

**Quantum Transport Properties and Scattering Mechanisms in Transition Metal
Dichalcogenides**

by

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ABSTRACT

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**Quantum Transport Properties and Scattering Mechanisms in Transition Metal
Dichalcogenides**

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Abstract here

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Table of Contents

List of Figures	v
List of Tables	vi
1 Introduction	1
1.1 Birth of Integrated Circuits	1
1.2 Graphene as a New Two-dimensional Material	1
1.2.1 Properties of Graphene	1
1.2.2 Band Structure of Graphene	2
1.3 Two-dimensional Materials: Transition Metal Dichalcogenides	3
1.3.1 Properties of Commonly Used transition metal dichalcogenides (TMDs)	3
1.3.2 Current State of TMDs	3
1.4 Hall Effect	3
1.4.1 Overview	3
1.4.2 Theoretical Background	3
2 Experimental Details	5
2.1 Substrate Preparation	5
2.1.1 Substrate Cleaning	5
2.2 Exfoliation	6
2.3 Device Synthesis	7
2.3.1 PDMS Transfer	7
2.3.2 Polycarbonate Pickup Method	8
2.4 Characterization	9
2.4.1 Optical Characterization	9
2.4.2 AFM Characterization	9
2.5 Device Fabrication	10
2.5.1 Device Design	10
2.5.2 Electron Beam Lithography	10
2.5.3 Metal Deposition	11
2.6 Electrical Measurements and Characterization	11
2.6.1 Measurement Devices	12
3 Intrinsic Channel Properties and Scattering Mechanisms	13
3.1 <i>p</i> -type WSe ₂ Semiconductor Contact Resistance	13
3.2 <i>p</i> -type WSe ₂ Hall Effect Measurements	15
3.3 <i>p</i> -type WSe ₂ Field-Effect Mobility	16
4 Future Works and Conclusion	20
4.1 Performance Limits in TMDs	20
4.2 Integer Quantum Hall Effects	20
4.3 Shubnikov-de Haas Oscillations	22

4.4 Limitations	22
References	23
Appendices	29

List of Figures

1.1	Lattice and band structure of graphene	2
1.2	Hexagonal lattice structure of TMD	3
1.3	Band structures of MoS ₂	4
2.1	Plain wafer	5
2.2	Schematic of Si/SiO ₂ substrate	5
2.3	Photolithography system	5
2.4	Alignment marks at varying magnifications	6
2.5	Exfoliation steps	7
2.6	Transfer stage setup	8
2.7	Optical microscope	9
2.8	AFM setup and AFM cantilever	10
2.9	Scanning electron microscope	11
2.10	Electron beam lithography patterns	11
2.11	Au/Ti deposited on device	12
2.12	Electrical measurement devices	12
3.1	Transmission line 0.05% Nb doped WSe ₂ channel device	14
3.2	Contact resistance of 0.05% Nb doped WSe ₂ channel device	14
3.3	Hall effect measurement diagram	16
3.4	Hall measurement data for 0.05% Nb doped WSe ₂ channel	17
3.5	WSe ₂ device with degenerately doped contacts and lightly doped channel	18
3.6	Degenerately doped contacts and lightly doped channel field-effect mobility	19
4.1	Example data of the Integer Quantum Hall Effect	21

List of Tables

1.1	Band gaps of typical TMDs and other materials	4
3.1	Summary of contact resistances for 0.05% Nb doped WSe ₂ channel	15

Chapter 1

Introduction

1.1 Birth of Integrated Circuits

The development of microelectronics revolutionized the world in the latter half of the twentieth century. The term semiconductor, in the sense it is known today, first appears in literature in 1911 [1]. Initially, work on the subject was rather pessimistic. However, in the years following Word War II breakthroughs began shed light on the possible applications and the underlying physics involved, such as the ideas of *intrinsic* and *extrinsic* semiconductors [2, 3, 4, 5].

