List of Figures

Figures	*Title*	*page*
1.1	The rapidly increasing number of publications on 2D materials with each passing year. (Source: Scopus)	1
1.2	Different classes of 2D materials varying from insulating to superconducting behavior. (Source: Kang et al., 2015)	2
1.3	Research done on 2D materials in different disciplines. (Source: Scopus)	3
1.4	The different spectral range covered by various 2D materials. (Source: Xia et al., 2014)	3
1.5	Milestones achieved in the field of 2D materials starting from graphene.	4
1.6	The first discovered 2D material, graphene, in 0D, 1D, and 3D forms. (Source: Geim and Navoselov, 2007)	5
1.7	Klein tunneling effect in graphene. (Source: Zhang et al., 2014)	6
1.8	(a) Schematic illustration and (b) resistivity changes upon exposure to different gas molecules on the graphene surface. (Source: Schedin et al., 2007)	7
1.9	(a) Schematic illustration of double-layer graphene photodetector. (b) Band alignment and photoinduced carrier movement between the bottom and top layer of graphene. (c) Gate tunable photocurrent with varying illuminating power. (d) Photoresponsivity at different values of laser power. (Source: Liu et al., 2014)	8
1.10	Applications of graphene in different fields. (Source: Barkan, 2019)	8
1.11	Different families of 2D materials beyond graphene. (Source: https://cen.acs.org/articles/95/i22/2-D-materials-beyond-graphene.html)	9
1.12	Schematic diagram of the hexagonal boron nitride crystal structure. (Source: https://www.substech.com/)	10
1.13	Side and top views of the crystal structure of few-layer phosphorene. (Source: Liu et al., 2014)	11
1.14	Schematic illustration of crystal structures of MAX phases. (Source: Dhakal et al., 2015)	11
1.15	Schematic view of borophene. The blue circles represent the atoms shared by three units. (Source: Piazza et al., 2014)	12
1.16	Different phases of TMDs due to different stacking positions of transition metal and chalcogen atoms. (a) 1H, (b) 1T, (c) distorted 1T, (d) 2H, and (e) 3R phases. (Source: Voiry et al., 2015)	13
1.17	The 3D schematic illustration of the crystal structure of MoS_2. Each layer of MoS_2 is 6.5 A˚ thick. (Source: Radisavljevic et al., 2011)	14
1.18	Year-wise number of publications in the field of optoelectronics for most widely used 2D materials. (Source: Thakar et al., 2020)	15
1.19	(a) Schematic illustration of monolayer MoS_2 based transistor. (b) AFM image and (c) height profile along the solid red line of mechanically exfoliated MoS_2 flake on SiO_2/Si substrate. (d) The Transfer characteristics depicting a very high value of on/off ratio (> 10^8) for V_{ds} = 500 mV. (e) Output characteristics at different top gate voltage. (Source: Radisavljevic et al., 2011)	15
1.20	(a) 3D schematic illustration of ultrasensitive MoS_2 photodetector. (b) Optical image of MoS_2 based transistor. (c) Photo-responsivity of the device depicting a value as high as 880 A/W under light irradiation of 150 pW. (d) Output characteristics of the device in the dark and under various light irradiation intensities. (Source: Lopez-Sanchez et al., 2013)	16
1.21	Comparison of sensing ability of 2- and 5- layer MoS_2 film upon exposure of varying concentration of (a) NH_3 and (b) NO_2. (Source: Late et al., 2013)	17
1.22	Schematic representation of MoS_2 based gas sensor. (b) The sensitivity of the device with varying concentrations of NO_2 at room temperature. (c) A liner fitting of between sensitivity and different concentrations of NO_2 using Langmuir's model. (d) Comparison of sensing performance of the device at room temperature and 100 ˚C. (e) Cycling text at 120	18

ppb NO_2 concentration to evaluate the stability of the device. (f) The selectivity of the device to different gas analytes. The inset shows the sensing response under mixed gas environment. (Source: Cho et al., 2015)
1.23 Different kinds of band alignments at the heterointerfaces. 19
1.24 (a) The classification of 2D materials based on their properties. (b-f) multifunctional heterostructures formed by combining different dimensional materials. (Source: Liu et al., 2016) 20
1.25 Schematic illustration of 0D materials, including fullerenes, tiny organic molecules, and quantum dots. (Source: Jariwala et al., 2016) 21
