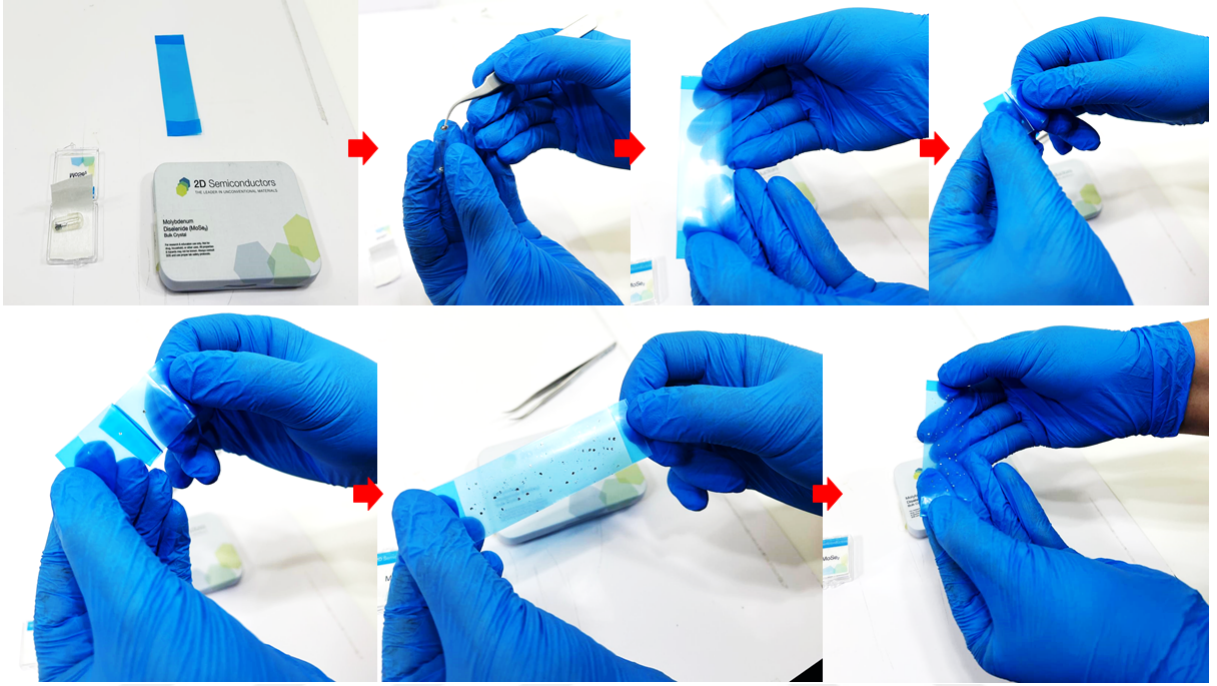


Figure 2.8: Step by step mechanical exfoliation of $MoSe_2$ with scotch tape

Second harmonic generation (SHG) and transition electron microscopy can be used to determine the ML flakes' crystallographic orientation. However, there is a statistical approach called the edge identification method to determine the in-plane cleavage behavior of the TMDs. Hence, the orientation of the edges of the materials [7].

As seen in figure 2.9, without the SHG, the edge of the flakes' orientation, whether an armchair or zigzag, can be identified with this statistical approach. If the angle between two flake edges is 60° or it multiplies, the possibility of a zigzag configuration will increase. Moreover, in the same research [7], they show that the clean edges are more likely zigzag edges by molecular dynamics simulations, as shown in figure 2.9.

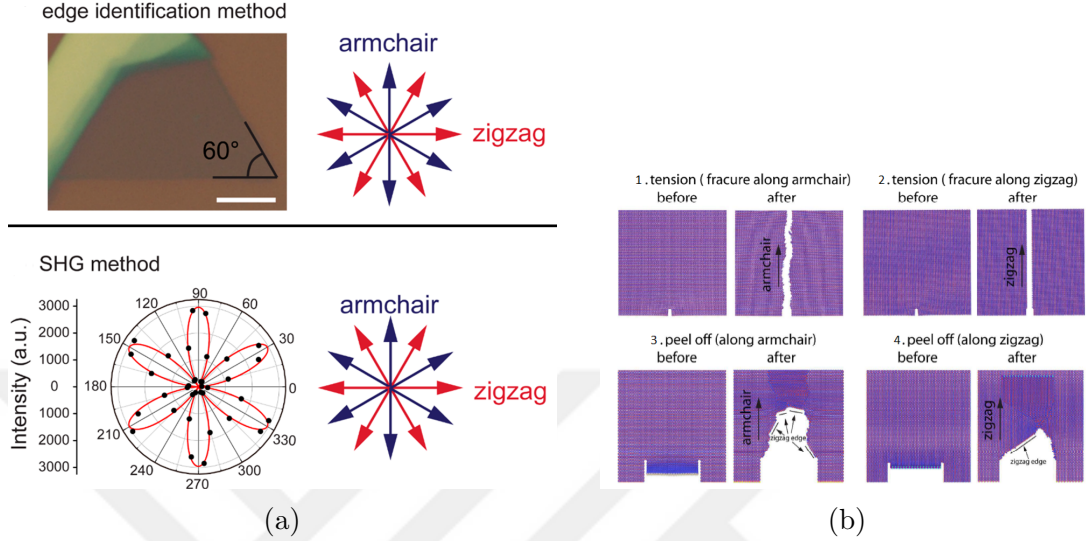


Figure 2.9: (a) Finding the MoS_2 flake's lattice direction by identifying the edge that has been cleaved at a 60° angle. The white scale bar is 2 μm (top), Finding the same MoS_2 flake's (top) lattice direction with the SHG method [7] (b)(1) MoS_2 monolayer flake is fracturing along armchair direction under the tension along zigzag direction. (2) MoS_2 monolayer flake is fracturing along a zigzag direction under the tension along the armchair direction. (3) MoS_2 monolayer flake is peeling off along the armchair direction. (4) MoS_2 monolayer flake is peeling off along zigzag direction [7].

Identifying the zigzag edges is essential because, during the stacking process of MLs to make heterostructures, the zigzag edges of the MLs should be aligned with the consideration of the angle which should close to magic twist angles 0 and 60° between the edges [7]. The aim here is to align the K and K' valleys (corners of the Brillouin zones of the materials, see figure 2.10) so that momentum mismatch can be avoided as much as possible and exciton formation in the type-II band alignment can occur more efficiently [23, 27, 8]. With different twist angles other than 0 or 60°, the valence band maximum and conduction band minimum may shift to the Γ/Λ (or Q) valleys; hence, due to significant interlayer hybridization, an indirect band gap can be formed [10, 28].

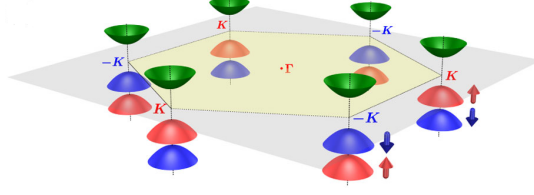


Figure 2.10: Diagram showing the band structure at the K points' band edges. Reprinted with permission from [8] as follows: Di Xiao, Gui-Bin Liu, Wanxiang Feng, Xiaodong Xu, and Wang Yao, Coupled Spin and Valley Physics in Monolayers of MoS2 and Other Group-VI Dichalcogenides, 108, 196802, 2012. Copyright 2012 by the American Physical Society.

Moreover, TMD monolayers have in-plane two π three rotational (C_3) symmetry with three rotation centers as metal site (M), chalcogen sites (X), and hollow center created by M and X (h) [29], figure 2.11 shows the schematic representation of the symmetry rotation centers. Because of these symmetry centers, the heterostructures of TMDs can have different types of lattice orientations.

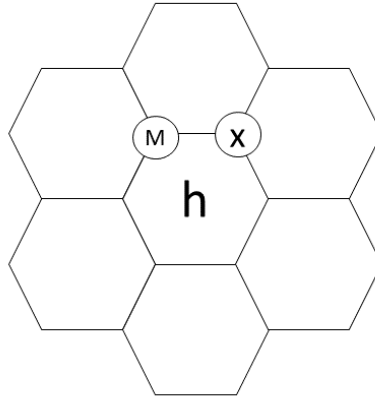


Figure 2.11: C_3 symmetry rotation centers as metal site (M), chalcogen site (X) and hollow center created by M and X (h) of TMDs

2.2 Intra- and Interlayer Excitons

An exciton is a hydrogen-like Coulomb-bound electron-hole pair [30, 31]. The excitons that form in the same layer are called intralayer excitons in TMD monolayers. Since the dimensionality modifies the bandgap of the van der Waals TMD materials, intralayer excitons in monolayers have a direct band gap caused by the in-plane confinement in the material [9, 32]. Hence, the Coulombic screening decreases because of the change in the dielectric environment. That results in higher exciton binding energies and an increase in the bandgap as well, see figure 2.12 (b); for example, the intralayer exciton of WS_2 in its ML form has a binding energy of 320 meV yet in bulk form the binding energy of the excitons is 50 meV[9].

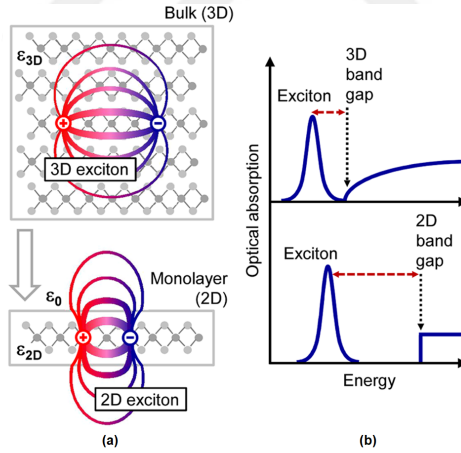


Figure 2.12: (a) Schematic of electrons and holes forming excitons in real space in three-dimensional bulk and quasi-two-dimensional monolayers. Changes in the dielectric environment are indicated by varying dielectric constants (ϵ_{3D} and ϵ_{2D}) and vacuum permittivity (ϵ_0). (b) Illustrates the impact of dimensionality on electronic and excitonic properties. The dashed red line shows that transitioning from 3D to 2D is expected to increase both the band gap and exciton binding energy. Reprinted with permission from [9] as follows: Alexey Chernikov, Timothy C. Berkelbach, Heather M. Hill, Albert Rigosi, Yilei Li, Burak Aslan, David R. Reichman, Mark S. Hybertsen, and Tony F. Heinz, Exciton binding energy and nonhydrogenic Rydberg series in monolayer WS_2 , 113, 076802, 2014. Copyright 2014 by the American Physical Society.

In TMD, monolayers and their heterostructures, intralayer excitons, and interlayer excitons can be observed. When the hole and the electron are formed in the same material layer, it is called an intralayer exciton; however, when the hole and the electron are located in different layers, the exciton is called interlayer. Interlayer excitons can be formed in the heterostructures of these monolayer TMDs. TMD excitons have strong oscillator strength and high exciton binding energy [33].

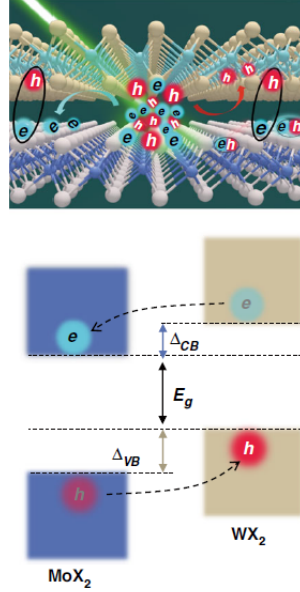


Figure 2.13: Diagrammatic representation of a TMD heterostructure's type-II band alignment (bottom) and side-view structure (top) [10]

Monolayer TMD heterostructures can be stacked vertically to observe interlayer excitons. Heterostructures can have three types of band alignments: straddling/type-I, staggered/type-II, and broken/type-III [10, 34, 35]. In TMDs, the theoretical and the experimental studies showed that the type-II band alignment occurs between conduction band minimum and valence band maximum [10]. In figure 2.13, a schematic of a type-II band alignment can be seen. When the charge transfer happens in the conduction band minimum and valence band maximum placed in opposite monolayers, the interlayer exciton formation can be observed in TMD heterostructures. Interlayer excitons have longer lifetimes than

intralayer excitons; because of the spatial separation of the electron and the hole, the wavefunctions of the hole and exciton overlap decrease [10, 35].

The Moiré superlattices (MSL) can be formed in TMD heterostructures due to a slight twist angle between the layers and the lattice mismatch. There are two types of stacking: near rhombohedral stacking (R-stacking) and near-hexagonal stacking (H-stacking). Multiple high-symmetry points can be identified in both stacking orders, as illustrated in Figure 2.11 [11].

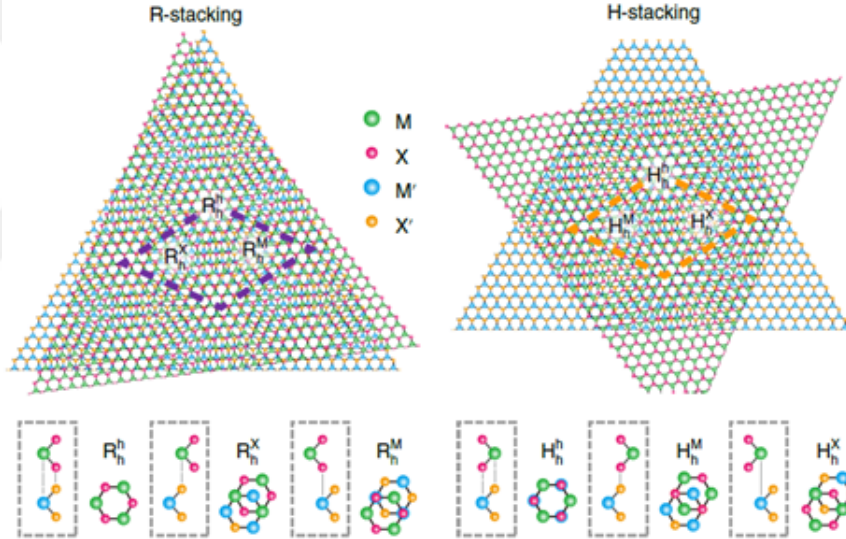


Figure 2.14: The image shows near-R-stacked and near-H-stacked MSLs, consisting of electron-layer MX_2 and hole-layer $M'X'_2$. The M, X, M', and X' atoms are represented by green, magenta, blue, and gold colors. Purple and orange dashed diamond shapes indicate one supercell of the R- and H-stacked MSL [11]. Reproduced with permission from Springer Nature.

These high symmetry points modify the optical selection rules in these MSLs [29, 11]. Since TMDs do not have inversion symmetry, spin-orbit coupling lifts the spin degeneracy; hence, the spin splitting occurs[23, 5].

Interlayer excitons formed in the same valley have suppressed exchange interactions and valley depolarization because of their spatially separate nature [36]. Hence, they can form spin singlet and triplet states at the H_h^h high symmetry points because of the spin-orbit coupling caused by transition metal (in the $MoSe_2/WSe_2$ heterostructures caused by Molybdenum [12]), subsequently undergoing radiative recombination (optically bright)[36, 37].

Since the twist angle between two MLs changes, the stacking changes, hence the high symmetry points. In the case of the $MoSe_2/WSe_2$ heterostructure, when the twist angle is close to 0° , the heterostructure becomes an R-stack when it is close to 60° , it becomes an H-stack. In H-stack, the spin-triplet state of the interlayer exciton is also bright with the spin-singlet state; however, in R-stack, only the singlet state is bright [12, 38].

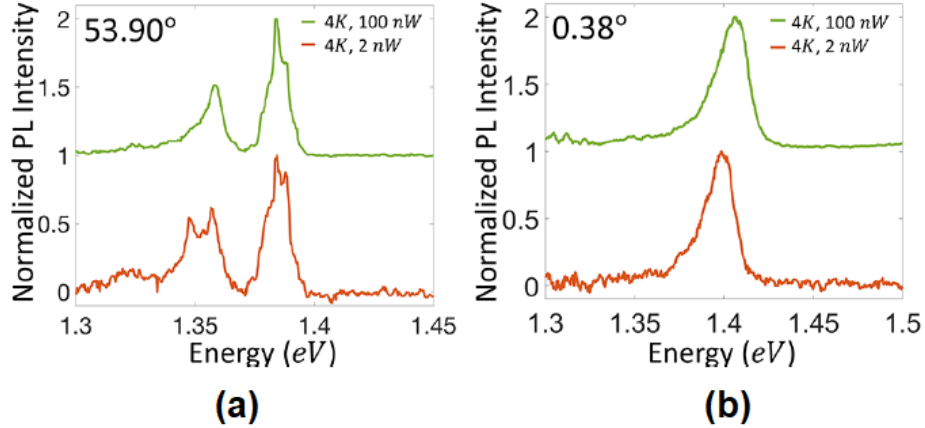


Figure 2.15: PL spectra of the MSL $MoSe_2/WSe_2$ heterostructure with a nonzero twist angle.(a) H-stacking $MoSe_2/WSe_2$ heterostructure with an optically bright spin-triplet state and spin-singlet interlayer exciton [12], (b) R-stacking $MoSe_2/WSe_2$ heterostructure with only optically bright spin-singlet state interlayer exciton [12]. Reprinted with permission from [12]. Copyright 2021 American Chemical Society.

During the stacking process of the MLs of TMDs, because of the inevitable lattice mismatch and twist angle, Moiré trapping sites can occur [12, 39]. Moiré trapping causes localization of the IEXs by modifying the band gap, creating

minibands [12, 39]. At the low-temperature excitation with low pump power, these localized IEXs can be observed [40]. Figure 2.16 shows the schematic representation of the Moiré trapping sites. When the mismatch angle is very close to zero, for instance, 0.15 degrees, these Moiré traps can be very deep; hence, they become observable even at room temperature [12]. Moreover, deep Moiré potentials like ≈ 100 meV can also localize intralayer excitons and trions [39].

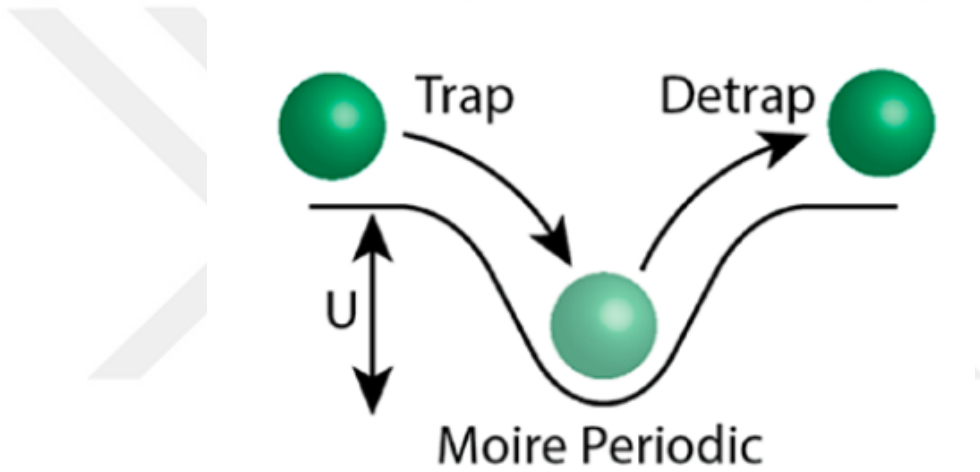


Figure 2.16: Schematic representation of the Moiré trapping and de-trapping process of the IEX. Reprinted with permission from [12]. Copyright 2021 American Chemical Society.

2.3 Fabry-Pérot Microcavities

An optical cavity is a resonator, a device that resonates only at specific frequencies for electromagnetic radiation. Those specific frequencies are called resonator modes or cavity modes of the cavity [41]. Different kinds of cavities exist, such as planar cavities, photonic crystals, microdisk cavities, etc. [42]. In our study, a FP cavity, a planar cavity, is used.

FP cavities consist of two distributed Bragg mirrors (DBRs), which are made out of $\lambda/4$ thick alternating layers of dielectric thin films with different refractive

indices, see figure 2.17 [43]. Where λ is the cavity's resonance frequency. Hence, they can be fabricated with desired properties, such as cavity modes. In the cavity mode, the reflectivity of the full cavity reaches its lowest value. The reason is because of constructive interference between the light reflected from all the interfaces of the layers within the cavity. As a result, the cavity reflects all wavelengths of light near the cavity mode [44].

Figure 2.17 (a) shows an example of a FP cavity. It has 8.5 periods on the bottom DBR and 7.5 on the top DBR. The period refers to the thin films with high and low refractive indices. The silicon dioxide layer in the middle is called the inner cavity. Figure 2.17 (b) shows the reflectivity of the full cavity, the black curve without the quantum well-embedded response, and the blue curve shows the quantum well-embedded response as seen there is an absorbance at the cavity mode.