The history of semiconductors and transistors is a well documented subject. The first transistor was constructed at Bell Labs in 1947 using polycrystalline germanium. Shortly thereafter one was developed using silicon. Throughout the following years, these devices were improved on by replacing polycrystalline with single crystals [6]. Then Jack Kilby demonstrated the first integrated circuit (IC) in 1958, for which he would win the Nobel Prize in physics [7, 8]. The scale of ICs grew rapidly in the subsequent years. Initially only a few transistors could fit on a chip (small-scale integration), in stark contrast to moder-day chips that contains billions of transistors [9, 10]. Growth continued at a rapid pace, but eventually it was realized that some limits, material and integration based, existed in silicon and other commonly used materials [11, 12]. In part, these limitations increased the interest in alternative materials. As a result widespread and renewed interest has led to a breadth information and results on a wide range of materials and their applications.

1.2 Graphene as a New Two-dimensional Material

Layered materials have existed for billions of years, and have been studied over the last few centuries [13, 14]. In recent decades the scientific study of graphite (3D) has led to new forms materials, such as carbon nanotubes (1D) and fullerenes (0D) [15, 16, 17]. However, only more recently have scientists began to understand the potential of such layered materials and their potential technological applications. After attempting unsuccessfully to synthesize few-layer graphite during the 1960s (only around 10-50 layers were able to be synthesized) a breakthrough was finally achieved [16]. This most notably began with the synthesis of monolayer graphene [18].

1.2.1 Properties of Graphene

To date, graphene's properties have been the focus of much research, both theoretical and experimental. It has been one of the primary driving forces in study of 'relativistic' condensed matter physics due to its low dimensionality and its band structure that allows electrons to mimic relativistic particles confirming

the appearance of several relativistic phenomena [19, 20, 21, 22]. In its most basic sense, graphene is composed of a single layer of carbon atoms arranged in two-dimensional honeycomb lattice (see fig. 1.1(a)) It has a Young's modulus of 100 GPa (several times more than steel) with a breaking force that is 13% of its Young's modulus [23, 24]. Its strength is due, in part, to its strong in-plane carbon (C) bonds. In addition, graphene can sustain elastic deformations of 20% due to its two-dimensional nature and it has high pliability [16]. These mechanical properties are of interest because graphene lies in the extreme ranges of many metrics considering its size and dimensionality.

Aside from its mechanical properties, graphene's transport properties were another reason why the material was so appealing. Graphene's mobility is several times that of silicon's. Experimental results have shown graphene mobility around $15,000 \text{ cm}^{-2}\text{V}^{-1}\text{s}^{-1}$ with a potential theoretical limit of $200,000 \text{ cm}^{-2}\text{V}^{-1}\text{s}^{-1}$ [25, 24]. The upper theoretical limit imposed on mobility is due to scattering, however, these high mobilities are achieved mainly because electrons in graphene act very much like photons in their mobility due to their lack of mass. This enables them to travel sub-micron distances without scattering [26]. In reality, there are other limiting factors that need to be considered such as the quality of graphene and scattering with the substrate, for example.

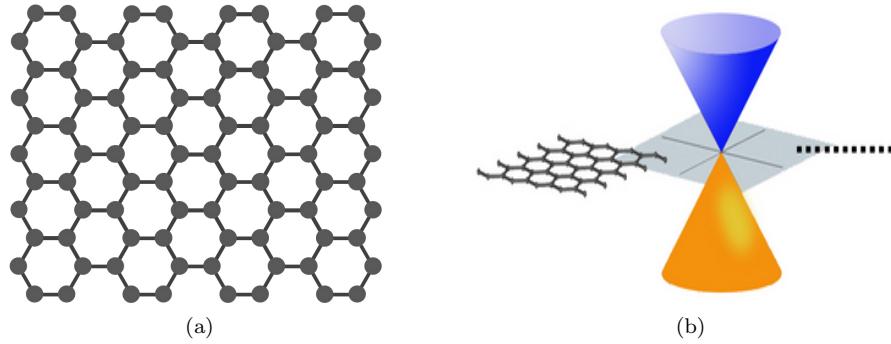


Figure 1.1: (a) Graphene: a layer of carbon atoms in a honeycomb lattice. (b) One of the most unusual features of graphene is that its conduction and valence bands meet at a point, meaning that in single-layer graphene there is no band gap (Figures obtained from [27]).