1.26 Schematic illustration of the MoS_2-graphene quantum dots based photodetector. (b) I_{ds}-V_{ds} characteristics of in presence and absence of 405 nm light irradiation for the only MoS_2 and MoS_2-graphene quantum dots based photodetector. (c) Gate tenability of photocurrent. (d) Photoresponsivity as a function of incident optical irradiation. (Source: Chen et al., 2015) 22
1.27 Band alignment at the MoS_2/PbS heterointerface. The photoinduced charge carriers get separated at the interface. (b) Comparison of photoresponsivity of only MoS_2 film and MoS_2/PbS hybrids. The inset shows the schematic representation of the MoS_2/PbS hybrid device. (Source: Kufer et al., 2015) 22
1.28 Energy band diagram at the TiO_2/WS_2 heterointerface. Schematic illustration of TiO_2/WS_2 nanohybrids in (b) air and (c) NH_3 molecules. (Source: Qin et al., 2017) 23
1.29 Photograph of Pd-MoS_2 nanocomposite sensing device. (b) SEM image of Pd-MoS_2 hybrids. (c) Sensing response of pristine MoS_2 and Pd-MoS_2 hybrid upon exposure to hydrogen. (d) Sensing response of Pd-MoS_2 hybrids upon exposure to different concentrations of hydrogen. (Source: Kuru et al., 2015) 24
1.30 Schematic illustration of 1D structures, including carbon nanotube, polymers, and Si nanowire. (Source: Jariwala et al., 2016) 25
1.31 Schematic representation of single-walled carbon nanotube/MoS_2 heterostructure. (b) Gate modulated photocurrent at different wavelengths. (Source: Jariwala et al., 2013) 25
1.32 Sensing performance of the MoS_2/carbon nanotube hybrid structure against (a) various concentrations of NO_2 gas and (b) 25 ppb concentration of NO_2 gas. (Source: Deokar et al., 2017) 26
1.33 Schematic diagram of Ag nanowire/WS_2 sensor. (b) The gas-sensing performance of the device against various concentrations of NO_2 at room temperature. (c) Comparison of sensing response of pristine WS_2 and Ag nanowire/WS_2 based gas sensors. (d) Sensing mechanism occurring at Ag nanowire/WS_2 based sensor. (Source: Ko et al., 2016) 26
1.34 Schematic representation of MoS_2/graphene phototransistor. The inset shows the TEM image of the heterostructure. (b) Photocurrent of MoS_2/graphene heterostructure under 650 nm light irradiation at different illumination intensities. (c) Measured photoresponsivity and (d) transient response of the device under 94 μW light illumination. (Source: Chen et al., 2016) 27
1.35 Schematic illustration of (a) top view and (b) side view of graphene–MoS_2–graphene heterostructure. (c) Current-voltage characteristic of the device in the dark and under 514 nm light irradiation. (Source: Yu et al., 2013) 28
1.36 (a) Schematic illustration and (b) SEM image of graphene/MoS_2 heterostructure based gas sensor. The sensitivity of the device under the varying concentration of (c) NO_2 and (d) NH_3 gas molecules. (Source: Cho et al., 2015) 28
1.37 Schematic illustration of 3D structures, including Si, GaAs, and amorphous oxides. (Source: Jariwala et al., 2016) 29
1.38 Schematic illustration of germanium-graphene heterostructure based photodetector. (b) Band alignment at germanium-graphene heterointerface depicting the movement of photoinduced charge carriers. (c) Photoresponsivity and (d) photoconductive gain of the device at different wavelengths under varying illumination intensities. (Source: Yang et al., 2017) 30
1.39 Schematic illustration and photoresponse of MoS_2/ZnO heterostructure based photodiode. The I_D-V_D characteristics without illumination and under 365 nm light irradiation with varying light intensities. (b) The photocurrent at a different value of light irradiation intensities at a different value of drain voltage. (Source: Xue et al., 2016) 31
1.40 3D schematic representation of MoS_2/VO_2 van der Waals heterostructure. (b) Band 32

	alignment at the MoS_2/VO_2 heterointerface when VO_2 is having insulating behavior and after making the transition from insulating to metallic phase. (c) Photoresponsivity as a function of temperature. The photoresponsivity saturates after the phase transition of VO_2 from insulating to metallic. (Source: Oliva et al., 2017)	
1.41	Optical image of graphene/si based gas sensor. (b) Band alignment at graphene/si heterointerface. The barrier height at the interface changes upon NO_2 and NH_3 exposure. (c) Comparison of sensing response in the only graphene and graphene/si based gas sensor upon NO_2 exposure. (d) Comparison of sensing response in the only graphene and graphene/si based gas sensor upon NH_3 exposure. (Source: Singh et al., 2014)	32