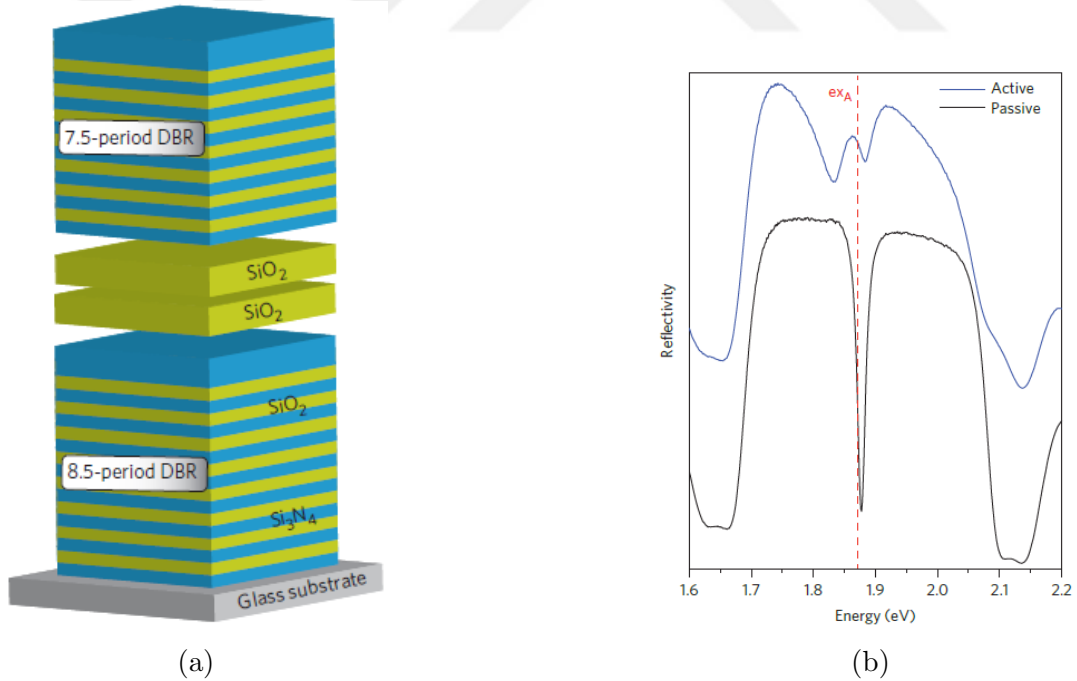


Figure 2.17: (a) An example of FP Cavity with $SiO_2 - Si_3N_4$ alternating layers. Reproduced with permission from Springer Nature. (b) The reflectivity spectra depict the absence of a MoS_2 monolayer in a passive location (indicated by the black line) and the presence of a MoS_2 monolayer in an active location (indicated by the blue line). The vertical red dashed line denotes the energy associated with the MoS_2 exciton (exA). [13]. Reproduced with permission from Springer Nature.

The cavity photons' loss can be interpreted using a cavity's quality factor. If the quality factor is high, the loss of cavity photons will decrease [45].

$$Q = \frac{w_0}{\Delta w} \quad (2.1)$$

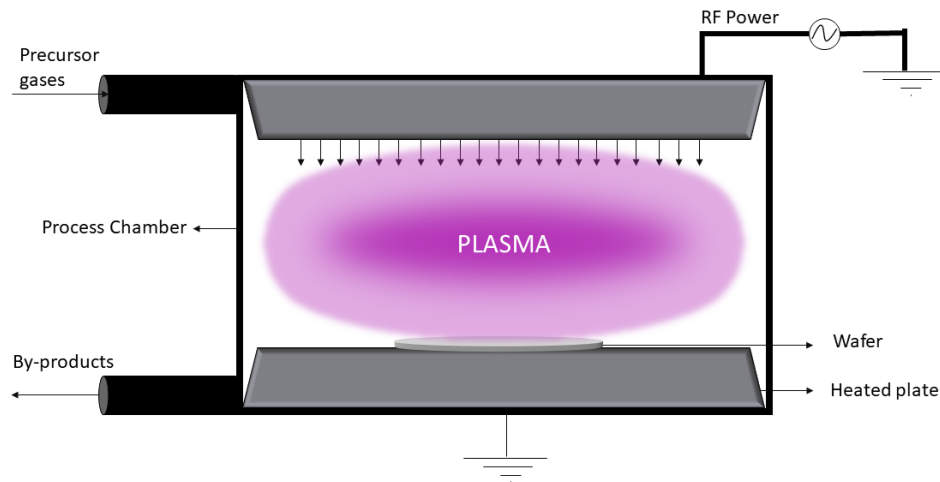
Where w_0 is the resonance wavelength of the full cavity equals 1.39eV, which is specifically for $MoSe_2 - WSe_2$ IEX emitter, and Δw is the full-width half maximum of the cavity mode reflectivity signal [45].

Plasma-enhanced chemical vapor deposition (PECVD) can be used to deposit the thin films; see figure 2.18 (a). The schematic representation of the PECVD can be seen in figure 2.18 (b) [46]. Plasma is created with a ground electrode and radio frequency (RF) powered electrode in PECVD [47]. The main difference from regular CVD is that the PECVD can operate at lower temperatures because of the effect of the plasma [47].

The alternating layers of thin films with different refractive indices are deposited onto the silicon wafer layer by layer to fabricate the bottom DBR first. After the fabrication of the bottom DBR, the quantum well is placed on top of the DBR, and the top DBR is fabricated using the same method.



(a)



(b)

Figure 2.18: (a) The real image of the Plasma-Therm PECVD and (b) The schematic representation of the PECVD.

2.4 Exciton Polaritons

An exciton is a dipole formed between an electron and a hole pair; therefore, it interacts with the electromagnetic fields of light [48]. This interaction can be evaluated by looking at the oscillator strength of the exciton [48]. The oscillator strength, f , is calculated as follows,

$$f = \frac{2m_{ex}w}{\hbar} |\langle u_v | r \cdot e | u_c \rangle|^2 \frac{V}{\pi a_B^3} \quad (2.2)$$

where w is the angular frequency of the electromagnetic field, m_{ex} is the effective mass of the exciton, $|u_c\rangle$ and $|u_v\rangle$ are the Bloch functions of the electron and hole, a_B is the Bohr radius of the exciton, and V is the quantized volume. The position operator " r " acts on the electromagnetic field, " e ," representing the transition of the dipole of electron and hole [48].

The enhancement of the interaction because of the confinement is interpreted with the term $\frac{V}{\pi a_B^3}$. The term shows that the exciton has increased electron-hole interaction rather than an unbound electron-hole pair [48].

The confined quantum well exciton has a smaller Bohr radius and higher binding energy; therefore, its oscillator strength is also higher than unbound electron-hole pairs; however, to efficiently couple this exciton to the electromagnetic field, the electromagnetic field should also be confined because its longer transverse coherence length. That is why microcavities must be used to confine light in z direction [48]. Moreover, the light propagates in free space and has a zero rest mass with linear dispersion. Yet, when the cavity confines light, it shows a parabolic dispersion relation with an effective mass m [49].

A photon and an exciton in a microcavity can be considered two coupled harmonic oscillators; a schematic representation can be seen in figure 2.19 [14, 50]. In a microcavity, the cavity photon and the exciton oscillate independently or form a new particle called an exciton-polariton in a strong coupling regime [51]. Strong coupling occurs between the photon and the exciton when the exchange

of light and matter energy is faster than their decay and dephasing rates[52, 53]. The two coupled oscillator model can explain the hybridization of these two new states [17].

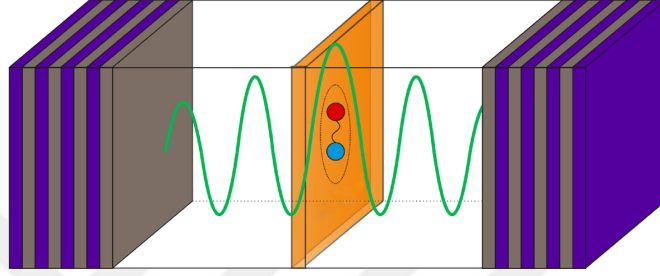


Figure 2.19: Full cavity with embedded quantum well [14]

In exciton-polariton, these two eigenfrequencies are called the upper and lower polariton branches, as shown in figure 2.20. Hence, two new eigenfrequencies occur proportionally to the coupling strength between the exciton and the cavity photon. These branches exhibit characteristics of both excitons and photons due to the hybridization of light and matter states [54].

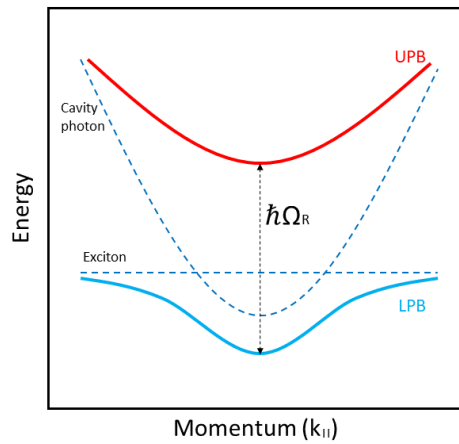


Figure 2.20: Exciton-Polariton Energy vs In-Plane momentum graph representation

The energy difference between the upper and lower polariton branches is called the Rabi splitting energy, and this phenomenon indicates that the system is in a strong coupling regime. Because excitons and photons are bosons, polaritons are also classified as bosons, known explicitly as composite bosons [51].

The system's angle-resolved photoluminescence or reflectivity spectrum is measured to determine the energies of the new eigenstates by analyzing the energy dispersion relation vs momentum of the cavity photons. The photon and exciton wavevectors are equal during the coupling, $k=k'$. Since the microcavity confines the out-of-plane momentum and the photon field in the z-direction, changes in the in-plane momentum can be detected using angle-resolved techniques. The reason is that the cavity photon's momentum is related to its angle, revealing exciton-polariton formation. This alteration in-plane momentum contributes to the total momentum, allowing the calculation of the energy of the upper and lower polariton branches. In this way, the dispersion relation of the polariton can be observed.

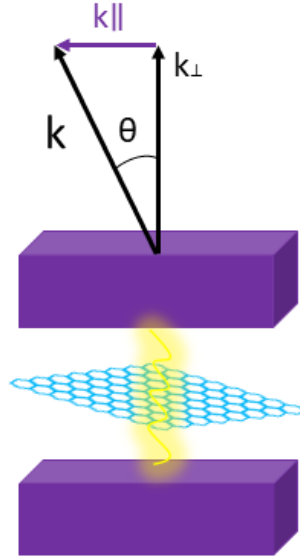


Figure 2.21: The schematic of the cavity photon coupled with TMD materials

The coupled cavity photon and TMD quantum well with the momentum of cavity photon are represented in figure 2.21. The momentum of the cavity photon can be calculated with the equation 2.3.

$$k_{total} = \sqrt{k_{\parallel}^2 + k_{\perp}^2} \quad (2.3)$$

The in-plane momentum ($\mathbf{k}_{\parallel}$) is minimal compared to the out-of-plane momentum, ($k_{\perp}$) $\mathbf{k}_{\parallel} \ll k_{\perp}$, Taylor series expansion can be used to approximate the photon's energy. The equation 2.4 shows the calculation of the cavity photon energy, where $\hbar$ is the reduced Planck constant, n_c is the refractive index of the cavity, and c is the speed of light in vacuum [48].

$$E_{cav}(k) = \frac{\hbar c}{n_c} \sqrt{k_{\parallel}^2 + k_{\perp}^2} \quad (2.4)$$

When factor out $k_{\perp}$ in the equation 2.4 to simplify, it becomes as the following equation 2.5 [48],

$$E_{cav}(k) = \frac{\hbar c}{n_c} k_{\perp} \sqrt{1 + \left(\frac{k_{\parallel}}{k_{\perp}}\right)^2} \quad (2.5)$$

Since $\mathbf{k}_{\parallel} \ll k_{\perp}$, the term $\left(\frac{\mathbf{k}_{\parallel}}{k_{\perp}}\right)^2$ is very small, and Taylor series expansion for $\sqrt{1+x}$ when x is very small can be calculated as follows,

$$\sqrt{1+x} \approx 1 + \frac{x}{2} \quad \text{for } |x| \ll 1 \quad (2.6)$$

When $x = \left(\frac{\mathbf{k}_{\parallel}}{k_{\perp}}\right)^2$. The following terms become approximately equal to each other [48].

$$\sqrt{1 + \left(\frac{\mathbf{k}_{\parallel}}{k_{\perp}}\right)^2} \approx 1 + \frac{1}{2} \left(\frac{\mathbf{k}_{\parallel}}{k_{\perp}}\right)^2 \quad (2.7)$$

Hence, after substituting the square root free term into the cavity photon energy formula in equation 2.5, the energy of the cavity photon can be calculated as follows [48],

$$E_{\text{cav}}(\mathbf{k}) \approx \frac{\hbar c}{n_c} k_{\perp} \left(1 + \frac{1}{2} \left(\frac{k_{\parallel}}{k_{\perp}} \right)^2 \right) \quad (2.8)$$

$$E_{\text{cav}}(\mathbf{k}) \approx \frac{\hbar c}{n_c} k_{\perp} + \frac{\hbar c}{2n_c k_{\perp}} \mathbf{k}_{\parallel}^2 \quad (2.9)$$

The term $\frac{\hbar c}{n_c} k_{\perp}$ can be identified as the energy of the photon at zero in-plane momentum ($\mathbf{k}_{\parallel} = 0$) [48].

$$E_{\text{cav}}(0) = \frac{\hbar c}{n_c} k_{\perp} \quad (2.10)$$

The second term represents the correction to the energy due to the small in-plane momentum [48].

$$E_{\text{cav}}(\mathbf{k}_{\parallel}) \approx E_{\text{cav}}(0) + \frac{\hbar c}{2n_c k_{\perp}} \mathbf{k}_{\parallel}^2 \quad (2.11)$$

This can be written with an effective mass of the cavity photon m_{cav} [55].

$$E_{\text{cav}}(\mathbf{k}_{\parallel}) \approx E_{\text{cav}}(0) + \frac{\hbar^2 \mathbf{k}_{\parallel}^2}{2m_{\text{cav}}} \quad (2.12)$$

$$m_{\text{cav}} = \frac{E_{\text{cav}}(0)}{c^2/n_c^2}. \quad (2.13)$$

Where the effective cavity photon mass m_{cav} is given by, and it is mainly on the order of $10^{-5}m_e$ [48, 55].

2.4.1 The Quantum Model of the Exciton-Polariton

The Hamiltonian of the polariton can be written as the following,

$$\hat{H}_{\text{pol}} = \hat{H}_{\text{cav}} + \hat{H}_{\text{exc}} + \hat{H}_{\text{Int}} \quad (2.14)$$

$$\hat{H}_{\text{pol}} = \sum E_{\text{cav}}(\mathbf{k}_{\parallel}, k_c) \hat{a}_{\mathbf{k}_{\parallel}}^{\dagger} \hat{a}_{\mathbf{k}_{\parallel}} + \sum E_{\text{exc}}(\mathbf{k}_{\parallel}) \hat{b}_{\mathbf{k}_{\parallel}}^{\dagger} \hat{b}_{\mathbf{k}_{\parallel}} + \sum g_0 (\hat{a}_{\mathbf{k}_{\parallel}}^{\dagger} \hat{b}_{\mathbf{k}_{\parallel}} + \hat{a}_{\mathbf{k}_{\parallel}} \hat{b}_{\mathbf{k}_{\parallel}}^{\dagger}) \quad (2.15)$$

where $\hat{a}_{\mathbf{k}_{\parallel}}^{\dagger}$ is the photon creation operator, k_c is the out of plane momentum $k_c = k \cdot \hat{z}$, $k_{\parallel}$ is the in plane momentum, $\hat{b}_{\mathbf{k}_{\parallel}}^{\dagger}$ is the exciton creation operator, g_0 is the dipole interaction strength of the exciton-photon [48].

The following transformation equations can be used to diagonalize the Hamiltonian [48].

$$\hat{P}_{k_{\parallel}} = X_{k_{\parallel}} \hat{b}_{k_{\parallel}} + C_{k_{\parallel}} \hat{a}_{k_{\parallel}} \quad (2.16)$$

$$\hat{Q}_{k_{\parallel}} = -C_{k_{\parallel}} \hat{b}_{k_{\parallel}} + X_{k_{\parallel}} \hat{a}_{k_{\parallel}} \quad (2.17)$$

Where $\hat{P}_{k_{\parallel}}$ is the creation, and $\hat{Q}_{k_{\parallel}}$ is the annihilation operators of the lower polariton branch and upper polariton branch, respectively. $C_{k_{\parallel}}$ and $X_{k_{\parallel}}$ are the Hopfield coefficients which represent the amplitudes of the upper and lower polariton branches because a polariton is a linear superposition of exciton and photon with the same $k_{\parallel}$. The sum of their squares must be equal to one [56, 48].

$$|X_{k_{\parallel}}|^2 + |C_{k_{\parallel}}|^2 = 1 \quad (2.18)$$

After diagonalization, the Hamiltonian of the polariton becomes as follows,

$$\hat{H}_{pol} = \sum E_{LP}(k_{\parallel}) \hat{P}_{k_{\parallel}}^{\dagger} \hat{P}_{k_{\parallel}} + \sum E_{UP}(k_{\parallel}) \hat{Q}_{k_{\parallel}}^{\dagger} \hat{Q}_{k_{\parallel}} \quad (2.19)$$

where $\hat{P}_{k_{\parallel}}^{\dagger}$ and the $\hat{Q}_{k_{\parallel}}^{\dagger}$ are the creation and annihilation operators.

When the energy of the upper polariton branch equals the lower polariton branch, the squares of Hopfield coefficients become 1/2, which also means the upper and lower polaritons are exactly half photon and half exciton. Hence, the energy of the lower and upper polariton becomes as follows.

$$E_{LP,UP}(k_{\parallel}) = \frac{1}{2}[E_{exc} + E_{cav} \pm \sqrt{4g_0^2 + (E_{exc} - E_{cav})^2}] \quad (2.20)$$

As the exciton and photon couple, the new polariton energies, upper and lower polariton branch anticross; in other words, strong coupling regimes are achieved in the system. It is only possible when $|E_{cav} - E_{exc}| \gg g_0$.

Since polariton is a quasi-particle, it has a lifetime. When the lifetime of the cavity photon and the exciton substitute the energy formula of the upper and lower polariton branches, it can be modified as follows,

$$E_{LP,UP}(k_{\parallel}) = \frac{1}{2}[E_{exc} + E_{cav} + i(\gamma_{cav} + \gamma_{exc}) \pm \sqrt{4g_0^2 + (E_{exc} - E_{cav} + i(\gamma_{cav} - \gamma_{exc}))^2}] \quad (2.21)$$

Where γ_{cav} is the decay rate or the linewidth of the cavity photon mode, and the γ_{exc} is the decay rate or the linewidth of the exciton. Half of the absolute value of the difference between the decay rates of the exciton and the cavity photon must be smaller than the coupling strength to achieve the anticrossing; see equation 2.22 [48].

$$g_0 \gg \frac{|\gamma_{cav} - \gamma_{exc}|}{2} \quad (2.22)$$

The Rabi frequency, Ω , which it is related to the Rabi splitting, $\hbar\Omega$, can be calculated as the following [55, 57],

$$\Omega = 2\sqrt{g_0^2 - \frac{1}{4}(\gamma_{cav} - \gamma_{exc})^2} \quad (2.23)$$

The Rabi splitting refers to the energy split between the upper and lower polariton branches [55].

Moreover, the coupling strength can also be approximately calculated as follows [55],

$$g_0 \approx \sqrt{\frac{2\Gamma_0 c N_{QW}}{n_c L_{eff}}} \quad (2.24)$$

$$\Gamma_0 = \frac{\pi e^2}{n_c 4\pi\epsilon_0 m_e c} f \quad (2.25)$$

Where the f is the oscillator strength, N_{QW} is the effective number of the quantum wells in the microcavity, L_{eff} is the effective cavity length, and n_c is the refractive index of the inner cavity. The effective cavity length can be calculated as follows [55, 58].

$$L_{eff} = L_c + \frac{\lambda_c}{2n_c} \frac{n_1 n_2}{|n_1 - n_2|} \quad (2.26)$$

Where n_1, n_2 are the refractive indices of the high and low refractive index material of the thin films in the DBR, L_c is the actual cavity length, and the λ_c is the wavelength of the cavity mode [55].

Chapter 3

Experimental Methods

This chapter details the fabrication and characterization techniques employed in this thesis. Mechanical exfoliation is initially discussed for cleaving TMD MLs from bulk form. Raman and Photoluminescence spectroscopy are explained with examples of the monolayer TMD flakes. Following this, the stacking monolayers with the Dry Transfer method and the determination of twist angle with the Edge Identification method are mentioned thoroughly. The fabrication process of FP planar microcavities using plasma-enhanced chemical vapor deposition (PECVD) is described, and high and low refractive index thin films are characterized via ellipsometry. The cross-section images are obtained with Focused Ion Beam (FIB), and the thicknesses are determined with Transmission Electron Microscopy (TEM). In the last section, schematics and authentic images of the optical setups used to measure the photoluminescence in the room and low temperatures, as well as the time-resolved photoluminescence spectroscopy for lifetime measurements of IEX, are explained.

3.1 Mechanical Exfoliation

Mechanical exfoliation is one of many methods to obtain ML flakes from TMD bulk crystals. This method is superior to others in achieving ML crystals with fewer defects. Furthermore, it requires a simple scotch tape for application; see figure 3.1.