1.2.2 Band Structure of Graphene

Despite its impressive properties, the main drawback of graphene is its lack of bandgap. As this became known, the prospect of using graphene for the fabrication to ICs became unlikely. In graphene the conduction and valence bands touch at a single point as shown in fig. 1.1(b) [28]. Ultimately, the lack of a bandgap means that the current on/off ratio is low and is unappealing for logical circuit applications [29]. However, graphene exhibits some interesting properties as a result of having no bandgap, particularly as it pertains to its optical properties. The material's band structure allows for absorption of light over a large range of the electromagnetic spectrum, ranging from infrared ($< 1.65 \text{ eV}$) to ultraviolet ($> 3.2 \text{ eV}$), offering potential electronic-photonic device applications [30, 31, 32]. Since a direct use in logical circuits is not practical researchers have moved on to look for 'two-dimensional materials beyond graphene.' Several attempts at some derivatives of graphene-like materials have been studied, but for the most part they do not seem promising for use in logical circuits [33, 34]. As of late, research has been concentrated on two-dimensional materials, namely transition metal dichalcogenides, as a candidate for applications in ICs.

1.3 Two-dimensional Materials: Transition Metal Dichalcogenides

Commonly referred to two-dimensional materials beyond graphene, TMD have garnered much interest. TMDs were studied previously, however, they have gained renewed interest due to their properties [35, 36, 37, 38]. TMDs consist of hexagonal layers of metal (M) atoms in between two layers chalcogen (X) atoms (see fig. 1.2(b)), such that the stoichiometry of the material is MX_2 [29]. The material is dependent on the type of transition metal, typically one of: molybdenum (Mo), tungsten (W), niobium (Nb), rhenium (Re), nickel (Ni), or vanadium (V), and two chalcogen atoms, typically one of: sulfur (S), selenium (Se), or tellurium (Te) [38, 39]. The most commonly studied variations of TMDs are molybdenum disulfide (MoS_2), tungsten diselenide (WSe_2), and tungsten disulfide (WS_2). These materials are commonly stacked together involving van der Waals interactions between adjacent sheets and covalent bonding within each individual sheet (see fig. 1.2(a)) [29]. TMDs exhibit a wide variety of properties, including either being a

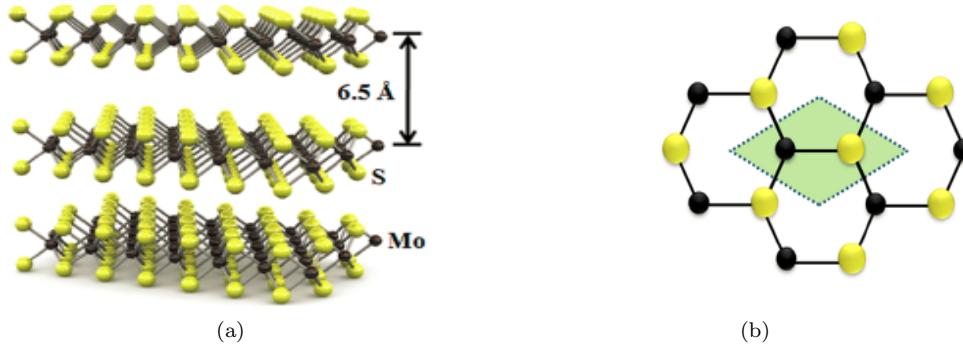


Figure 1.2: (a) The atomic structure of a layered TMD, depicting MoS_2 . Each sheet is composed of three atoms with Mo sandwiched in between two S atoms, S-Mo-S. (b) Top view of a TMD (MoS_2) lattice. (Figures obtained from [40])

metal or insulator, and displaying the topological insulator effect, superconductivity, and thermoelectricity [41, 42, 43, 44].