1.42	Some of the key application of Heterostructures heaving atleast on 2D component.	34
2.1	Illustration of the mechanical exfoliation process of MoS2. (a) Commercially available crystal of molybdenite (SPI Supplies). (b) Optical image of scotch tape after making contact with the crystal. (c) Repeated peeling off the scotch tape. (d) Further thinning down of MoS2 flakes after repeated peeling process. (e) Heating of the substrate in the air at 100 ˚C prior to removal of tape from the substrate. (f) Optical image of SiO2/Si after the MoS2 exfoliation process.	37
2.2	Optical microscope images of several contacted MoS2 flakes. All patterns were fabricated using the 0.6 μm resolution lens. (a), (b): Flake 1. (c), (d): Flake 2. (e), (f): Flake 3.	38
2.3	Schematic illustration of conventional chemical vapor deposition (CVD) technique for the growth of MoS2 film.	39
2.4	Optical image of chemical vapor deposition (CVD) system.	39
2.5	Working principle of magnetron sputtering technique. (Source: https://www.adnano-tek.com/magnetron-sputtering-deposition-msd.html)	40
2.6	Experimental setup of magnetron sputtering system.	41
2.7	Schematic illustrations of fabrication process of growth process of MoS_2 film by Sputtering coupled with sulfurization technique. (a) The pre-deposited Mo film through DC sputtering technique. (b) Sulfurization of pre-deposited Mo film in sulfur-rich environment. (c) MoS_2 film grown on the substrate.	42
2.8	SEM images and corresponding EDX spectra of grown MoS2 through sputtering coupled with sulfurization technique. (a) Sample 2, (b) sample 3, and (c) sample 7.	43
2.9	Experimental setup of scanning electron microscopy (SEM) system.	44
2.10	Working principle of SEM. (Source: https://en.wikipedia.org/wiki/Scanning_electron microscope)	44
2.11	Working principle of AFM. (Source: http://physics.usask.ca/~chang/homepage/STMAFM/STMAFM.html)	45
2.12	Instrument setup of atomic force microscopy (AFM) technique.	46
2.13	Occurrence of Rayleigh and Raman scattering (Stokes and Anti-Stokes). (Source: https://www.edinst.com/blog/what-is-raman-spectroscopy/)	47
2.14	Instrument setup of Raman spectroscopy. (Courtesy: Material Research Centre, MNIT Jaipur)	48
2.15	Experimental setup of UV/Vis spectrophotometer.	48
2.16	Principle of operation of UV/Vis spectrophotometer.	49
2.17	Schematic illustration of Bragg's x-ray diffraction process	50
2.18	Optical image of the X-Ray diffraction (XRD) system.	50
2.19	Schematic representation of working principle of the XPS technique. (Source: https://en.wikipedia.org/wiki/X-ray_photoelectron_spectroscopy)	51
2.20	Photograph of the X-ray photoelectron spectroscopy system, which is used for this thesis work. (Courtesy: Thin Film Devices & Metrology Section, CSIR-NPL New Delhi)	52
2.21	Schematic representation of the core electrons ejection process during XPS measurement.	52
2.22	Schematic representation of valance electrons ejection process during XPS measurement.	53
2.23	Photograph of thermal evaporation system.	53
2.24	Working principle of thermal evaporation system. (Source: http://www.semicore.com/thin-film-deposition-thermal-evaporation)	54
2.25	Sequential steps followed during the photolithography process.	55
2.26	Photograph of Mask Aligner: Optical Lithography.	55
2.27	Instrument setup for electrical characterization.	56
2.28	Experimental setup of customized gas sensing setup used in this thesis work.	57
3.1	Band-diagram of MoS_2 and WS_2 (a) before making contact and (b) after making contact. Once the contact took place, the Fermi level becomes constant. (c) Type-II band alignment	60

	at MoS_2/WS_2 heterointerface depicting efficient hole transfer from MoS_2 to WS_2. (Source: Hong et al., 2014)	
3.2	Optical microscopy image taken on the stacked MoS_2/Si heterojunction. (c) Raman spectra of the exfoliated FL-MoS_2 obtained using 514nm laser excitation.	61
3.3	AFM characterization. (a) AFM image of the exfoliated FL-MoS_2 on Si and (b) the corresponding height profile along the dotted line.	61
3.4	XPS survey scan of (a) Si, (b) MoS_2, and (c) MoS_2/Si.	62
3.5	The x-ray photoelectron spectroscopy (XPS) spectra for (a) isolated MoS_2 flake, also providing the binding energy separation between Mo 3d core-level and valence band spectra (b) Si substrate, also representing the binding energy separation between Si 2p core-level and valence band spectra and (c) stacked MoS_2/Si heterojunction, showing the binding energy separation between Mo 3d and Si 2p core-levels.	63