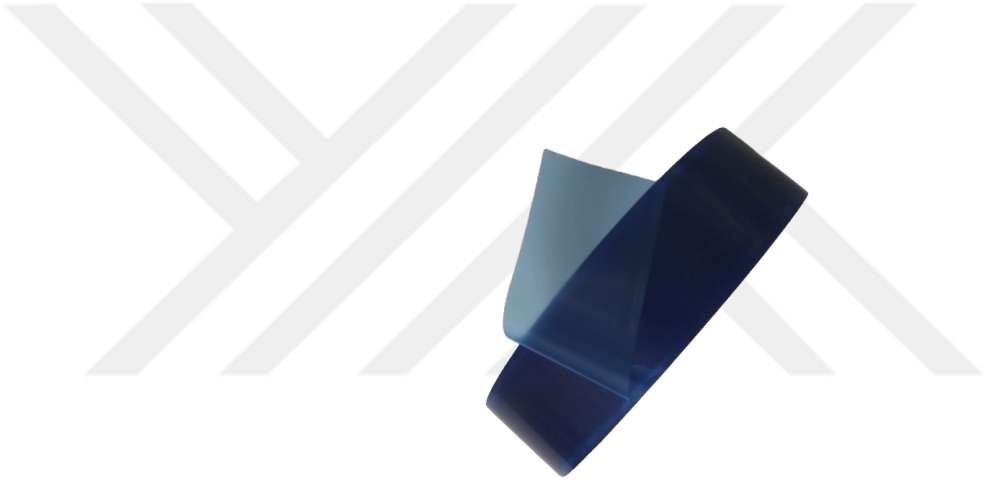


Figure 3.1: The scotch tape used for mechanical exfoliation

The $MoSe_2$ and WSe_2 bulk crystals are purchased from 2D semiconductors company USA. The exfoliated crystals on scotch tape can be seen in figure 3.2.

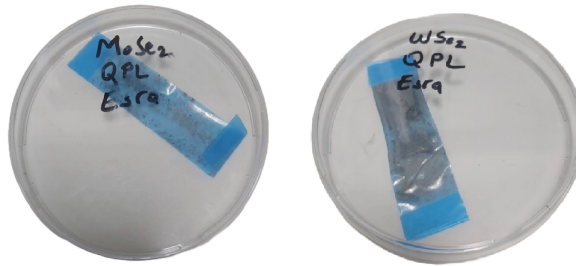
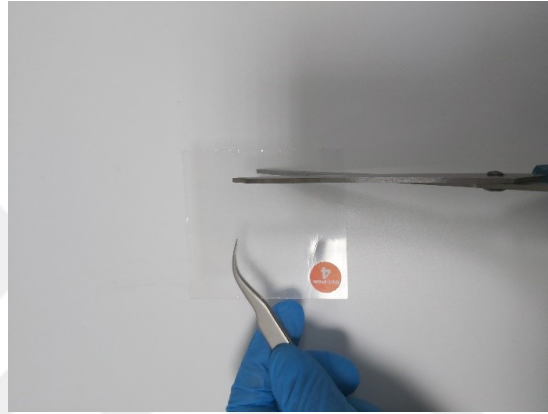


Figure 3.2: The $MoSe_2$ and the WSe_2 exfoliated crystals on scotch tape

After exfoliation, a piece of Polydimethylsiloxane sheet (PDMS) is cut with scissors and put on the edge of a slide; it can be seen in figure 3.3 (a) and (b) respectively. Then, the scotch tape with exfoliated flakes is pressed on the PDMS sheet to transfer ML flakes to the PDMS, or the tape can be pressed directly on silicon wafers. It depends on the application.



(a)



(b)

Figure 3.3: (a) Cutting a PDMS sheet with a scissor and (b) A piece of PDMS sheet on a slide.

The slides for $MoSe_2$ and WSe_2 should be kept separate to avoid contamination.

3.2 Raman Spectroscopy and PL Measurement

3.2.1 WSe_2 Monolayer

On a silicon substrate, the MLs look transparent and darker. The increase in the number of layers makes the flake a more prominent green; after a certain point, the transparency disappears, and flakes become light green and opaque till they become gold, see figure 3.4. Gold/yellowish flakes are considered bulk.

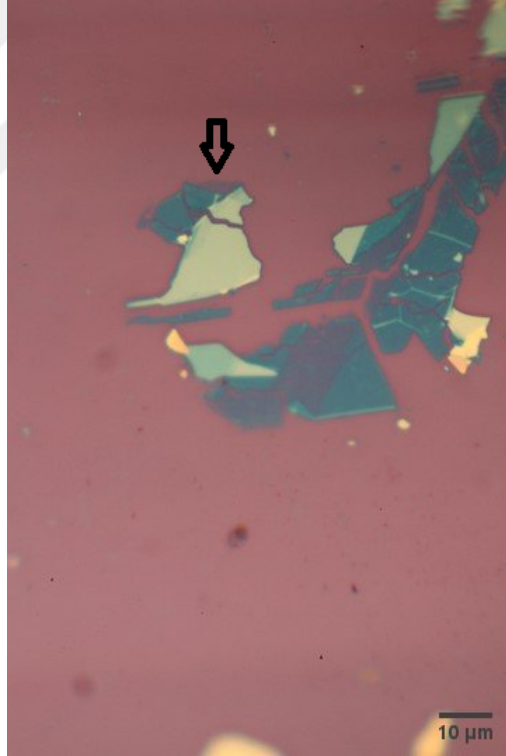


Figure 3.4: WSe_2 ML on silicon substrate pointed by an arrow.

The Raman shift and PL spectra of the WSe_2 ML in figure 3.4 can be seen in figure 3.5. As seen in the figure, the data matches the literature data.

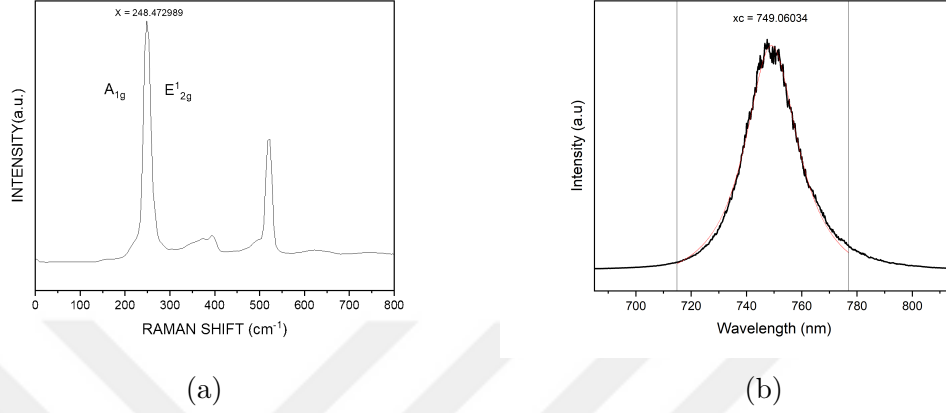


Figure 3.5: (a) Raman shift of the WSe_2 ML in figure 3.4, the laser power was 150 μ W, laser wavelength was 532 nm, and (b) Raman shift of the WSe_2 ML in figure 3.4, the laser power was 150 μ W, laser wavelength was 532 nm

3.2.2 $MoSe_2$ Monolayer

On PDMS, the flakes with a different number of layers can be differentiated with the optical microscope by looking at their shade of color, see figure 3.6. The darker, more transparent flakes are likely MLs or bilayers. To confirm the layer number, Raman spectroscopy and the PL spectrum of the flake can be measured.

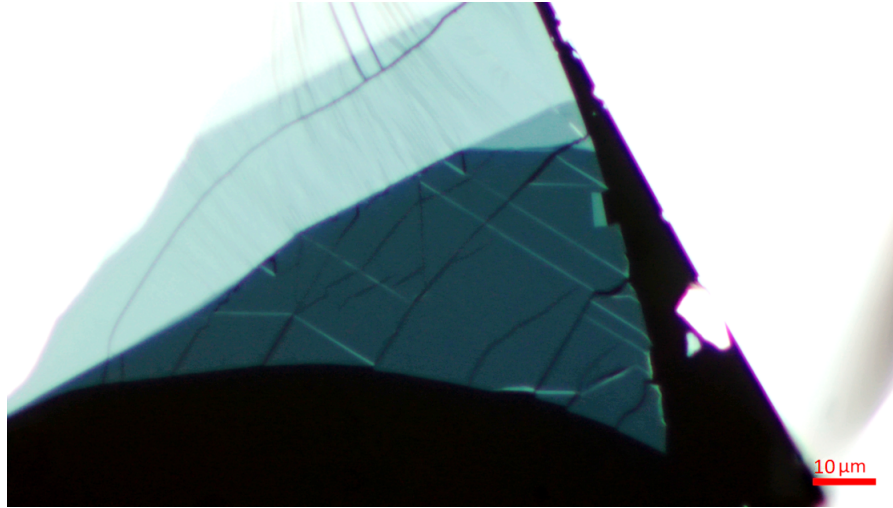


Figure 3.6: $MoSe_2$ ML on PDMS substrate

The Raman shift and the PL spectra of the monolayer in figure 3.6 can be seen in figure 3.7.

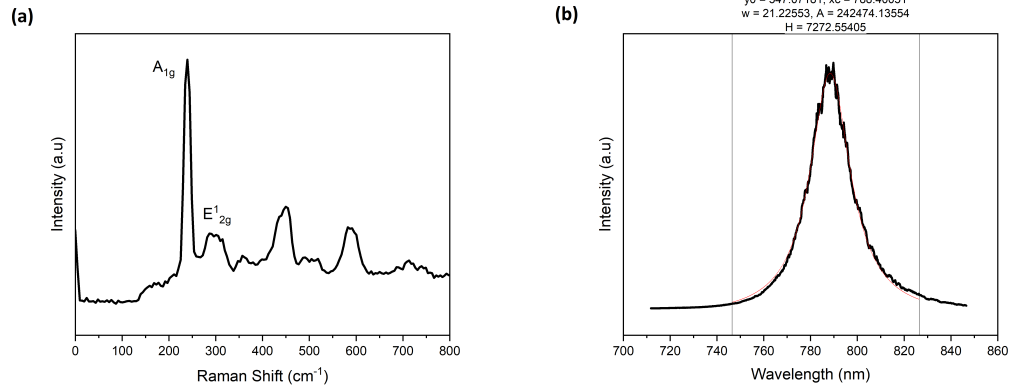
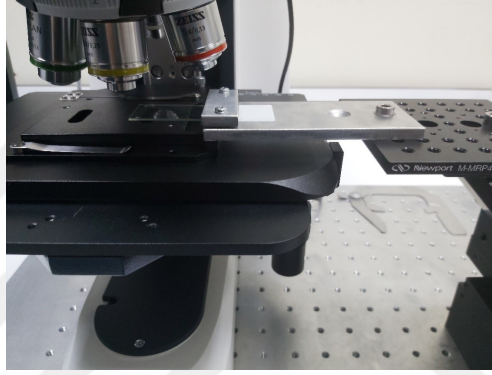


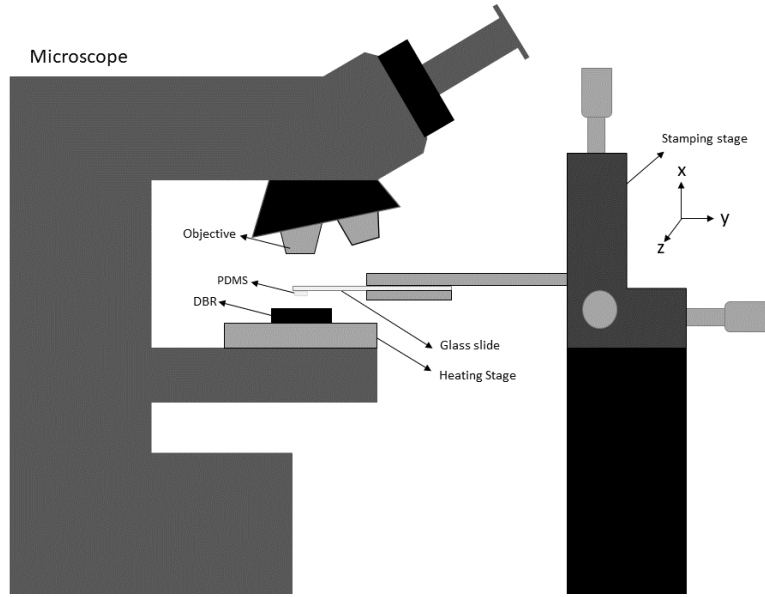
Figure 3.7: (a) Raman shift of the $MoSe_2$ ML in figure 3.7, the laser power was 150 μ W, laser wavelength was 532 nm, and (b) PL spectrum of the $MoSe_2$ ML in figure 3.7, Lorentz fitted, the laser power was 150 μ W, laser wavelength was 532 nm .

3.3 Stacking MLs with Dry Transfer Method

Dry transfer is a method used to transfer flakes from PDMS to substrates directly, such as silicon, quartz, etc., using an optical microscope with a transfer set. To transfer the flakes from PDMS to the substrate, the slide that holds the PDMS is inserted in the downward transfer set; see figure 3.8.



(a)



(b)

Figure 3.8: (a) Real image of the Dry Transfer Setup, and (b) The schematic representation of the Dry Transfer Setup

Then, the optic microscope focuses on the substrate's transfer area, and the slide is pushed down to the substrate. The whole process can be done with the help of a heating pad set at a temperature below 150 °C due to the glass transition temperature of the PDMS for a smooth transfer without any fracturing in the flake.

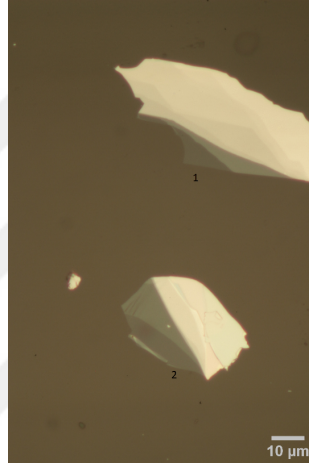


Figure 3.9: WSe_2 MLs on PDMS sheet

During the transfer between the PDMS and the substrate, a meniscus occurs at the first contact, and it moves in the same direction as the PDMS and can be seen in figure 3.10.

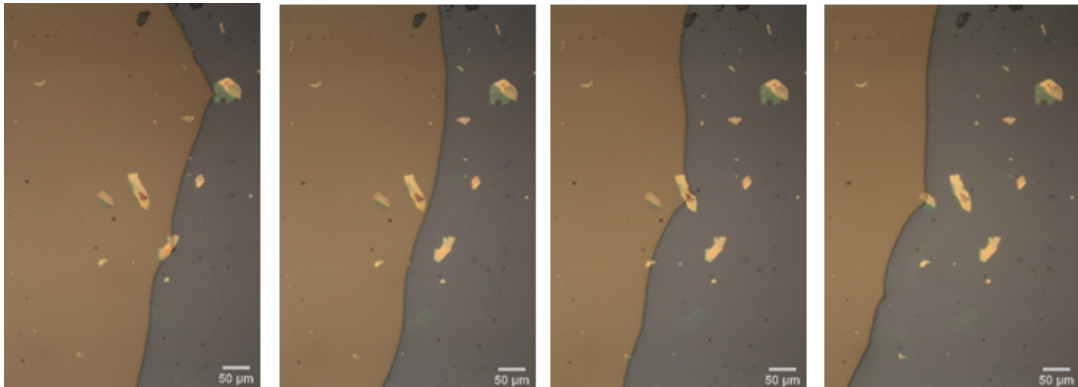


Figure 3.10: The meniscus of the PDMS during transfer

After the transfer, the ML WSe_2 flakes look dark green on the silicon substrate, as seen in figure 3.11.

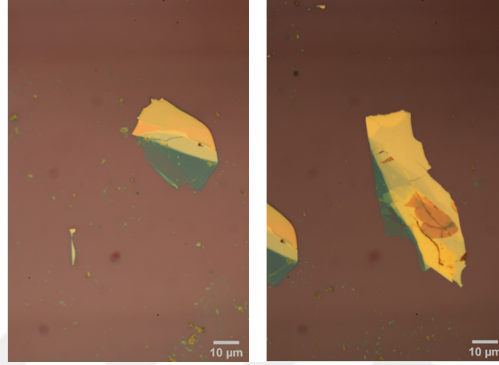


Figure 3.11: After transfer, WSe_2 flakes on silicon substrate

After the transfer of the first ML, the second ML is transferred on top of the first layer using the same dry transfer method; hence, the heterostructure can be fabricated. Also, the angle between monolayers can be predetermined, and a transfer can be made accordingly. For that kind of transfer, a rotation stage would be helpful. Below, you can see the $MoSe_2$ ML 3.12.

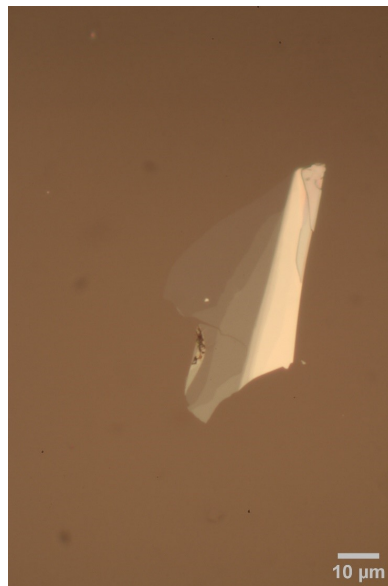


Figure 3.12: ML $MoSe_2$ on PDMS sheet

PL spectroscopy characterizes the heterostructures after the second ML is dry transferred. After the characterization, the heterostructure can be seen in figure 3.13.

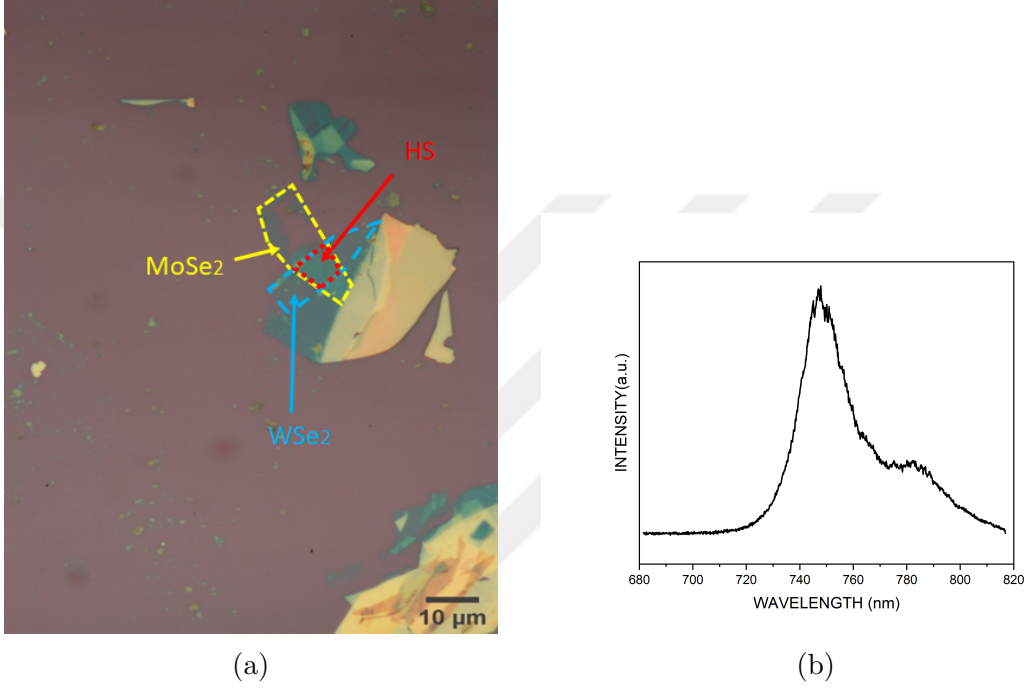


Figure 3.13: (a) HS labeled region is the heterostructure area, and (b) PL results of the heterostructure in (a)

3.3.1 The Edge Identification Method

Before stacking the MLs to fabricate the HSs, the orientation of the lattice of the TMD MLs should be determined. There are several methods to identify the lattice orientation, such as second harmonic generation (SHG) spectroscopy and the edge identification method (EIM) [7], which is a statistical approach. The SHG method is a non-invasive and straightforward method. Still, it requires a unique optical setup, which makes the identification of the orientation of the flake more expensive than the edge identification method. The EIM is also a non-invasive method and a more cost-effective solution.

Identifying the crystal structure of the ML flakes before fabrication is crucial to maintaining the angle between the layers, as it can impact the intensity of the peaks in the photoluminescence spectrum. Unintended mismatch of the crystal lattices of the MLs can be avoided by transferring the MLs while carefully considering the angle between their edges during the heterostructure fabrication process.

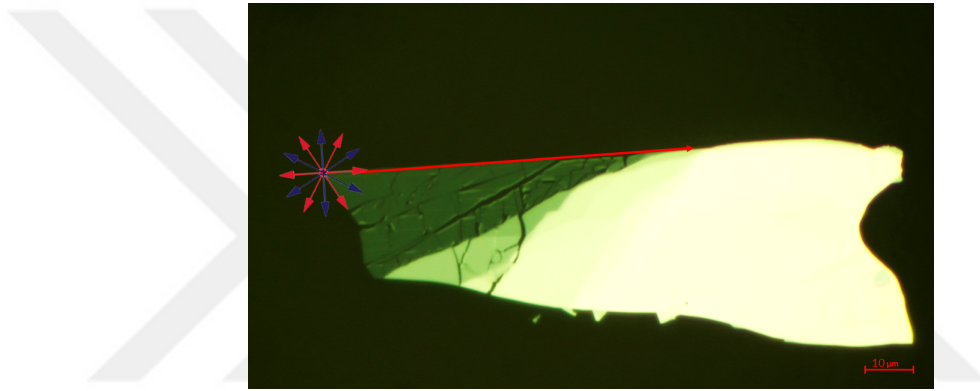


Figure 3.14: $MoSe_2$ ML flake with identified zigzag edge

As you can see in figure 3.14, the edges of the $MoSe_2$ ML flake are determined with the help of an arrow pattern. The angle between the red arrows is 60° ; therefore, based on the statistics, the two edges that align with the red arrows should have a zigzag edge structure. In figure 3.15, you can see the WSe_2 flake, which has two zigzag edges, as well as the $MoSe_2$ flake in 3.32.