1.3.1 Properties of Commonly Used TMDs

As stated in sec. 1.2.2, one important property as it pertains to applications for logical circuits is the material's band structure. One of the main reasons TMDs have been so extensively studied lately is due to the fact that, unlike graphene, they do exhibit a band gap. The band gaps in some commonly used TMDs is interesting because of the transition from an indirect to a direct band gap as the layered thickness decreases. Fig. 1.3 illustrates this, for bulk and few-layer MoS_2 there is an indirect band gap while for monolayer MoS_2 there is a direct band gap. This unusual structure results in some unique optical and properties.

1.3.2 Current State of TMDs

1.4 Hall Effect

Give a brief overview of the history

1.4.1 Overview

1.4.2 Theoretical Background

Some useful references for this section [55, 56, 57, 58, 59, 60]

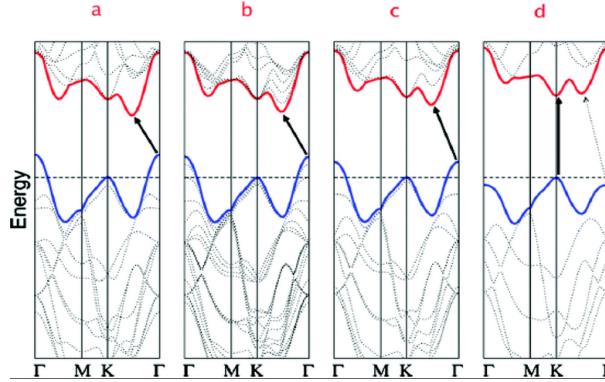


Figure 1.3: Calculated band structures of (a) bulk MoS₂, (b) four-layer MoS₂, (c) bilayer MoS₂, and (d) monolayer MoS₂. Here the solid arrows indicate the lowest energy transitions. (Taken from [45], originally appeared in [46])

2D material	theoretical E_g (eV)	experimental E_g (eV)
graphene	0	0
bilayer graphene	0	0
bulk <i>h</i> -BN	-	5.97 [47]
monolayer <i>h</i> -BN	-	6.07 [48]
few layer (2-5) <i>h</i> -BN	-	5.92 [49]
bulk MoS ₂	1.2 ^{a,b} [50, 51]	1.0-1.29 ^b [50, 51]
monolayer MoS ₂	\sim 1.90 ^{a,c} [52]	\sim 1.90 ^b [52]
bulk WS ₂	\sim 1.30 ^{a,b} [50, 53]	\sim 1.35 ^c [50, 53]
monolayer WS ₂	\sim 2.10 ^{a,c} [54]	-

^a Theoretical calculations based on first-principles calculations using density functional theory (DFT).

^b Indirect band gap semiconductor.

^c Direct band gap semiconductor.

Table 1.1: Summary of the band gaps of typical monolayer, bilayer, and bulk TMDs and *h*-BN materials. Table adapted from ref. [29].

Chapter 2

Experimental Details

2.1 Substrate Preparation

Using degenerately doped silicon dioxide (SiO_2) wafers that are 270 nm thick as pictured in fig. 2.1 and the subsequent substrate's schematic in fig. 2.2, there are several preliminary steps needed prior to device fabrication. For easy identification of locations on the substrate alignment marks are placed on the wafer using photolithography. There is a main alignment mark pictured in figs. 2.4(a) and 2.4(b) which allows for quicker identification during electron beam lithography, for example. The alignment marks are in a grid pattern with the coordinate $(0,0)$ at the center, stretching to $(\pm 6, \pm 6)$ in both the right and left directions. In each of these coordinate locations there smaller alignment marks evenly spaced within them as shown in figs. 2.4(c) and 2.4(d). Next, gold (Au) is deposited on the surface of the wafer, a process that will be explained in more detail in sec. 2.5.



Figure 2.1: Plain, polished uncut Si/SiO_2 wafer.



Figure 2.2: Schematic of Si/SiO_2 substrate.