3.6	(a) and (b) Ultraviolet photoelectron spectroscopy (UPS) spectra for deposited MoS_2 flake and Si substrate.	64
3.7	Type-II band alignment schematic representation at MoS_2/Si heterojunction.	64
3.8	PL spectra of molecular beam epitaxially grown GaN film.	66
3.9	FE-SEM images of MoS_2 deposited on GaN film (a-c) at the central zone with different magnifications and (d) at the MoS_2/GaN edge.	66
3.10	(a) Raman spectra of MoS_2/GaN interface measured by using a 514 nm laser excitation. (b) XRD patterns of MoS_2/GaN heterojunction.	67
3.11	XPS survey scan of (a) GaN, (b) MoS_2, and (c) MoS_2/GaN.	67
3.12	(a) Mo 3d core-level and valence band spectra obtained from MoS_2 film. (b) Ga 2p core-level and valence band spectra of molecular beam epitaxially grown GaN film. (c) Spectra of Ga 2p and Mo 3d core-level acquired on the stacked MoS_2/GaN heterojunction. Core-levels and valence band spectra are recorded from X-ray photoelectron spectroscopy (XPS) measurement.	68
3.13	(a) S 2p core-level and valence band spectra of MoS_2 Film. (b) Ga 2p and S 2p core-level spectra obtained from stacked MoS_2/GaN heterojunction.	69
3.14	Ultraviolet photoelectron spectroscopy (UPS) spectra of (a) as-grown MoS_2 film and (b) molecular beam epitaxially grown GaN film.	70
3.15	Schematic illustration depicting experimentally derived type-I band alignment at FL-MoS_2/GaN heterojunction.	71
3.16	Comparison of interfacial electronic movement at (a) MoS_2/p-Si and (b) MoS_2/GaN heterojunctions.	72
4.1	(a) Three-dimensional model of FL-MoS_2 photodetector with an experimental setup, (b) Raman spectrum of mechanically exfoliated FL-MoS_2 flake, (c) AFM image of FL-MoS_2 flake on a Si surface forming a heterojunction, and (d) the height profile taken along the dotted line in the AFM image.	75
4.2	(a) AFM image of FL-MoS_2 flake on a Si surface forming a heterojunction, and (b) the height profile taken along the dotted line in the AFM image.	75
4.3	(a) AFM image of FL-MoS_2 flake on a Si surface forming a heterojunction, and (b) the height profile taken along the dotted line in the AFM image.	76
4.4	I-V characteristics of the MoS_2/Si heterojunction in the dark and under light irradiation of different wavelengths for (a) p-type and (b) n-type devices.	77
4.5	The I-V characteristics of pure Si only with Al contacts in the dark and under light irradiation of different wavelengths using a linear scale for (a) p-Si (b) n-Si device.	77
4.6	I-V characteristics of pure Si only with Al contacts in the dark and under light irradiation of different wavelengths using a log scale for (a) p-Si (b) n-Si device.	77
4.7	Schematic illustration of two Auger-type relaxation processes: (a) Auger recombination (AR) and (b) inverse Auger recombination (IAR) resulted from photon absorption on MoS_2/Si heterojunction.	79
4.8	(a) Fitting of experimental data of the n-Si device to Eq. (5.3) in the dark and under light irradiation of different wavelengths. The symbols are experimental data points, and solid lines are fitted to the data according to Bardeen's model. (b) Calculated responsivity and detectivity at different wavelengths.	80
4.9	Photoswitching characteristic under different wavelength light irradiation at the bias voltage of -2 V (a) p-Si (b) n-Si device.	81
4.10	Energy band diagram of (a) MoS_2/p-Si and (b) MoS_2/n-Si at heterojunction interface under reverse bias condition under photo-excitation. The arrows indicate the transport of electrons and holes across the heterojunction.	82

5.1	(a), (b) SEM micrographs of the MoS_2 film grown on a GaN substrate indicating interconnected MoS_2 flakes.	85
5.2	(a,b) Optical images of the MoS_2 thin film grown homogenously on GaN substrate, the scratch in penal (b) was intentionally introduced to show the color contrast.	85
5.3	(a) The XRD patterns and (b) Raman spectra of the MoS_2/GaN heterojunction.	85
5.4	An x-ray photoelectron spectroscopy (XPS) analysis of (a) the Mo 3d and (b) the S 2p core regions of MoS_2.	86
5.5	(a) Schematic diagram of the MoS_2/GaN heterojunction photodetector device. (b) Current-voltage characteristics of the device without illumination.	86
5.6	(a) UV-Vis absorption spectra of MoS_2/GaN heterojunction. (b) Current-voltage characteristics of the device in dark and under various light intensities.	86