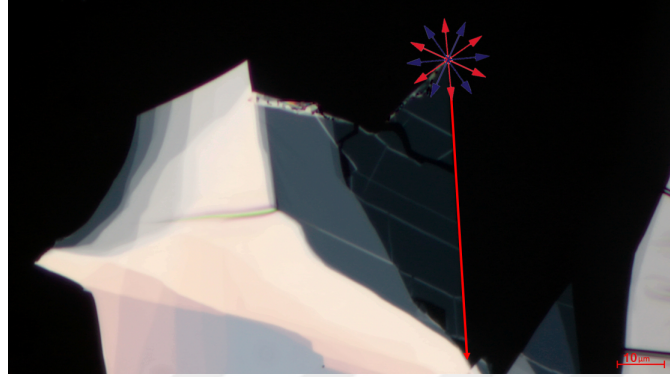


Figure 3.15: WSe_2 ML flake identified with zigzag edges

The angle between edges of MLs should be close to 0° or 60° degrees; these angles are referred to as magic twist angles, and lattice matching improves as the angle between the edges approaches these values, as mentioned in Chapter 2.

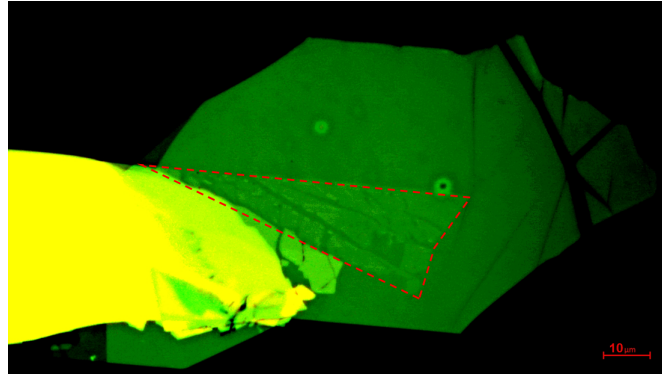


Figure 3.16: The area shown with dashed line is $MoSe_2$ flake on top of the few-layers hBN flake

Moreover, the HSs should be encapsulated with a dielectric material like hBN to isolate it from the surfaces of DBRs and converge the decrease in the exciton linewidths to the Fourier limit [59]. To do that, first, a few layers of hBN flake transfer onto the DBR by utilizing the dry transfer method; an example can be seen in figure 3.16, the $MoSe_2$ flake transferred on top of the hBN flake firstly. Then WSe_2 flake transferred to make the HS, as shown in figure 3.17. After that, a thin layer of hBN flake transferred on top of the HS region; see figure 3.18.

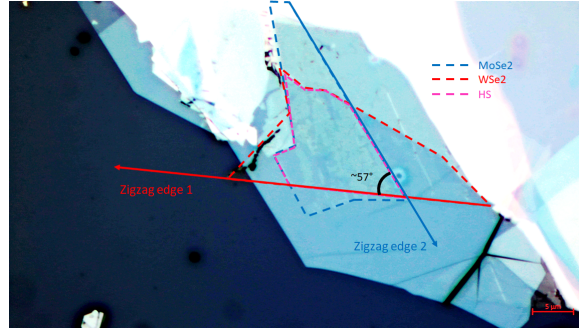


Figure 3.17: HS closed with a thin layer of hBN flake shown with the red dashed line

The HS in figure 3.17 is stacked with approximately 57 degrees in between two zigzag edges of the $MoSe_2$ flake and WSe_2 flake.

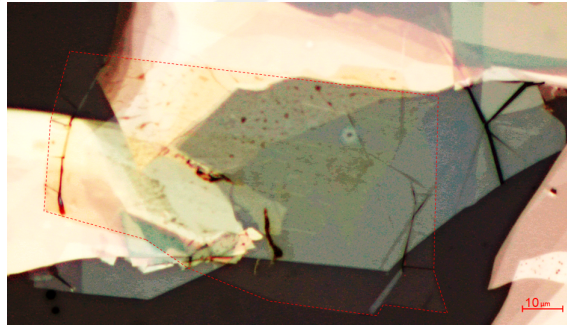


Figure 3.18: HS closed with a thin layer of hBN flake

In the given figure, labeled as Figure 3.19 (a), an HS is displayed that was created without utilizing the edge identification method. Consequently, the lattice mismatch between the two edges is unknown. Using the edge identification method, Figure 3.19 (b) displays an HS with an approximate angle of 0° between the two edges.

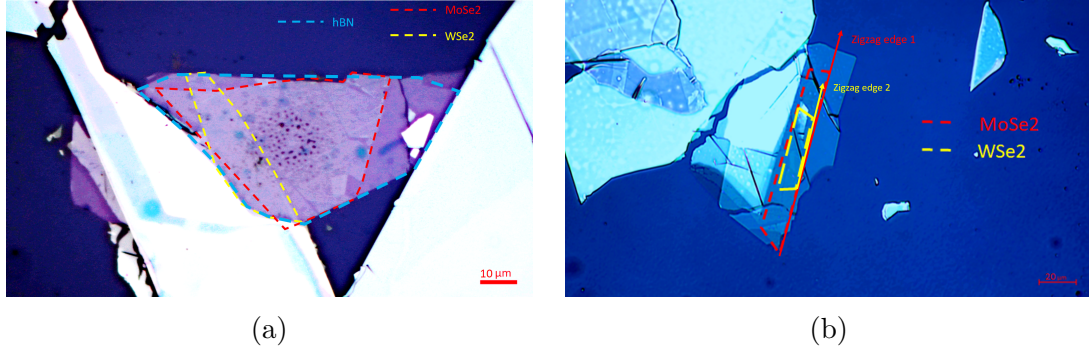


Figure 3.19: (a) HS image with dashed lines on bottom DBR, and (b) HS fabricated by utilizing the edge identification method on bottom DBR

In figure 3.20, some other examples of HSs can be seen. The HS in 3.20 (a) is stacked without considering the angle between monolayers. The heterostructure in 3.20 (b) has 57° between monolayers, which makes it H-stack. The HSs in 3.20 (c) and (d) are R-stack because the angle between their monolayers is close to zero.

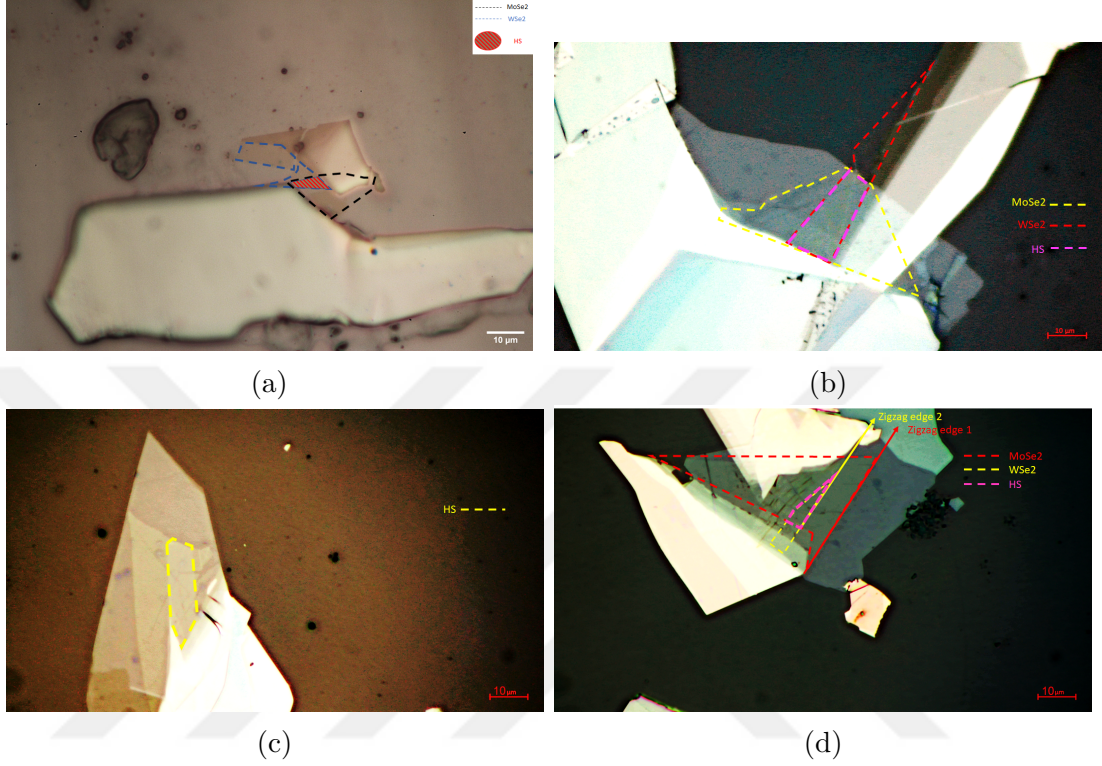


Figure 3.20: (a, b, c, d) Other examples of HSs by utilizing Dry Transfer method

3.4 Fabrication and Characterization of The Fabry-Pérot Cavities

To build a FP resonator, two distributed Bragg mirrors (DBRs) are fabricated on top of each other. The following equation determines the layer thicknesses of the SiO_2 and Si_3N_4 [43].

$$d = \frac{\lambda}{4n} \quad (3.1)$$

Where in the equation 3.1 λ is the wavelength of the interlayer exciton resonance, and n is the refractive index of the layers when the λ is equal to the exciton peak. This refractive index data is obtained with an Ellipsometer; the refractive index vs wavelength graphs can be seen in figure 3.22.

The refractive index of the Si_3N_4 for 885 nm, which is the interlayer exciton peak wavelength, is 1.7955, and the refractive index of the SiO_2 for the interlayer exciton peak wavelength is 1.463. For eight periods, the schematics of the bottom DBR can be seen in figure 3.21.

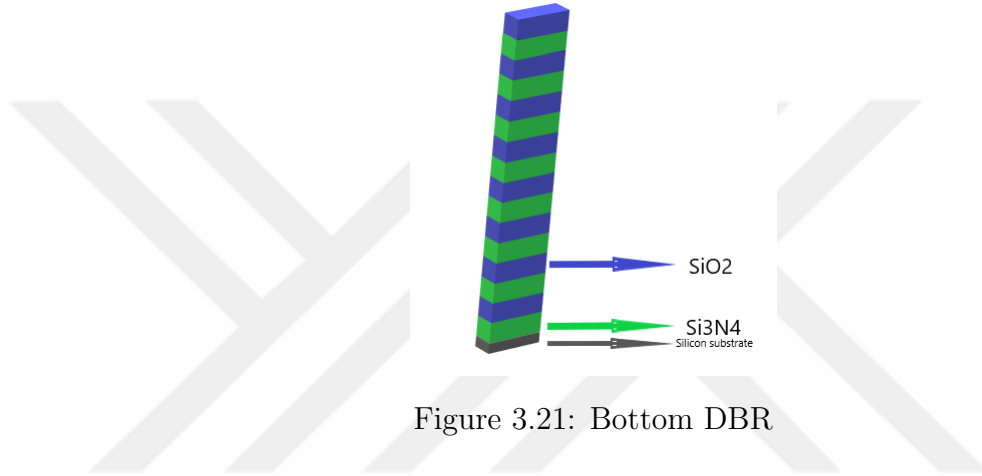


Figure 3.21: Bottom DBR

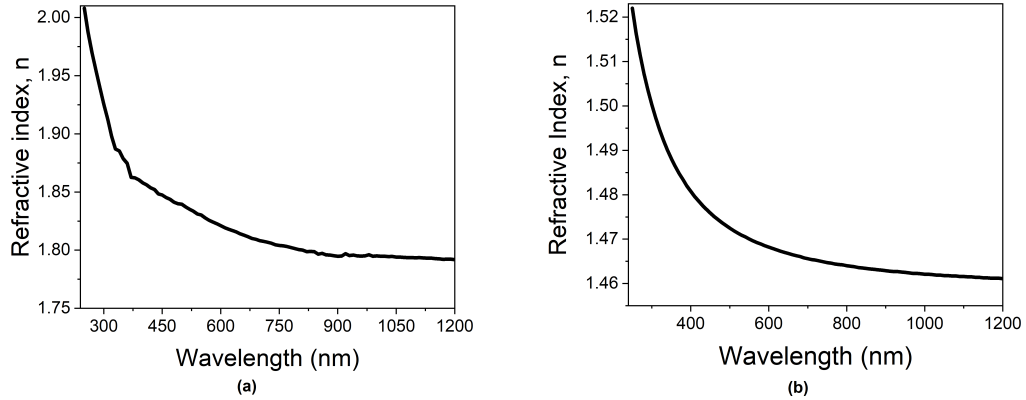


Figure 3.22: (a) The refractive index of the Si_3N_4 obtained with Ellipsometer, and (b) The refractive index of the SiO_2 obtained Ellipsometer.

After determining all the parameters of the DBR, it is fabricated using a plasma-enhanced chemical vapor deposition (PECVD) system; the thin films are coated onto a piece of silicon substrate, as shown in figure 3.23. After fabrication, the cross-section image of the DBR is obtained using Focused Ion Beam Lithography (FIB) to see the layer formation, which can be seen in figure 3.24.

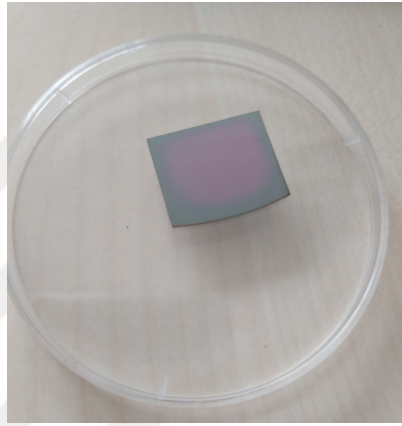


Figure 3.23: The bottom DBR

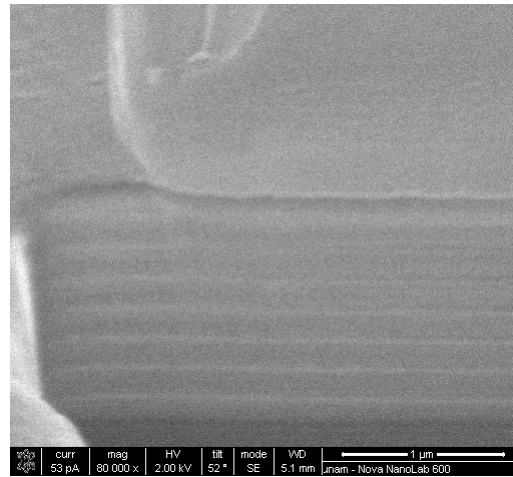


Figure 3.24: Cross-section image of the bottom DBR obtained with FIB

The thin films deposited to fabricate the DBR characterized with Transmission Electron Microscopy (TEM) showed that the first PECVD recipe did not achieve the target thicknesses, as shown in figure 3.25. For Si_3N_4 , the radio frequency power (RF power) was 20 watts, and for the SiO_2 was 70 watts. The process time for Si_3N_4 was 10 minutes 20 seconds and for SiO_2 was 5 minutes 15 seconds.

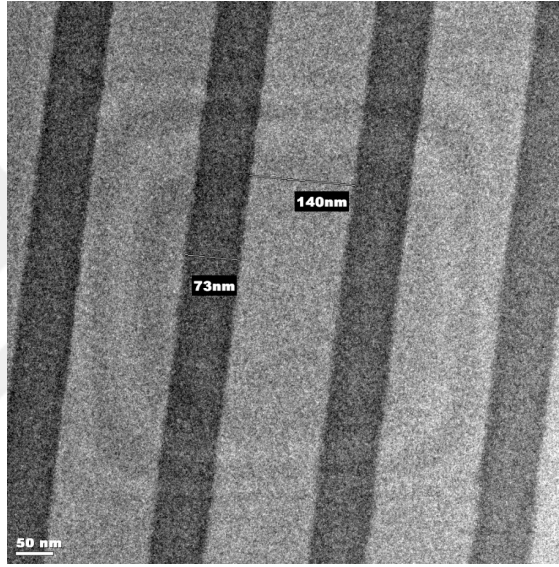


Figure 3.25: TEM image of the DBR with scaled layers of thin films

Initial measurements showed deviations from the target values: SiO_2 layers were 140 nm instead of the intended 151 nm, and Si_3N_4 layers were 73 nm instead of 123 nm. Consequently, the PECVD growth times for SiO_2 and Si_3N_4 layers were adjusted based on TEM results. Initially, the deposition times were one minute for a 12 nm Si_3N_4 thin film and 3.5 minutes for a 100 nm SiO_2 thin film. Post-optimization, the deposition times were revised to 8.5 seconds per nanometer for Si_3N_4 and 2.25 seconds per nanometer for SiO_2 . No other process parameters were altered to maintain control over the deposition process.

As you can see in figure 3.26, the colors of the two DBRs are distinctively different. The second DBR is fabricated with 70 W RF power for SiO_2 , and the processing time was 5 minutes 45 seconds; for Si_3N_4 , the RF power was 20 W, and the processing time was 17 minutes 45 seconds to achieve 123 nm for Si_3N_4 and 150 nm for SiO_2 .

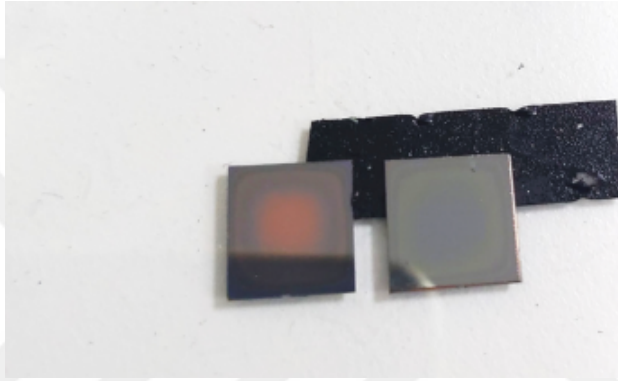


Figure 3.26: The color difference between bottom DBRs fabricated with two different PECVD recipes

The TEM images indicate that the second PECVD recipe did not precisely achieve the target thicknesses of 150 nm for SiO_2 and 123 nm for Si_3N_4 , as shown in figure 3.27. However, the thicknesses achieved were very close to the target thicknesses. Such deviations are not uncommon, given the nanometer scale of the thin films. Furthermore, achieving deposition precision within 5-10 nm is inherently challenging.

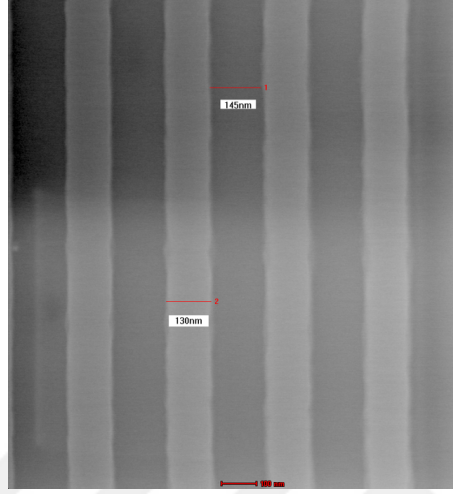


Figure 3.27: The TEM results of the second DBR, target thicknesses which were 150 nm for SiO_2 and 123 nm for Si_3N_4

To determine the cavity mode of the second DBR and its stopband, the reflectivity of the full cavity is measured; a full cavity has 16 periods of 32 layers of thin films, which can be seen in figure 3.28.

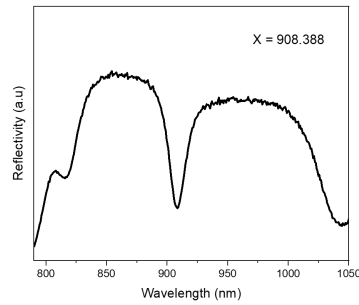


Figure 3.28: The stopband of the full cavity, the DBR of this cavity is shown in figure 3.27.

The DBR's cavity mode initially resonated at 908 nm; however, a resonating wavelength between 885-890 nm was required for cryogenic experiments due to the interlayer exciton resonance wavelength. By using this data, two new DBRs are fabricated, first to converge the interlayer exciton resonance wavelength at a cryogenic temperature which is approximately 890 nm; after multiple different combinations of the various thicknesses, the recipe was fixed at 18 minutes 10 seconds for the Si_3N_4 layer and 5 minutes 54 seconds for the SiO_2 layer for low-temperature DBR. You can see the stop band of the final low-temperature cavity in figure 3.29 (a). For the room temperature measurements, the time of deposition is changed to 17 minutes 28 seconds for the Si_3N_4 layer, and for the SiO_2 layer, the time of deposition is changed to 6 minutes 41 seconds, you can see the stop band in figure 3.29 (b).

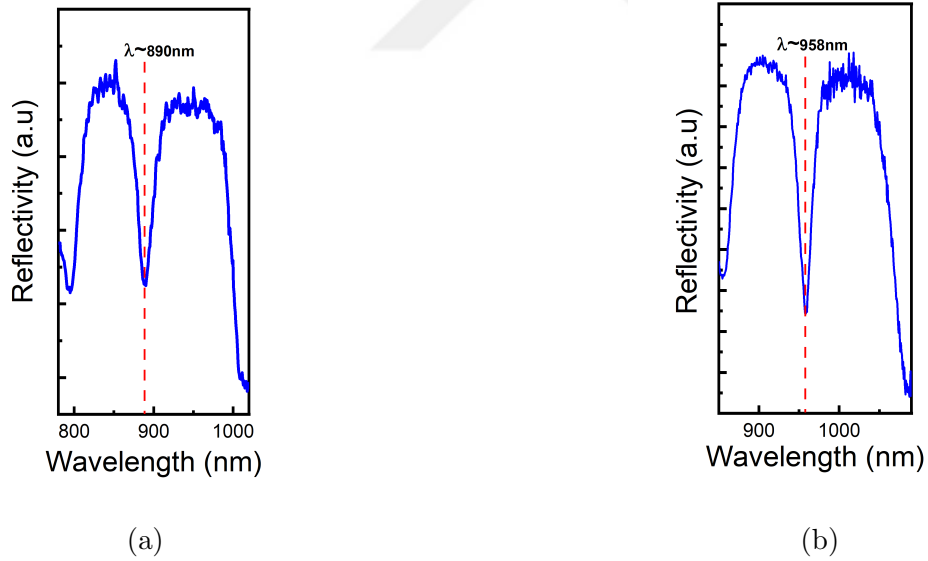


Figure 3.29: (a) The DBR stop band, the resonating wavelength at 890 nm for low-temperature interlayer exciton measurements, and (b) The DBR stop band, the resonating wavelength at 958 nm for room-temperature interlayer exciton measurements

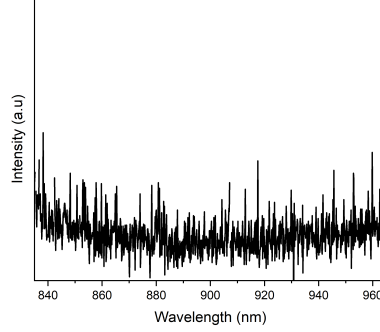


Figure 3.30: The bare DBR PL response

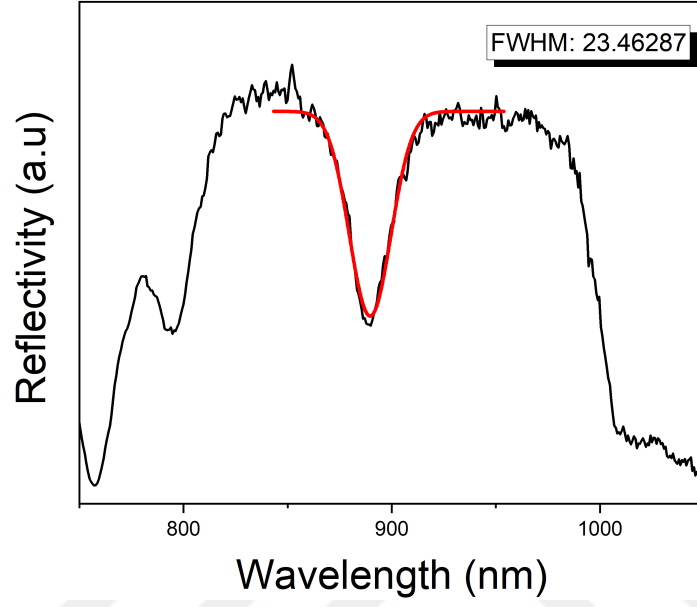
Moreover, the bare DBR PL was measured to see if a signal came from the interlayer exciton region. As seen in figure 3.30, there is no signal without any quantum wells embedded inside the cavity.

In figure 3.31, the stop bands of both cavities are fitted with the Gauss function with Origin Lab software; according to these results, the low-temperature cavity has approximately 23 nm full-width half maximum value (FWHM), and the room temperature cavity has approximately 20 nm FWHM. The quality factors can be calculated as follows,

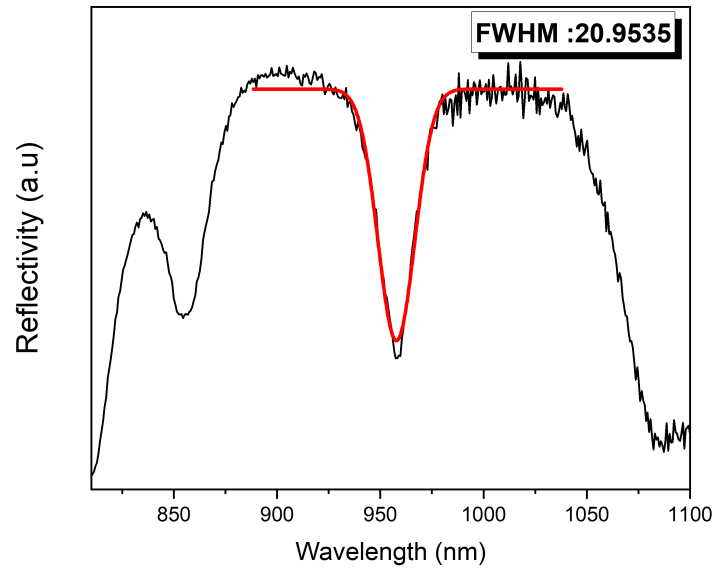
$$Q_{890} = \frac{890nm}{23nm} = 38.7 \quad (3.2)$$

$$Q_{958} = \frac{958nm}{20nm} = 47.9 \quad (3.3)$$

as seen in the equations 3.2 and 3.3.



(a)



(b)

Figure 3.31: (a) The stopband of the full cavity fitted with Gauss function resonating at 890 nm approximately with FWHM at 23 nm, and (b) The stopband of the full cavity fitted with Gauss function resonating at 958 nm approximately with FWHM 20 nm

Finally, it is essential to note that the color of thin films varies based on their thickness. This can be a qualitative way to guess the approximate thicknesses of the thin films; see figure 3.32.

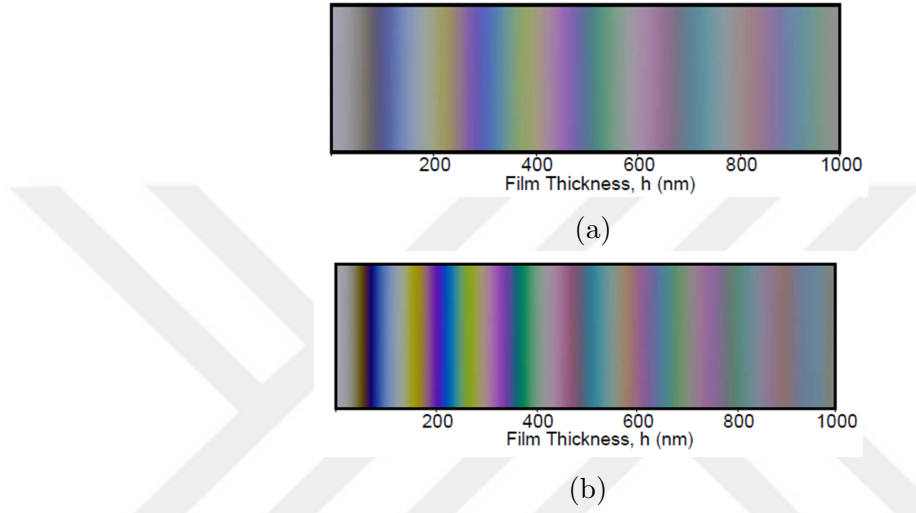


Figure 3.32: (a) SiO_2 thin film color change according to the thickness [15], and (b) Si_3N_4 thin film color change according to the thickness [15]

After finishing the HSs, the top layer of DBR is made with PECVD. The schematic of the full cavity embedded with the HS can be seen in figure 3.33.

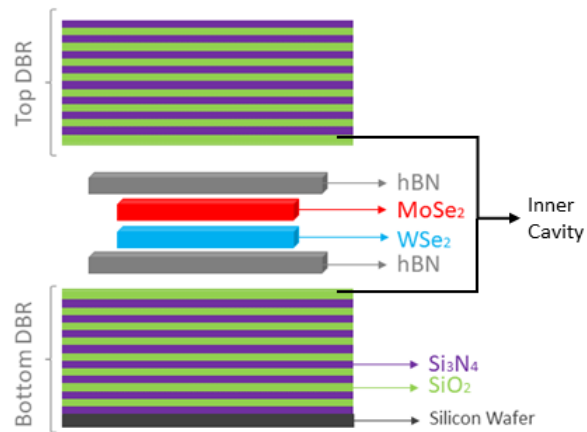


Figure 3.33: The schematic of full cavity embedded with HS

3.5 Optical Setups

3.5.1 Room Temperature Angle Resolved Photoluminescence Spectrometer

A home-built room temperature angle-resolved photoluminescence spectrometer (ARPS) is used for measuring the room temperature PL of the HSs embedded in room-temperature cavities mentioned in Chapter 3. Figure 3.34 (a) shows an actual real image of the room temperature angle resolved PL spectroscopy setup, and figure 3.34 (b) shows the schematic of the room temperature angle resolved PL spectroscopy setup. It consists of a ThorLabs single-axis, 1/2" to the point of rotation goniometric stage for changing the angle of the sample, an objective, collimator lenses for focusing the laser, a beam splitter (BS), mirrors to navigate the laser, a spectrograph, and a laser source with 532 nm wavelength.

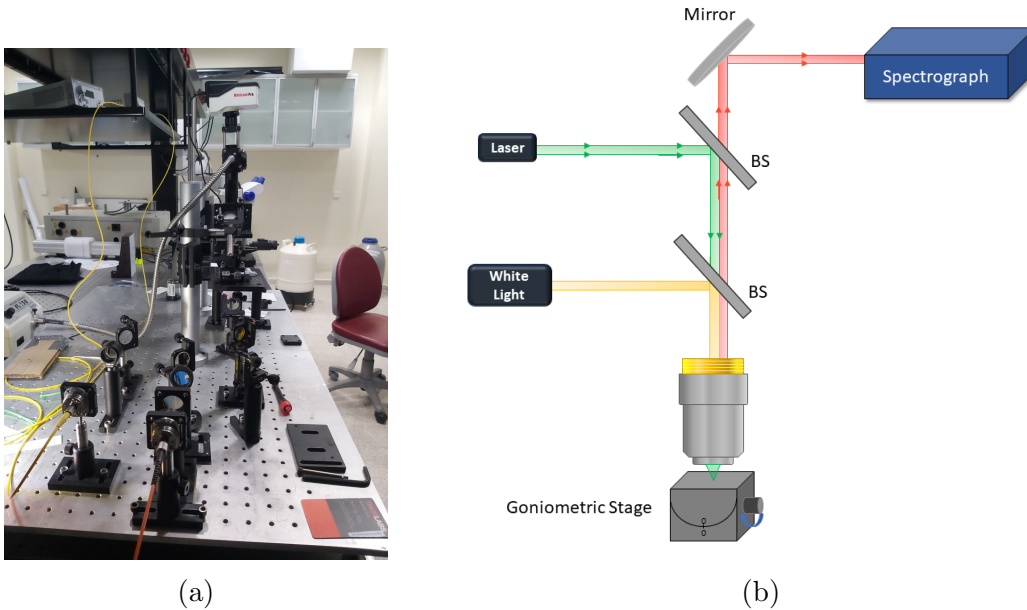


Figure 3.34: (a) Room temperature optic PL setup ARPS, and (b) Schematic of ARPS optical setup

3.5.2 Micro PL Optical Setup

The low-temperature measurements are conducted using a custom-built micro PL optic setup in our laboratory. The setup comprises a closed-cycle cryostat (attoDRY 1000), a spectrograph, and an optical breadboard with lenses, filters, and light sources [16, 60], which can be seen in the figure 3.35. The setup facilitates low-temperature PL spectroscopy measurements, including time-resolved PL spectroscopy. Operating within a cryostat capable of reaching temperatures as low as 3.5 K enables incremental temperature adjustments, facilitating comprehensive temperature-dependent measurements. The laser utilized was a 532 nm picosecond pulsed laser diode capable of operating in both continuous wave (CW) and pulsed wave modes; hence, PL spectra of the heterostructures are measured with the continuous wave mode, and the time-resolved PL measurements are done with the pulsed mode.

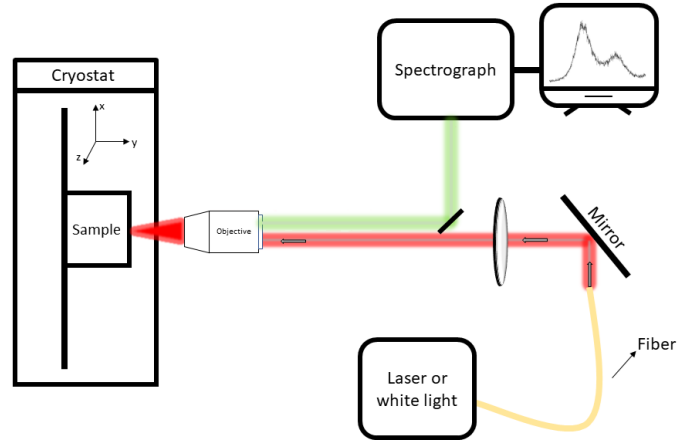
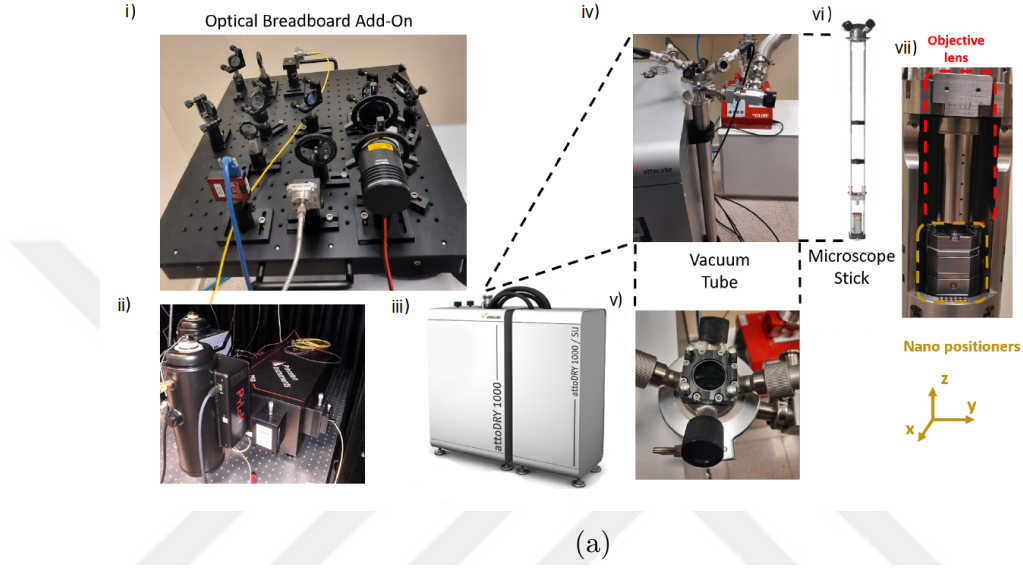


Figure 3.35: (a) Micro PL Optic Setup: i) The optical breadboard setup includes the white light source, laser source, and camera, ii) the spectrograph, iii) the cryostat (attoDRY 1000), iv) the vacuum tube, v) the window allowing light transmission, along with upper and lower cage plates and a microscope holder, vi) the objective lens and the sample holder with a nano-positioner [16] and (b) The schematic presentation of the micro PL setup

3.5.3 Time resolved Photoluminescence Spectroscopy

Time-resolved photoluminescence is used to measure the lifetime of the IEX. The time-correlated single photon counting (TCSPC) system (HydraHarp 400) is used, which counts the fluorescence photons with an Avalanche Photodiode detector (APD). APD converts the photons into electrical signals. Not after all cycles should a photon necessarily be emitted. After many cycles, the histogram of photons is taken as raw data and fitted with an exponential decay function to obtain the lifetime of the IEX. The slow step of the decay is considered the lifetime of the IEX. In figure 3.36, the schematic of the TCSPC system can be seen [61].

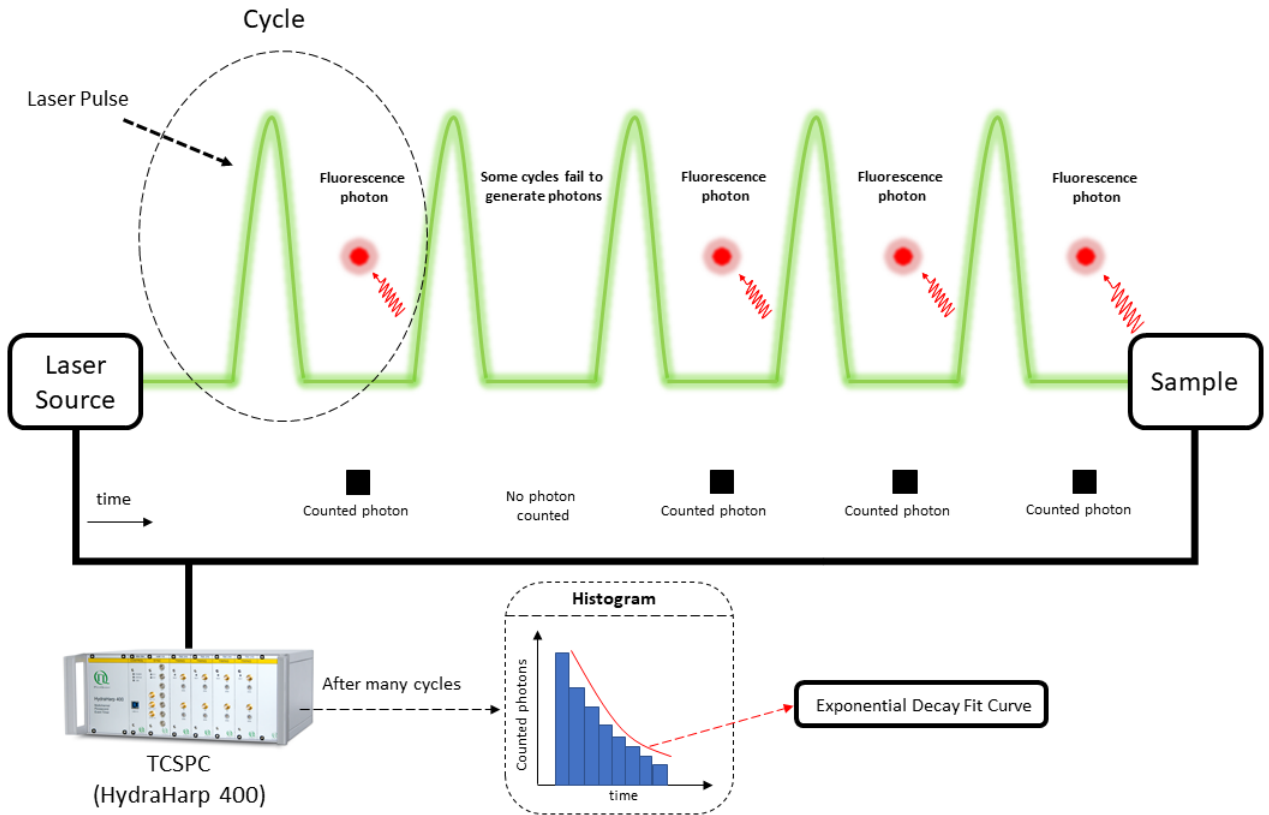


Figure 3.36: The time-resolved photoluminescence setup schematic.

Chapter 4

Results and Discussion

This chapter presents the results obtained from the room and low-temperature photoluminescence spectra measurements of TMD heterostructures fabricated using the dry transfer method, which is mentioned in Chapter 3. The measurements were conducted at room and low temperatures to explore the TMD heterostructures' optical properties inside a planar microcavity and the IEX's photophysics under different conditions. Enabling the micro PL setup, the PL measurements are obtained, and the lifetime of the spin-singlet state of the IEX is measured using time-resolved PL spectroscopy. Finally, the chapter concludes with the discussion section, where the results are evaluated, and it outlines possible directions for further research and suggests improvements and new experiments that could build upon the findings presented in this study.

4.1 Results

4.1.1 Room Temperature PL Measurements

The room temperature PL measurements of the IEXs were done using the PL mode of the Raman spectroscopy and ARPS. The optical microscope image of the fabricated R-type heterostructure is shown in figure 4.1 (a), and its intralayer and the interlayer exciton regions are shown in figure 4.1 (b) and (c), respectively. These measurements are obtained using the PL mode of the Raman spectrograph without the top DBR at $150 \mu\text{W}$. As seen in figure 4.1(c), at room temperature, the PL peak of the IEX is present but with low intensity at around 970 nm peak position; it should be noted that these measurements were conducted without the top DBR.