5.7	(a) The current–voltage characteristics of the MoS_2/GaN heterojunction based photodetector measured in the dark and under light irradiation with different light intensities under reverse bias condition. (b-f) Photo switching characteristic of the device under 365 nm light irradiation with different light intensities at the bias voltage of 0.5, 1, 1.5, and 2V. The light was turned on/off regularly to measure the time-dependent response of the photodetector. (d) Shows the enlarged rising and falling edges with a very fast rise (τ_{rise}) and decay (τ_{decay}) time of 5.3 and 5.6 ms, respectively.	88
5.8	(a) The photocurrent as a function of light-intensity under different voltage bias conditions. (b) Responsivity and detectivity measured at the bias of 1 V with different incident light-intensity.	89
5.9	(a) Energy band diagram of the MoS_2/GaN heterojunction under equilibrium and (b) under reverse bias condition showing the process of charge transfer at the junction between MoS_2 and GaN by light illumination. The bandgap (E_g) of MoS_2 and n-GaN is 1.3 and 3.4 eV, respectively. E_C, E_F, and E_V denote the bottom of the conduction band, Femi level, and top of valence band, respectively.	90
5.10	Barrier height as a function of light intensity at the MoS_2 and GaN interface.	91
6.1	A comparative analysis of popular gas sensors using metal oxide and 2D heterostructures.	94
6.2	Fabrication flowchart of MoS_2/GaN heterojunction gas sensor.	95
6.3	(a) Schematic diagram of the fabricated device. (b) optical micrograph of at MoS_2/GaN heterointerface, affirming the large-scale growth of MoS_2 on GaN substrate.	96
6.4	(a) 3D schematic illustration of the MoS_2/GaN heterojunction based sensor under hydrogen molecules. (b) Current-voltage characteristics of MoS_2/GaN heterojunction using log and linear scale at room temperature.	96
6.5	(a) Current-voltage characteristics of MoS_2/GaN heterojunction measured at different temperatures in (a) absence and (b) presence of hydrogen.	96
6.6	Dynamic response resistance to different concentration of hydrogen at (a) R.T., (b) 100 °C, (C) 150 °C, and (d) 180 °C. (e) Cyclic test with 0.1% hydrogen concentration at 150 °C. (f) The sensitivity of the MoS_2/GaN sensor as a function of hydrogen concentration at different values of temperature. The sensitivity curves of MoS_2- only and GaN only at an optimum temperature of 150 °C have also been added as control samples.	98
6.7	I–V characteristics measured in absence and presence of 1% hydrogen at 150 °C for (a) only MoS_2 film (b) only GaN film.	99
6.8	Dynamic response resistance upon hydrogen exposure at 150 °C for (a) only MoS_2 film (b) only GaN film.	99
6.9	Change in barrier height at MoS_2/GaN interface as a function of hydrogen concentration at different values of temperature.	100
6.10	The schematic illustration of the energy band diagram of MoS2/GaN heterostructure under reverse bias condition (a) without and (b) with hydrogen exposure. The dotted red line depicts an increase in barrier height in the presence of hydrogen. E_V, E_C, and E_F symbolize the valance band maximum, conduction band minimum, and Fermi level, respectively.	101
7.1	Three-dimensional schematic diagram of the device.	104
7.2	Current–voltage characteristics for (a) Al/p-Si/Al and Al/n-Si/Al configuration and (b) Au/MoS_2/Au configuration.	104
7.3	Current-voltage characteristics measured at different temperatures (100, 200, 300, 400 and 500 K) for (a) MoS_2/p-Si device (b) MoS_2/n-Si device. The inset shows current on a log scale.	105
7.4	FE-SEM images of MoS_2 deposited on GaN film (a) at the edges and (b,c,d) at the central zone with different magnifications.	107
7.5	(a) Schematic diagram of the fabricated device (b,c) Optical image and Raman spectra of MoS_2/GaN heterojunction (d) Tapping-mode AFM image of MoS_2 film grown on GaN substrate. Inset shows the 3D image of a grown MoS_2 film.	107

7.6 (a) Current-voltage characteristics of MoS_2/GaN heterojunction using log and linear scale at room temperature (b) Energy band diagram of MoS_2/GaN heterojunction. 108
7.7 (a) Current-voltage characteristics of MoS_2/GaN heterojunction using log scale at different temperatures. (b) The bar graph depicting the variation in barrier height and ideality factor with temperature. 108