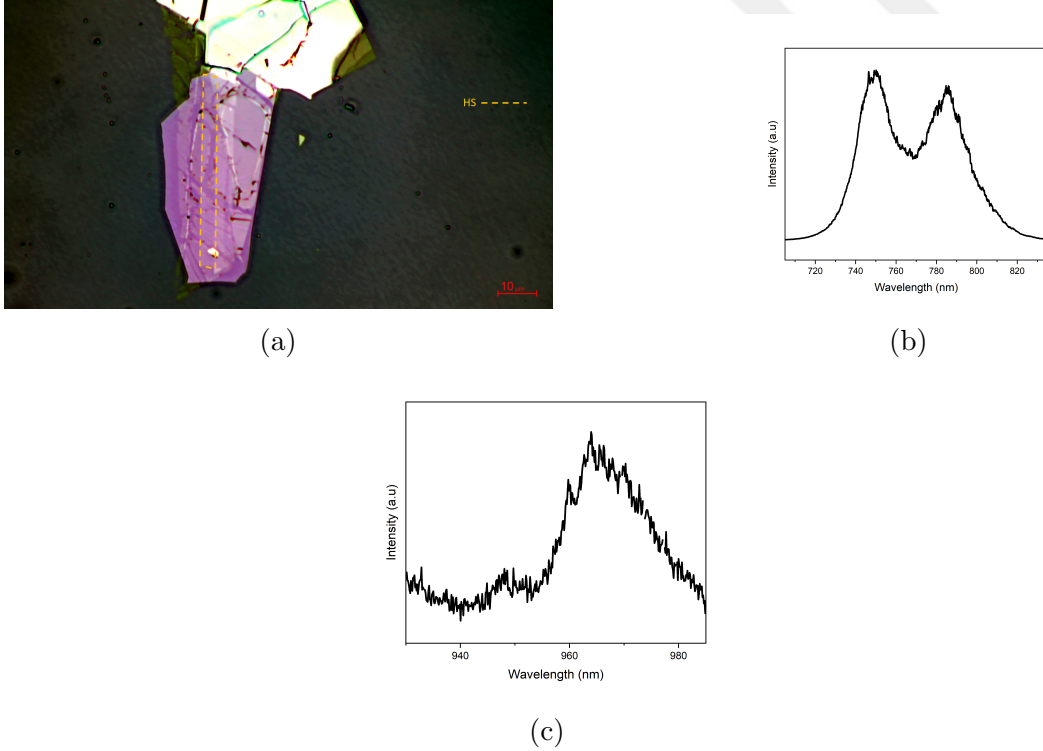


Figure 4.1: (a) The $WSe_2 - MoSe_2$ HS with nearly 0° twist angle on bottom DBR, (b) The PL spectrum of the intralayer exciton region of the heterostructure in (a) at room temperature, and (c) The PL spectrum of the interlayer exciton region of the heterostructure in (a) at room temperature

The IEX peak of another HS is shown in figure 4.2 (a) was not detected using our home-built ARPS setup, which can be seen in figure 4.2 (c). This absence can be attributed to the presence of the top DBR layer on the sample, which likely caused the low intensity of the IEX, rendering it undetectable. Also, at high temperatures, such as room temperature, the intensity of IEX is expected to be lower due to the increased phonon interactions. Moreover, the intralayer exciton region is measured as seen in figure 4.2 (b), the WSe_2 and $MoSe_2$ PL peaks are seen clearly at 755 and 790 nm, respectively, which are consistent with our previous PL data obtained individually from the MLs. All the measurements were done with ARPS $\approx 100\mu$ W laser power was used.

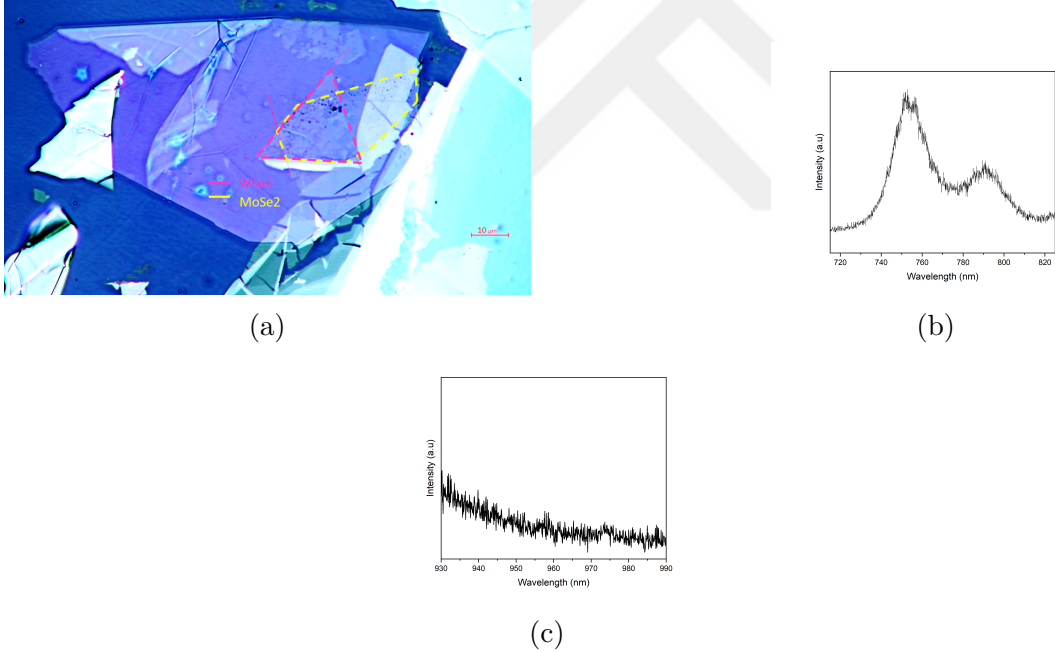


Figure 4.2: (a) The $WSe_2 - MoSe_2$ HS with nearly 57° twist angle on bottom DBR, (b) The PL spectrum of the intralayer exciton region of the heterostructure in (a) at room temperature measured with home-built ARPS setup, (c) The PL spectrum of the interlayer exciton region of the heterostructure in (a) at room temperature measured with home-built ARPS setup

Although the IEX was not detected at a 0-degree incident angle, further investigations were conducted. The PL of the IEX region was measured at various angles, including -10, -5, 0, 5, and 10 degrees. The ARPS measurements with changing angle of the HS in figure 4.2 (a) are shown in figure 4.3. As seen in the graph with the changing angle, there is no change in the IEX region. Hence, it is clear that even with the changing angle, the intensity of IEX remains undetected.

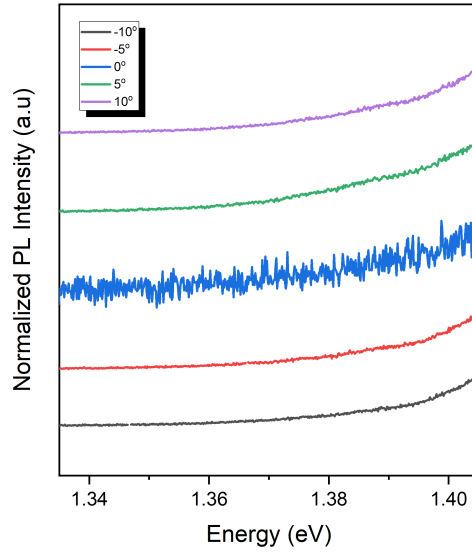
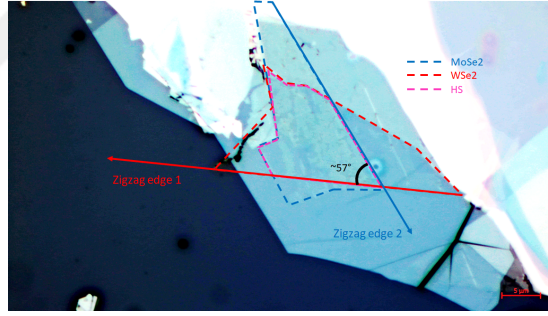


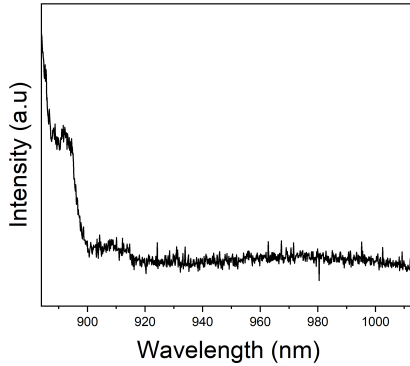
Figure 4.3: The ARPS measurements of the HS in figure 4.2 (a).

4.1.2 Low-Temperature PL Measurements

More HSs were fabricated on DBRs, which were fabricated explicitly for low-temperature PL measurements. Figure 4.4 (a) shows the image of one of these HSs (HS1), and the PL result of the IEX region is demonstrated in 4.4 (b). The angle between two zigzag edges of the MLs is approximately 57° . As seen in figure 4.4 (b), there is no prominent interlayer exciton peak at room temperature, but there is a broad low-intensity peak. After the room temperature characterization, the sample is put into our close-cycle Attocube AttoDRY 1000 cryostat. The PL spectrum is measured under 3.5 K. The PL of the intra- and interlayer exciton regions can be seen in figure 4.5. At 3.5 K, interlayer excitons become observable due to the suppression of phonon interactions.



(a)



(b)

Figure 4.4: (a) The optical image of HS1 for low-temperature measurements and (b) The PL result of the interlayer exciton region of the HS1 in (a) at room temperature.

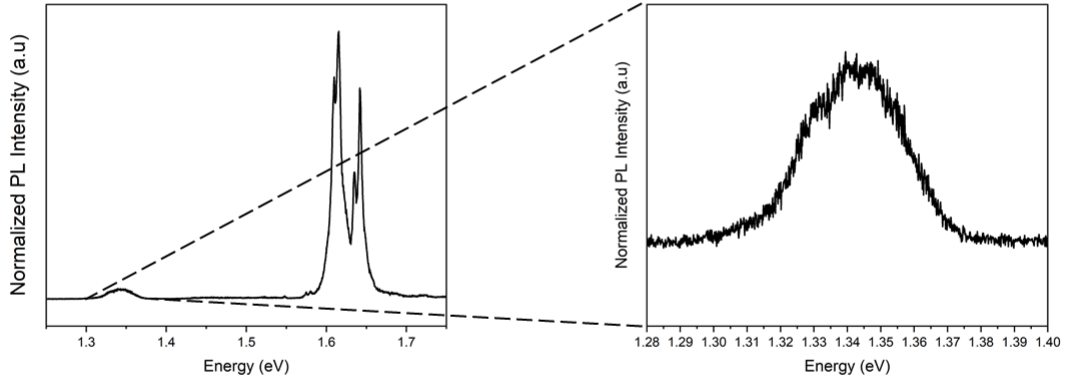


Figure 4.5: Intra- and interlayer region of the PL emission shown in 4.4 (a)

The PL peaks from the different regions of the same heterostructure at 3.5 K can be seen in figure 4.6 (a, b, c, d). As seen in figure 4.6 (a, b, c, d), the H-stack crystal structure is achieved in most parts of the HS1; the spin-triplet and spin-singlet states of IEX are bright due to the spin-orbit coupling happening in the conduction band of the $MoSe_2$ [12, 62, 63]. The energy differences between the spin-triplet and spin-singlet IEXs measured in the 27-34 meV range are slightly higher than previously reported values. At 3.5 K, in many experimental studies, the splitting between spin-singlet and spin-triplet IEX in $MoSe_2 - WSe_2$ HSs without the cavity is reported as approximately 24-26 meV [64, 65, 66, 37, 38, 67].

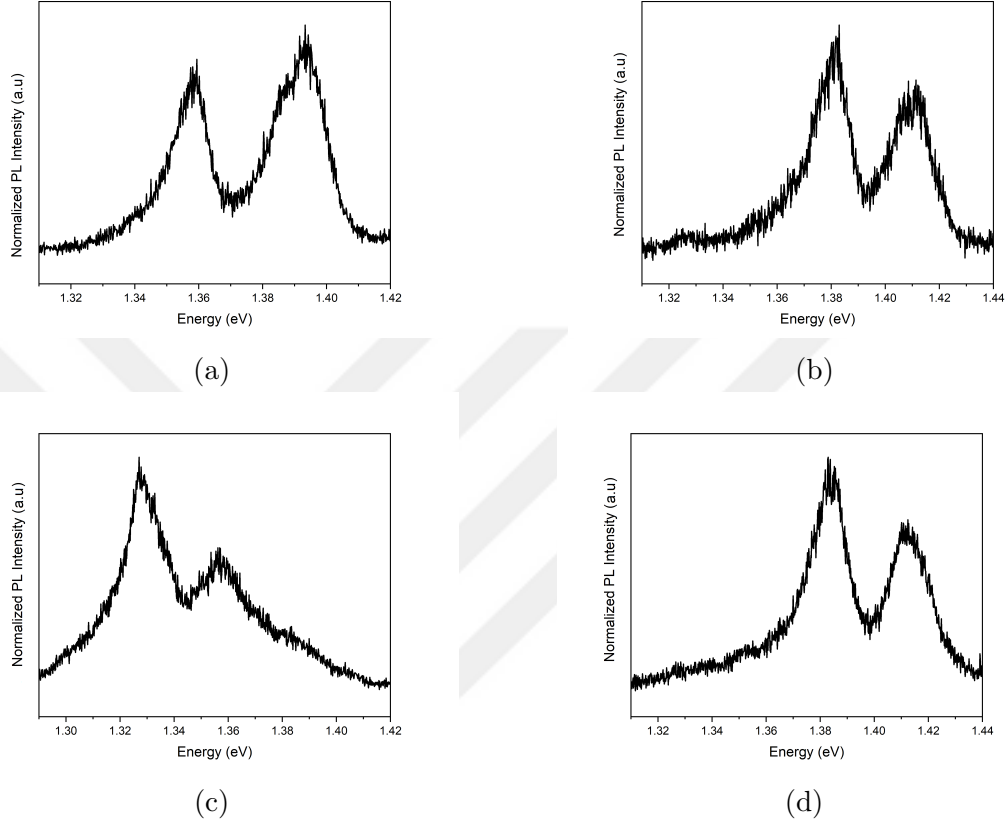


Figure 4.6: (a, b, c, d) The interlayer exciton peaks of the HS1 in figure 4.4 (a) obtained from different regions.

After measuring the IEX PL emission, for example, in figure 4.6 (a), its temperature dependence was also studied for further investigation. As seen in figure 4.7, the intensity of IEX changes with the changing temperature. The PL intensity is not normalized to know the intensity change with the changing temperature. For 3.5 and 10 Kelvin, the intensity of the spin-triplet IEX does not change significantly; at 30 Kelvin, the intensity of the spin-triplet IEX starts decreasing. At 50 Kelvin, the spin-triplet IEX became hard to recognize. However, it seems like the intensity of the spin-singlet IEX is not changing much for 3.5, 10, and 30 Kelvin; at 50 Kelvin, it decreases by approximately 50 percent. The reason is the thermal population of the lowest state at the low temperatures, which is the spin-triplet IEX is higher; with the increasing temperature, the number of thermally excited interlayer excitons increase and fill the high-energy spin-singlet states [37]. Moreover, rising temperatures also stimulate nonradiative decay channels as well.

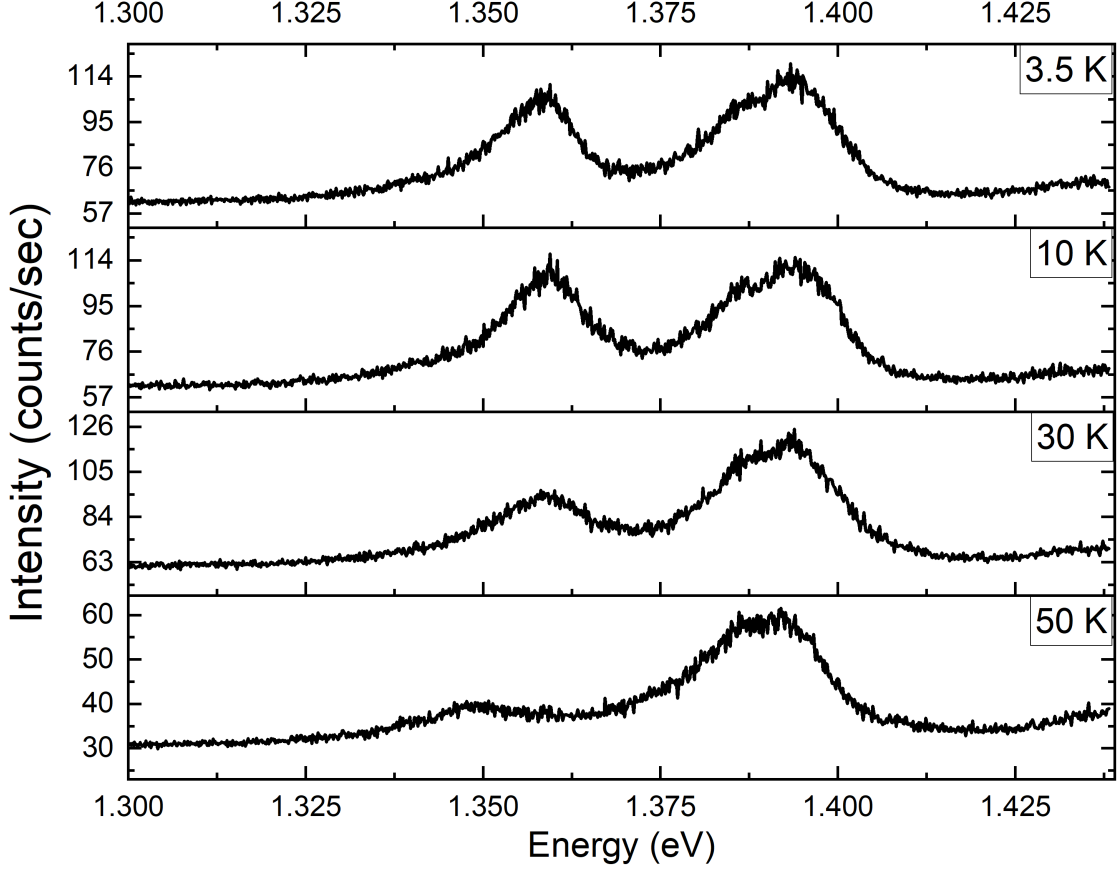


Figure 4.7: The temperature dependency of the PL emission of the HS1 in figure 4.6 (a) with the non-normalized PL intensity.

The spin-singlet IEX has a PL emission at 1.39 eV, the same as the FP cavity's resonance. This indicates that the interlayer exciton can couple to the cavity resonance, leading to the formation of exciton-polaritons. Upon close observation of the spin-singlet interlayer exciton peak in figure 4.9 (a), it can be seen that there is a small energy split in the spin-singlet IEX peak. The splitting is calculated, and the relation between the temperature and the splitting is shown in figure 4.9 (b). At 3.5 K, the splitting is nearly 9 meV, as shown in figure 4.9 (a). Although this might not seem sufficient for the splitting between two branches of an exciton-polariton, since the oscillator strength of interlayer exciton is much lower than the intralayer exciton, 9 meV Rabi splitting could be possible.

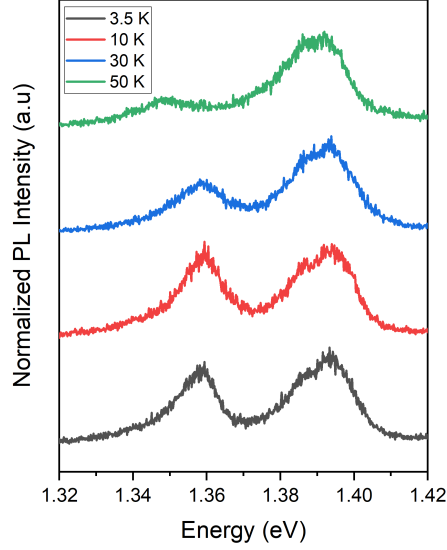


Figure 4.8: Temperature dependence of the PL emission of the HS1 in figure 4.6 (a)

Moreover, the coupling strength is directly related to the square root of the oscillator strength of the interlayer exciton [55]; the Rabi splitting is expected to be less than the intralayer exciton polariton's Rabi splitting. It is also clear that to safely attribute this splitting to exciton-polariton formation, one has to do whole angle resolved PL measurements as performed for intralayer excitons of TMD materials as in the study of Liu et al. [13].

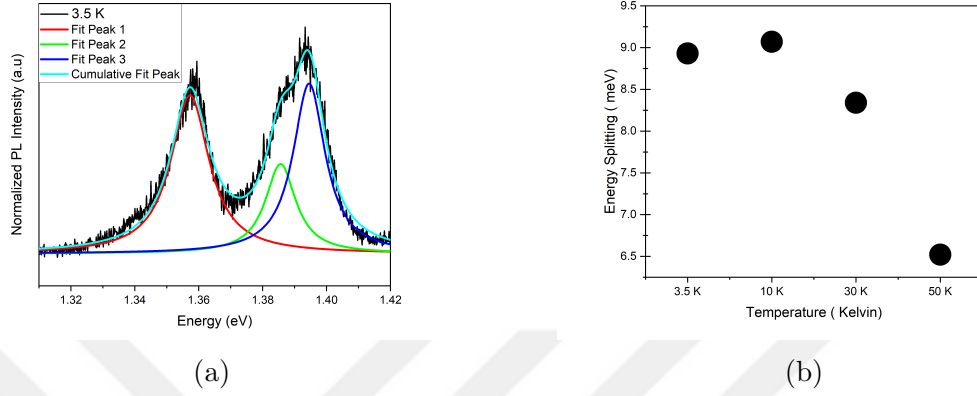


Figure 4.9: (a) The PL emission of the HS1 in figure 4.6 (a) at 3.5 K, fitted with Lorentz function and (b) The splitting in the spin-singlet IEX peak in (a) as 3.5 K to 50 K.

Furthermore, the pump power dependence of IEXs is also probed, as shown in figure 4.10 and 4.11; the PL emission is taken in another region on the HS1. With the increase in laser power, the split in energy between the spin-triplet and the spin-singlet IEX is decreasing. There was a significant blueshift in the spin-triplet IEX and the spin-singlet IEX due to the increasing IEX density, as shown in figure 4.10 and 4.11. The spin-triplet IEX shifts ≈ 15 meV from $1 \mu\text{W}$ to 1 mW , and the spin-singlet IEX shifts ≈ 3 meV. This indicates that with increasing power, the spin-triplet IEX density increases more than the spin-singlet IEX density because spin-triplet IEX has the lowest energy configuration. Increasing the density of IEXs with increased laser power also increases the dipole-dipole interactions because of the spatial separation of electron and hole in IEX; they have a permanent dipole moment [68, 29]. Moreover, as previously reported by Miller et al. [69], and Rivera et al. [70], the intensity of the spin-triplet state IEX is more prominent than the spin-singlet state IEX at low pump power excitations because the lower spin-split band of MoSe_2 fills first, which matches our results as seen in the figure 4.10.

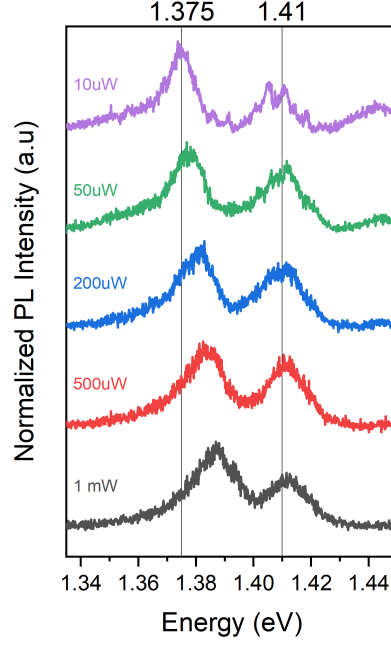


Figure 4.10: The IEX measured in HS1 with different laser powers, the lines added as a reference to follow the blueshift.

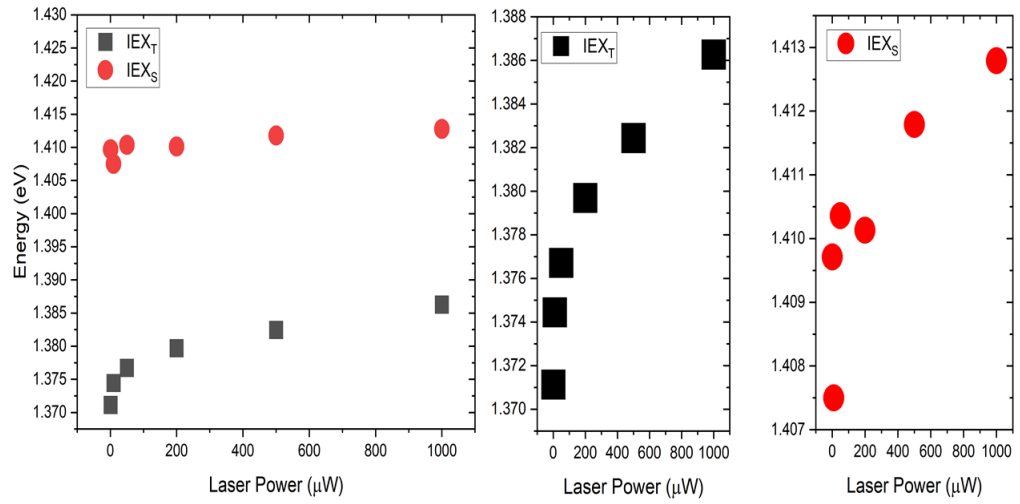


Figure 4.11: The IEX peaks positions measured in HS1 vs. Laser power graph to observe the evolution of blueshift.

The lifetime of the spin-singlet state of the IEX was also measured for the second heterostructure (HS2). The optical microscope image of the HS can be seen in figure 4.12. This HS is stacked at approximately 57 degrees like the HS1. In figure 4.13 (a), the PL result of the IEX can be seen; the intensity of the spin-triplet state is lower than the spin-singlet state. They are separated from each other with an energy difference of ≈ 22 meV. The lifetime of the spin-singlet state IEX is measured with the help of a bandpass filter; the spin-singlet state IEX is isolated.

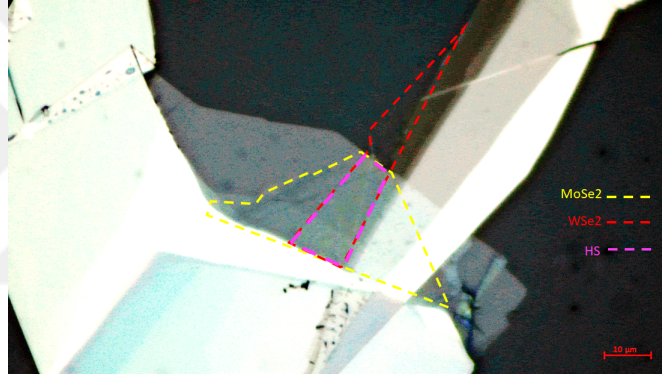
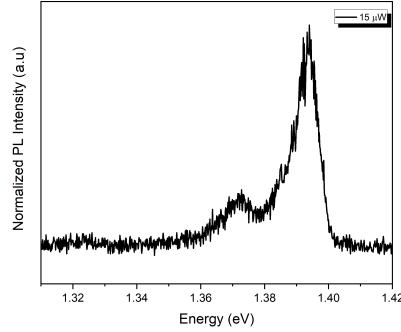


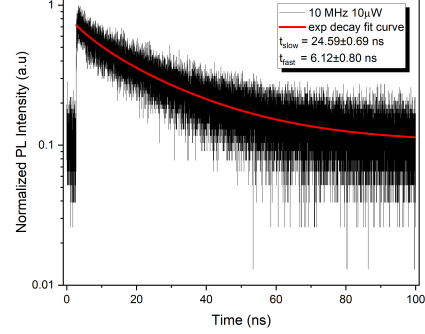
Figure 4.12: The optical microscope image of the second heterostructure.

The oscillator strength of the IEX is a few orders of magnitude lower than the intralayer exciton; hence, the recombination lifetime of IEX is longer than the intralayer exciton [10]. The previously reported data shows that the lifetime of the IEXs can be in the range of a few ns to μ s [68, 69, 70, 71, 72]. Moreover, the lifetime of IEX can be changed for many reasons, such as external electric and magnetic fields, twist angle between the MLs, temperature, pump power, and optical cavities [10]. Therefore, lifetime can be measured differently, even for the same kind of HSs.

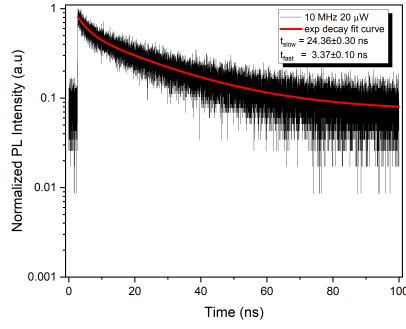
As seen in figure 4.13 (b),(c), and (d), the lifetime measurements are done with different powers to see the power dependence on IEX lifetime. At 10 μ W and 20 μ W, the slow component of the decay is around 24 ns with the indicated errors; the lifetime is close to the previously reported data [73], the fast component of decay seems to decrease from 6 to 3 ns for 10 μ W and 20 μ W respectively. They



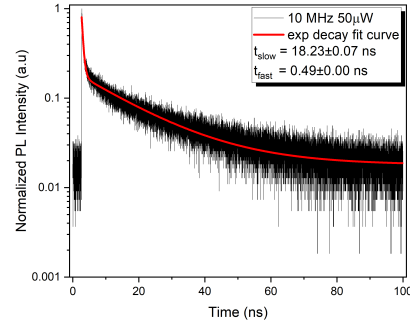
(a)



(b)



(c)



(d)

Figure 4.13: (a) IEX PL Peak and (b,c,d) The Lifetime measurements of the spin-singlet IEX peak resonates around 1.39 eV.

are fitted with the biexponential decay equation in 4.1,

$$y = y_0 + A_1 e^{-x/t_1} + A_2 e^{-x/t_2} \quad (4.1)$$

where y_0 is the offset, A_1 and A_2 are the amplitudes, t_1 and t_2 are the time constants. When the laser power increases to 50 μW , the slow component of the decaying of the spin-singlet IEX decreases to approximately 18 ns, and the fast component decreases to 0.49 ns. The fast and slow components of the decay of the IEX cannot be associated with a specific decay mechanism only. However, it can safely be commented that both nonradiative and radiative recombination affect lifetimes' fast and slow components. With that, some particular recombination mechanisms affect one component more than the other; for example, the

fast decay component decreases significantly with increasing pump power, and the slow component stays almost the same. The reason is that the fast decay component of the decay mechanism is heavily dominated by Auger-assisted annihilation (exciton-exciton annihilation) [74]. The exciton-exciton annihilation is nonradiative recombination; hence, the lifetime is expected to decrease with increasing density of excitons [75, 74]. In figure 4.14, the decrease in the lifetime's slow and fast components is seen, while the slow component does not decrease too much; on the contrary, the fast component seems to decrease significantly. Hence, the results are consistent with previously reported data [74].

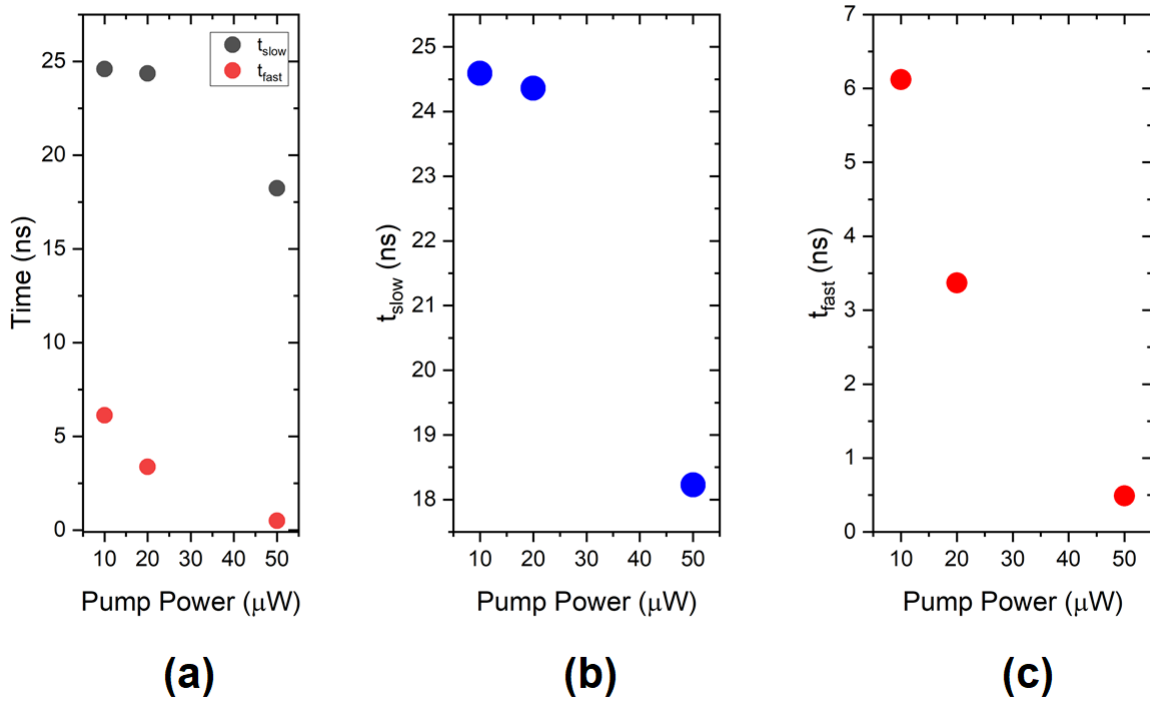
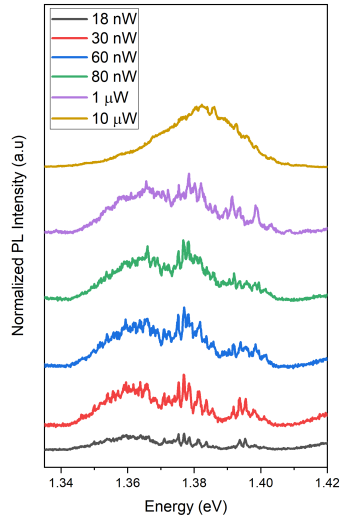
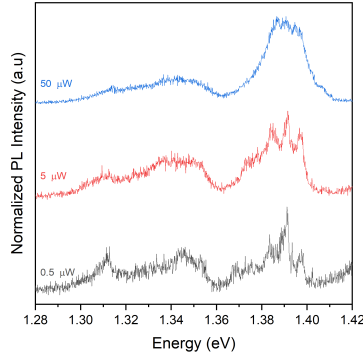


Figure 4.14: (a) Lifetimes of IEX's slow and fast components in figure 4.13 (a) vs. pump power, (b) Slow time component of the lifetimes of the IEX vs. pump power and (c) Fast time component of the lifetimes of the IEX vs. pump power

The moiré trapping sites were also formed in HS2, and the moiré localized sharp and narrow PL emission peaks were observed under low pump power and temperature as observed in the previous study of Seyler et al. [76]. As seen in figure 4.15, with the increasing power, the moiré trapped localized IEXs are changing into delocalized IEXs. The sharp peaks belong to the PLs of the trapped IEXs. The pump power dependence of the $MoSe_2-WSe_2$ moiré IEXs is explored in the study of Förg et al. [77], it has been shown that with the increasing pump power, the intensity of moiré IEXs are decreasing as in our case same results are obtained.



(a)



(b)

Figure 4.15: (a, b) IEX region PL results with changing laser power obtained from different regions of the HS2 with visible Moiré trapped IEX.

4.2 Discussion

In this thesis, we aimed to study the photophysics of IEXs in $MoSe_2-WSe_2$ TMD heterostructures coupled to FP microcavities. By reaching the strong coupling regime of cavity quantum electrodynamics, we targeted the formation of interlayer exciton polariton in these structures.

The TMDs are unique materials that provide an excellent medium to work many-body physics. The direct band gap in their monolayers allows them to have excitons with high binding energies. Moreover, stacking TMD monolayers on top of each other enables the formation of long-lived IEXs. Because of the spatial charge separation of the electron and the hole, IEXs have longer lifetimes, which makes them ideal candidates for exciton-polariton formation studies.

We have fabricated TMD heterostructures inside the FP microcavities using the combination of well-known dry transfer method and the noninvasive, cost-effective edge identification method. Then, we performed both room and low-temperature PL spectroscopy. Firstly, we investigated the temperature dependence of the IEX PL emission. Our findings showed that an increase in temperature significantly impacts the spin-triplet IEX more than the spin-singlet IEX. The intensity of spin-triplet IEX diminishes at temperatures above 30 K, while the spin-singlet IEX remains unaffected. Also, the pump power dependence of the IEX showed that an increase in laser power induces a blueshift in both spin states of the IEX. The spin-triplet IEX exhibits a 15 meV blueshift, whereas the spin-singlet IEX shows a 3 meV blueshift in the same pump power range. This difference is attributed to the lower energy of the spin-triplet IEX, which leads to a higher density of interlayer excitons with the increasing pump power, subsequently enhancing dipole-dipole interactions and resulting in the observed blueshift. The PL emission dynamics of IEXs have also been studied, and the spontaneous emission lifetime of the IEXs was measured. The lifetime's fast and slow decay components have been identified within the corresponding time constants of a few and a few tens of nanoseconds.

We also emphasize that an approximately 9 meV splitting on the spin-singlet IEX PL emission was observed in one of the studied emitters, which might be attributed to exciton-polariton formation at $k_{\parallel}$ when it is equal to zero. However, to safely attribute this splitting in the energy of the PL peak of the IEX to the exciton-polariton formation, one needs to perform the whole angle-resolved PL measurement. Previous reports have documented exciton-photon coupling in low-Q cavities with intralayer excitons [21], exhibiting prominent Rabi splitting energies. Due to the significantly lower oscillator strength of IEXs compared to intralayer excitons, lower Rabi splitting energies are expected, as discussed in Chapter 4.

Future research can focus on measuring the whole angle-resolved spectrum of the coupling of the IEX with light to confirm exciton-polariton formation. Modifying the cavity design to achieve high-Q cavities is also possible. Additionally, a comprehensive investigation of the effects of these microcavities on both spin-triplet and spin-singlet states of IEXs can be investigated.

Bibliography

- [1] Q. H. Wang, K. Kalantar-Zadeh, A. Kis, J. N. Coleman, and M. S. Strano, “Electronics and optoelectronics of two-dimensional transition metal dichalcogenides,” *Nature Nanotechnology*, vol. 7, pp. 699–712, Nov 2012.
- [2] R. Wang, Y. Yu, S. Zhou, H. Li, H. Wong, Z. Luo, L. Gan, and T. Zhai, “Strategies on phase control in transition metal dichalcogenides,” *Advanced Functional Materials*, vol. 28, no. 47, p. 1802473, 2018.
- [3] A. Splendiani, L. Sun, Y. Zhang, T. Li, J. Kim, C.-Y. Chim, G. Galli, and F. Wang, “Emerging photoluminescence in monolayer mos₂,” *Nano Letters*, vol. 10, pp. 1271–1275, Apr 2010.
- [4] K. F. Mak, C. Lee, J. Hone, J. Shan, and T. F. Heinz, “Atomically thin mos₂: A new direct-gap semiconductor,” *Phys. Rev. Lett.*, vol. 105, p. 136805, Sep 2010.
- [5] R. Roldán, J. A. Silva-Guillén, M. P. López-Sancho, F. Guinea, E. Cappelluti, and P. Ordejón, “Electronic properties of single-layer and multilayer transition metal dichalcogenides mx₂ (m = mo, w and x = s, se),” *Annalen der Physik*, vol. 526, no. 9-10, pp. 347–357, 2014.
- [6] P. Tonndorf, R. Schmidt, and P. Bottger, “Photoluminescence emission and raman response of monolayer mos₂, mose₂ and wse₂,” *Reflectance Confocal Microscopy of Cutaneous Tumors, Second Edition*, vol. 21, 2017.

- [7] Y. Guo, C. Liu, Q. Yin, C. Wei, S. Lin, T. B. Hoffman, Y. Zhao, J. H. Edgar, Q. Chen, S. P. Lau, J. Dai, H. Yao, H.-S. P. Wong, and Y. Chai, “Distinctive in-plane cleavage behaviors of two-dimensional layered materials,” *ACS Nano*, vol. 10, pp. 8980–8988, Sep 2016.
- [8] D. Xiao, G.-B. Liu, W. Feng, X. Xu, and W. Yao, “Coupled spin and valley physics in monolayers of mos2 and other group-vi dichalcogenides,” *Physical review letters*, vol. 108, p. 196802, May 2012.
- [9] A. Chernikov, T. C. Berkelbach, H. M. Hill, A. Rigosi, Y. Li, B. Aslan, D. R. Reichman, M. S. Hybertsen, and T. F. Heinz, “Exciton binding energy and nonhydrogenic rydberg series in monolayer ws_2 ,” *Phys. Rev. Lett.*, vol. 113, p. 076802, Aug 2014.
- [10] Y. Jiang, S. Chen, W. Zheng, B. Zheng, and A. Pan, “Interlayer exciton formation, relaxation, and transport in tmd van der waals heterostructures,” *Light: Science & Applications*, vol. 10, p. 72, Apr 2021.
- [11] D. Huang, J. Choi, C.-K. Shih, and X. Li, “Excitons in semiconductor moiré superlattices,” *Nature Nanotechnology*, vol. 17, pp. 227–238, Mar 2022.
- [12] Z. Li, X. Lu, D. F. Cordovilla Leon, Z. Lyu, H. Xie, J. Hou, Y. Lu, X. Guo, A. Kaczmarek, T. Taniguchi, K. Watanabe, L. Zhao, L. Yang, and P. B. Deotare, “Interlayer exciton transport in mose2/wse2 heterostructures,” *ACS Nano*, vol. 15, pp. 1539–1547, Jan 2021.
- [13] X. Liu, T. Galfsky, Z. Sun, F. Xia, E.-c. Lin, Y.-H. Lee, S. Kéna-Cohen, and V. M. Menon, “Strong light–matter coupling in two-dimensional atomic crystals,” *Nature Photonics*, vol. 9, pp. 30–34, Jan 2015.
- [14] C. Schneider, M. M. Glazov, T. Korn, S. Höfling, and B. Urbaszek, “Two-dimensional semiconductors in the regime of strong light-matter coupling,” *Nature Communications*, vol. 9, p. 2695, Jul 2018.
- [15] J. Henrie, S. Kellis, S. M. Schultz, and A. Hawkins, “Electronic color charts for dielectric films on silicon,” *Opt. Express*, vol. 12, pp. 1464–1469, Apr 2004.

- [16] H. Korkut, “Photophysics of quantum emitters in hexagonal boron nitride,” Master’s thesis, Bilkent University, 2022.
- [17] L. Novotny, “Strong coupling, energy splitting, and level crossings: A classical perspective,” *American Journal of Physics*, vol. 78, pp. 1199–1202, 11 2010.
- [18] K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva, and A. A. Firsov, “Electric field effect in atomically thin carbon films,” *Science*, vol. 306, no. 5696, pp. 666–669, 2004.
- [19] R. K. Goyal, *Nanomaterials and Nanocomposites: Synthesis, Properties, Characterization Techniques, and Applications (1st ed.)*. CRC Press, 2017.
- [20] I. Sarpkaya, “4 - optical properties of semiconducting transition metal dichalcogenide materials,” in *Synthesis, Modeling, and Characterization of 2D Materials, and Their Heterostructures* (E.-H. Yang, D. Datta, J. Ding, and G. Hader, eds.), Micro and Nano Technologies, pp. 57–75, Elsevier, 2020.
- [21] X. Liu and V. M. Menon, “Control of light-matter interaction in 2d atomic crystals using microcavities,” *IEEE Journal of Quantum Electronics*, vol. 51, no. 10, pp. 1–8, 2015.
- [22] V. Ardizzone, L. D. Marco, M. D. Giorgi, L. Dominici, D. Ballarini, and D. Sanvitto, “Emerging 2d materials for room-temperature polaritonics,” *Nanophotonics*, vol. 8, no. 9, pp. 1547–1558, 2019.
- [23] S. Manzeli, D. Ovchinnikov, D. Pasquier, O. V. Yazyev, and A. Kis, “2d transition metal dichalcogenides,” *Nature Reviews Materials*, vol. 2, p. 17033, Jun 2017.
- [24] D. Voiry, A. Mohite, and M. Chhowalla, “Phase engineering of transition metal dichalcogenides,” *Chem. Soc. Rev.*, vol. 44, pp. 2702–2712, 2015.
- [25] K. Kang, S. Chen, and E.-H. Yang, “12 - synthesis of transition metal dichalcogenides,” in *Synthesis, Modeling, and Characterization of 2D Materials, and Their Heterostructures* (E.-H. Yang, D. Datta, J. Ding, and G. Hader, eds.), Micro and Nano Technologies, pp. 247–264, Elsevier, 2020.

- [26] K. S. Novoselov, D. Jiang, F. Schedin, T. J. Booth, V. V. Khotkevich, S. V. Morozov, and A. K. Geim, “Two-dimensional atomic crystals,” *Proceedings of the National Academy of Sciences*, vol. 102, no. 30, pp. 10451–10453, 2005.
- [27] L. Zhang, Z. Zhang, F. Wu, D. Wang, R. Gogna, S. Hou, K. Watanabe, T. Taniguchi, K. Kulkarni, T. Kuo, S. R. Forrest, and H. Deng, “Twist-angle dependence of moiré excitons in $\text{ws}_2/\text{mose}_2$ heterobilayers,” *Nature Communications*, vol. 11, p. 5888, Nov 2020.
- [28] P. Merkl, F. Mooshammer, S. Brem, A. Girnghuber, K.-Q. Lin, L. Weigl, M. Liebich, C.-K. Yong, R. Gillen, J. Maultzsch, J. M. Lupton, E. Malic, and R. Huber, “Twist-tailoring coulomb correlations in van der waals homobilayers,” *Nature Communications*, vol. 11, p. 2167, May 2020.
- [29] P. Rivera, H. Yu, K. L. Seyler, N. P. Wilson, W. Yao, and X. Xu, “Interlayer valley excitons in heterobilayers of transition metal dichalcogenides,” *Nature Nanotechnology*, vol. 13, pp. 1004–1015, Nov 2018.
- [30] S. Ovesen, S. Brem, C. Linderälv, M. Kuisma, T. Korn, P. Erhart, M. Selig, and E. Malic, “Interlayer exciton dynamics in van der waals heterostructures,” *Communications Physics*, vol. 2, p. 23, Feb 2019.
- [31] Y. Liu, Y. Gao, S. Zhang, J. He, J. Yu, and Z. Liu, “Valleytronics in transition metal dichalcogenides materials,” *Nano Research*, vol. 12, pp. 2695–2711, Nov 2019.
- [32] E. C. Regan, D. Wang, E. Y. Paik, Y. Zeng, L. Zhang, J. Zhu, A. H. MacDonald, H. Deng, and F. Wang, “Emerging exciton physics in transition metal dichalcogenide heterobilayers,” *Nature Reviews Materials*, vol. 7, pp. 778–795, Oct 2022.
- [33] J. Yu, X. Kuang, J. Zhong, L. Cao, C. Zeng, J. Ding, C. Cong, S. Wang, P. Dai, X. Yue, Z. Liu, and Y. Liu, “Observation of double indirect interlayer exciton in wse_2/ws_2 heterostructure,” *Opt. Express*, vol. 28, pp. 13260–13268, Apr 2020.

- [34] K. Xu, Y. Xu, H. Zhang, B. Peng, H. Shao, G. Ni, J. Li, M. Yao, H. Lu, H. Zhu, and C. M. Soukoulis, “The role of anderson’s rule in determining electronic, optical and transport properties of transition metal dichalcogenide heterostructures,” *Phys. Chem. Chem. Phys.*, vol. 20, pp. 30351–30364, 2018.
- [35] A. Ciarrocchi, F. Tagarelli, A. Avsar, and A. Kis, “Excitonic devices with van der waals heterostructures: valleytronics meets twistrionics,” *Nature Reviews Materials*, vol. 7, pp. 449–464, Jun 2022.
- [36] L. Zhang, R. Gogna, G. W. Burg, J. Horng, E. Paik, Y.-H. Chou, K. Kim, E. Tutuc, and H. Deng, “Highly valley-polarized singlet and triplet interlayer excitons in van der waals heterostructure,” *Phys. Rev. B*, vol. 100, p. 041402, Jul 2019.
- [37] T. Wang, S. Miao, Z. Li, Y. Meng, Z. Lu, Z. Lian, M. Blei, T. Taniguchi, K. Watanabe, S. Tongay, D. Smirnov, and S.-F. Shi, “Giant valley-zeeman splitting from spin-singlet and spin-triplet interlayer excitons in wse2/mose2 heterostructure,” *Nano Letters*, vol. 20, no. 1, pp. 694–700, 2020. PMID: 31865705.
- [38] H. Fang, Q. Lin, Y. Zhang, J. Thompson, S. Xiao, Z. Sun, E. Malic, S. P. Dash, and W. Wiczorek, “Localization and interaction of interlayer excitons in mose2/wse2 heterobilayers,” *Nature Communications*, vol. 14, p. 6910, Oct 2023.
- [39] E. Blundo, F. Tuzi, S. Cianci, M. Cuccu, K. Olkowska-Pucko, Ł. Kipczak, G. Contestabile, A. Miriametro, M. Felici, G. Pettinari, T. Taniguchi, K. Watanabe, A. Babiński, M. R. Molas, and A. Polimeni, “Localisation-to-delocalisation transition of moiré excitons in wse2/mose2 heterostructures,” *Nature Communications*, vol. 15, p. 1057, Feb 2024.
- [40] E. Liu, E. Barré, J. van Baren, M. Wilson, T. Taniguchi, K. Watanabe, Y.-T. Cui, N. M. Gabor, T. F. Heinz, Y.-C. Chang, and C. H. Lui, “Signatures of moiré trions in wse2/mose2 heterobilayers,” *Nature*, vol. 594, pp. 46–50, Jun 2021.

- [41] S. M. Dutra, *Cavity Quantum Electrodynamics: The Strange Theory of Light in a Box*, pp. 17–67. John Wiley Sons, Ltd, 2004.
- [42] A. H. W. E. Choi, *Handbook of Optical Microcavities*. Jenny Stanford Publishing, 1st ed. ed., 2014.
- [43] G. Panzarini, L. C. Andreani, A. Armitage, D. Baxter, M. S. Skolnick, V. N. Astratov, J. S. Roberts, A. V. Kavokin, M. R. Vladimirova, and M. A. Kaliteevski, “Cavity-polariton dispersion and polarization splitting in single and coupled semiconductor microcavities,” *Physics of the Solid State*, vol. 41, pp. 1223–1238, Aug 1999.
- [44] D. Angelakis, *Quantum Simulations with Photons and Polaritons: Merging Quantum Optics with Condensed Matter Physics*. Springer International Publishing AG, 01 2017.
- [45] T. Christopoulos, O. Tsilipakos, G. Sinatkas, and E. E. Kriezis, “On the calculation of the quality factor in contemporary photonic resonant structures,” *Opt. Express*, vol. 27, pp. 14505–14522, May 2019.
- [46] B. Chen, F. Tay, and C. Iliescu, “Development of thick film pecvd amorphous silicon with low stress for mems applications,” *Proceedings of SPIE - The International Society for Optical Engineering*, vol. 7269, 12 2008.
- [47] T. Lazerand and D. Lishan, “Silicon nitride for mems applications: Lpcvd and pecvd process comparison,” *MEMS Journal*, 01 2014.
- [48] H. Deng, H. Haug, and Y. Yamamoto, “Exciton-polariton bose-einstein condensation,” *Rev. Mod. Phys.*, vol. 82, pp. 1489–1537, May 2010.
- [49] S. Ghosh, R. Su, J. Zhao, A. Fieramosca, J. Wu, T. Li, Q. Zhang, F. Li, Z. Chen, T. Liew, D. Sanvitto, and Q. Xiong, “Microcavity exciton polaritons at room temperature,” *Photonics Insights*, vol. 1, p. R04, 1 2022.
- [50] D. Novko and V. Despoja, “Cavity exciton polaritons in two-dimensional semiconductors from first principles,” *Phys. Rev. Res.*, vol. 3, p. L032056, Sep 2021.

- [51] A. Kavokin and G. Malpuech, *Cavity Polaritons*. Thin Films and Nanostructures, Elsevier Science, 2003.
- [52] J. G. Pascual, *Polaritonic Chemistry Manipulating Molecular Structure Through Strong Light-Matter Coupling*. PhD thesis, Universidad Autónoma de Madrid, 2020.
- [53] D. S. Dovzhenko, S. V. Ryabchuk, Y. P. Rakovich, and I. R. Nabiev, “Light–matter interaction in the strong coupling regime: configurations, conditions, and applications,” *Nanoscale*, vol. 10, pp. 3589–3605, 2018.
- [54] X. Liu, *Control of Exciton Photon Coupling in Nano-structures*. PhD thesis, City University of New York, 2014.
- [55] C. Schneider, K. Winkler, M. D. Fraser, M. Kamp, Y. Yamamoto, E. A. Ostrovskaya, and S. Höfling, “Exciton-polariton trapping and potential landscape engineering,” *Reports on Progress in Physics*, vol. 80, p. 016503, nov 2016.
- [56] J. Hopfield, “Theory of the contribution of excitons to the complex dielectric constant of crystals,” *Physical Review - PHYS REV X*, vol. 112, pp. 1555–1567, 12 1958.
- [57] R. Holmes and S. Forrest, “Strong exciton–photon coupling in organic materials,” *Organic Electronics*, vol. 8, no. 2, pp. 77–93, 2007. Coupled States of Excitons, Photons, and Plasmons in Organic Structures.
- [58] E. Cotta and P. Roma, “Determination of oscillator strength of confined excitons in a semiconductor microcavity,” *Condensed Matter Physics*, vol. 17, 07 2014.
- [59] C. Qian, M. Troue, J. Figueiredo, P. Soubelet, V. Villafañe, J. Beierlein, S. Klemmt, A. V. Stier, S. Höfling, A. W. Holleitner, *et al.*, “Lasing of moiré trapped mose2/wse2 interlayer excitons coupled to a nanocavity,” *Science Advances*, vol. 10, no. 2, p. eadk6359, 2024.

- [60] H. Korkut and I. Sarpkaya, “Controlling the photoluminescence of quantum emitters in hexagonal boron nitride by external magnetic fields,” *2D Materials*, vol. 10, no. 1, p. 015004, 2022.
- [61] *Picoquant HydraHarp 400, Picosecond Histogram Accumulating Real-time Processor, Time-Correlated Single Photon Counting System with USB Interface, User’s Manual and Technical Data, Version 3.0.0.4.*
- [62] H. Yu, G.-B. Liu, and W. Yao, “Brightened spin-triplet interlayer excitons and optical selection rules in van der waals heterobilayers,” *2D Materials*, vol. 5, no. 3, p. 035021, 2018.
- [63] A. Kormányos, V. Zólyomi, N. D. Drummond, and G. Burkard, “Spin-orbit coupling, quantum dots, and qubits in monolayer transition metal dichalcogenides,” *Physical Review X*, vol. 4, no. 1, p. 011034, 2014.
- [64] P. Rivera, J. R. Schaibley, A. M. Jones, J. S. Ross, S. Wu, G. Aivazian, P. Klement, K. Seyler, G. Clark, N. J. Ghimire, J. Yan, D. G. Mandrus, W. Yao, and X. Xu, “Observation of long-lived interlayer excitons in monolayer mose2–wse2 heterostructures,” *Nature Communications*, vol. 6, p. 6242, Feb 2015.
- [65] M. A. Durmuş and I. Sarpkaya, “Quantum beats between spin-singlet and spin-triplet interlayer exciton transitions in wse2–mose2 heterobilayers,” *Nano Letters*, vol. 24, no. 19, pp. 5767–5773, 2024. PMID: 38639575.
- [66] A. T. Hanbicki, H.-J. Chuang, M. R. Rosenberger, C. S. Hellberg, S. V. Sivaram, K. M. McCreary, I. I. Mazin, and B. T. Jonker, “Double indirect interlayer exciton in a mose2/wse2 van der waals heterostructure,” *ACS Nano*, vol. 12, no. 5, pp. 4719–4726, 2018. PMID: 29727170.
- [67] L. Zhang, R. Gogna, G. W. Burg, J. Horng, E. Paik, Y.-H. Chou, K. Kim, E. Tutuc, and H. Deng, “Highly valley-polarized singlet and triplet interlayer excitons in van der waals heterostructure,” *Physical Review B*, vol. 100, no. 4, p. 041402, 2019.

- [68] P. Nagler, F. Mooshammer, J. Kunstmann, M. V. Ballottin, A. Mitioglu, A. Chernikov, A. Chaves, F. Stein, N. Paradiso, S. Meier, G. Plechinger, C. Strunk, R. Huber, G. Seifert, D. R. Reichman, P. C. M. Christianen, C. Schüller, and T. Korn, “Interlayer excitons in transition-metal dichalcogenide heterobilayers,” *physica status solidi (b)*, vol. 256, no. 12, p. 1900308, 2019.
- [69] B. Miller, A. Steinhoff, B. Pano, J. Klein, F. Jahnke, A. Holleitner, and U. Wurstbauer, “Long-lived direct and indirect interlayer excitons in van der waals heterostructures,” *Nano letters*, vol. 17, no. 9, pp. 5229–5237, 2017.
- [70] P. Rivera, J. R. Schaibley, A. M. Jones, J. S. Ross, S. Wu, G. Aivazian, P. Klement, K. Seyler, G. Clark, N. J. Ghimire, J. Yan, D. G. Mandrus, W. Yao, and X. Xu, “Observation of long-lived interlayer excitons in monolayer mose2–wse2 heterostructures,” *Nature Communications*, vol. 6, p. 6242, Feb 2015.
- [71] P. Nagler, M. V. Ballottin, A. A. Mitioglu, F. Mooshammer, N. Paradiso, C. Strunk, R. Huber, A. Chernikov, P. C. Christianen, C. Schüller, *et al.*, “Giant magnetic splitting inducing near-unity valley polarization in van der waals heterostructures,” *Nature communications*, vol. 8, no. 1, p. 1551, 2017.
- [72] C. Jiang, W. Xu, A. Rasmita, Z. Huang, K. Li, Q. Xiong, and W.-b. Gao, “Microsecond dark-exciton valley polarization memory in two-dimensional heterostructures,” *Nature communications*, vol. 9, no. 1, p. 753, 2018.
- [73] M. A. Durmuş, K. Demiralay, M. M. Khan, Ş. E. Atalay, and I. Sarpkaya, “Prolonged dephasing time of ensemble of moiré-trapped interlayer excitons in wse2-mose2 heterobilayers,” *npj 2D Materials and Applications*, vol. 7, p. 65, Sep 2023.
- [74] C.-S. Cai, W.-Y. Lai, P.-H. Liu, T.-C. Chou, R.-Y. Liu, C.-M. Lin, S. Gwo, and W.-T. Hsu, “Ultralow auger-assisted interlayer exciton annihilation in ws2/wse2 moire heterobilayers,” *Nano Letters*, vol. 24, no. 9, pp. 2773–2781, 2024.

- [75] D. Erkensten, S. Brem, and E. Malic, “Exciton-exciton interaction in transition metal dichalcogenide monolayers and van der Waals heterostructures,” *Physical Review B*, vol. 103, no. 4, p. 045426, 2021.
- [76] K. L. Seyler, P. Rivera, H. Yu, N. P. Wilson, E. L. Ray, D. G. Mandrus, J. Yan, W. Yao, and X. Xu, “Signatures of moiré-trapped valley excitons in $\text{MoSe}_2/\text{WSe}_2$ heterobilayers,” *Nature*, vol. 567, no. 7746, pp. 66–70, 2019.
- [77] M. Förg, A. S. Baimuratov, S. Y. Kruchinin, I. A. Vovk, J. Scherzer, J. Förste, V. Funk, K. Watanabe, T. Taniguchi, and A. Högele, “Moiré excitons in MoSe_2 - WSe_2 heterobilayers and heterotrilayers,” *Nature communications*, vol. 12, no. 1, p. 1656, 2021.

Appendix A

Two Oscillator Model

When the two oscillators are not coupling with each other ($\kappa = 0$, see figure A.1, where κ is the coupling strength), the eigenfrequencies can be calculated by the following formula in the equationA.1[17]. Those oscillators should be considered as the cavity photon and the exciton in the exciton-polariton case.

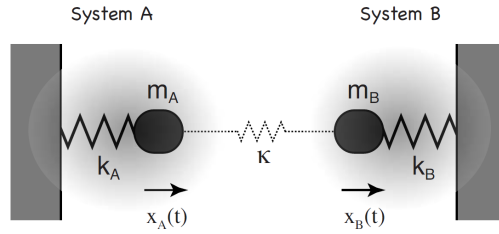


Figure A.1: Illustration of strong coupling, mechanical oscillators with masses m_i and spring constants k_i demonstrate frequency shifts and characteristic frequency splitting as a result of their coupling (κ) [17].

$$w_A = \sqrt{\frac{k_A}{m_A}}, w_B = \sqrt{\frac{k_B}{m_B}} \quad (\text{A.1})$$

When the two oscillators are coupling ($\kappa \neq 0$), there will be motion between them. The equation of the motions can be found in equation A.2 & 2.3 [17].

$$m_A \ddot{x} + k_A x_A + \kappa(x_A + x_B) = 0 \quad (\text{A.2})$$

$$m_B \ddot{x} + k_B x_B + \kappa(x_A + x_B) = 0 \quad (\text{A.3})$$

The desired solution should be composed as $x_i(t) = x_i^0 \exp[-i w_{\pm} t]$, here the $w_{\pm}$ represents the two new eigenstates of the polariton[17]. With this approach, the equation A.1 can be written as matrix as $M[x_A^0, x_B^0]^T = 0$. The solution for new eigenfrequencies for polariton branches can be calculated as in the equation A.4; this solution is only valid when the $\det[M]=0$, [17]. Otherwise, the solution would only be valid for trivial solutions.

$$w_{\pm}^2 = \frac{1}{2}[w_A^2 + w_B^2 \pm \sqrt{(w_A^2 - w_B^2)^2 + 4\Gamma^2 w_A w_B}] \quad (\text{A.4})$$

Where in equation A.4, the spring constants are appointed as $k_A = k_0$ and $k_B = k_0 + \Delta k$, and masses are $m_A = m_B = m_0$. Δk represents the increase from $-k_0$ to k_0 . While the w_B increases from 0 to $\sqrt{2}w_0$, w_A remains constant. The oscillator curves meet at $\Delta k = 0$ when no coupling occurs; in the case of coupling, anticrossing between oscillator curves appears; this anticrossing equals frequency splitting as in equation A.5.

$$[w_+ - w_-] = \Gamma \quad (\text{A.5})$$

When $\kappa \neq 0$, the frequencies of the eigenfrequencies become as in the equation A.6.

$$w_A = \sqrt{\frac{k_A + \kappa}{m_A}}, w_B = \sqrt{\frac{k_B + \kappa}{m_B}} \quad (\text{A.6})$$

In the case of strong coupling, an anticrossing behavior occurs between two new eigenstates; the frequency splitting can be calculated as follows A.7.

$$\Gamma = \frac{\sqrt{\kappa/m_A}\sqrt{\kappa/m_B}}{\sqrt{w_A w_B}} \quad (\text{A.7})$$

Since an increase in one leads to a corresponding increase in the other and vice versa, there is a linearly proportional relation between frequency splitting and the coupling strength ($\Gamma \propto \kappa$) [17].

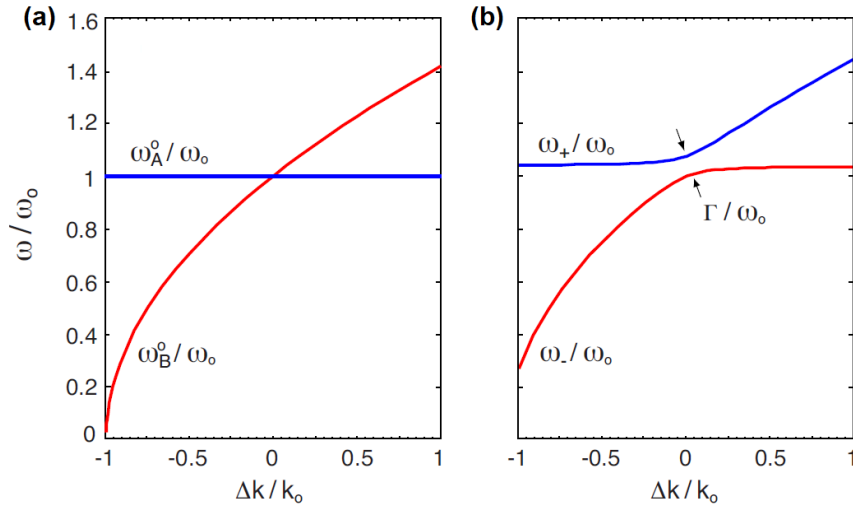


Figure A.2: Eigenfrequencies of two uncoupled oscillators $\kappa=0$, masses are equal and spring constants are k_0 and $k_0+\Delta k$. (b) When coupling strength $\kappa=0.08k_0$ the anticrossing occurs [17]

With the consideration of damping in the system, when the frictional terms $\gamma_A \dot{x}_A$ and $\gamma_B \dot{x}_B$ are added to the system, the eigenvalues become complex frequency eigenvalues[17]. The imaginary part of these complex frequency eigenvalues is the line widths. When there is robust damping, the split in frequency becomes harder to observe. Hence, the splitting in these frequencies has to be higher than the total amount of the linewidths for a strong coupling to be achieved[17].

$$\frac{\Gamma}{\frac{\gamma_A}{m_A} + \frac{\gamma_B}{m_B}} > 1 [17] \quad (\text{A.8})$$