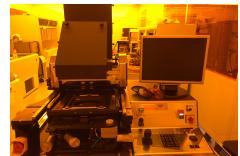


Figure 2.3: Photolithography system for creating alignment marks on substrates.

2.1.1 Substrate Cleaning

Beginning with a cut SiO_2 substrate with a deposited Au layer. To remove the Au layer, the substrate is first soaked in acetone for approximately 5-10 minutes then washed using isopropanol (IPA) and dried with nitrogen (N_2) gas. Next, the substrate is placed in acetone and sonicated for 15 minutes. Then sonicated once more but in IPA this time with a repetition of washing and drying step using IPA and N_2 as described above in between each sonication. In order to remove remaining organic matter on the surface of the substrate, the substrate is annealed under vacuum at 600°C for 10 minutes and passing forming gas for 2 of the 10 minutes. Forming gas is a mixture of H_2 and an inert gas, usually N_2 [61]. In addition to annealing the substrate for cleanliness, in certain cases when a higher degree of cleanliness is desired the substrate can be treated with oxygen plasma cleaning.

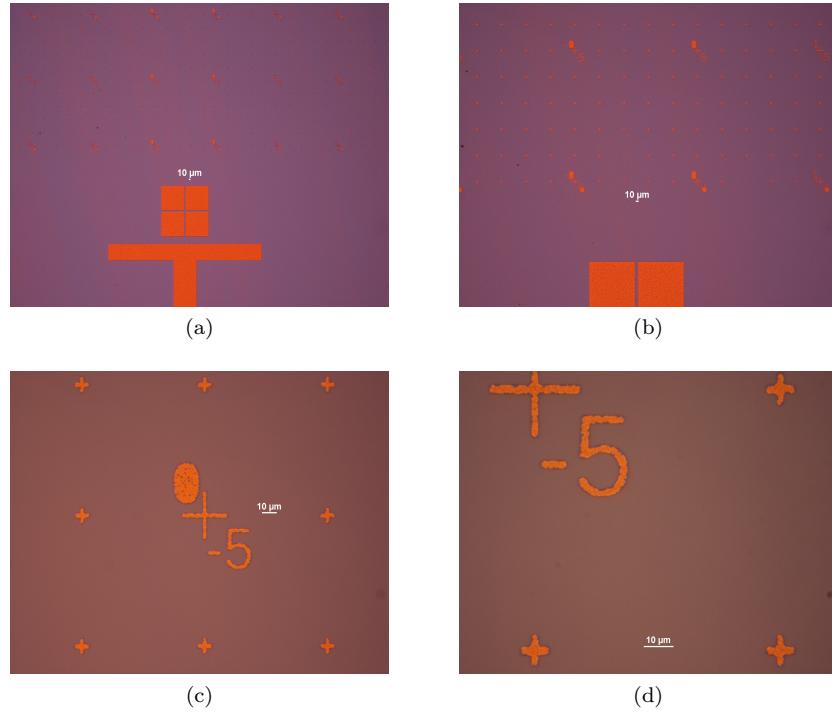


Figure 2.4: (a) Main alignment mark at 5x magnification. (b) Main alignment mark at 10x magnification. (c) Coordinate mark $(0, -5)$ at 50x magnification. (d) Coordinate mark $(0, -5)$ at 100x magnification.

2.2 Exfoliation

To synthesize samples the most common and often most effective method used is mechanical exfoliation, a technique made famous by the 2004 Novoselov et al. paper. The process involves using Scotch tape to repeatedly cleave layers of MoS₂ or some other TMD. Starting with a crystal of a particular TMD (see fig. 2.5(a)), placing it on a piece of Scotch tape (see fig. 2.5(b)). Then taking another piece of tape and pressing in on the crystal that is on the first piece of tape, being sure to press hard and firm on the crystal. The tape is then lifted up and this process is repeated until the whole piece of tape is filled with small samples of the TMD (see fig. 2.5(c)). At the end of this process it is expected that there are a wide range of mixture of sample sizes in terms of area and in term of thickness as well, where thicknesses of < 3 nm are not uncommon. To better characterize the samples the optical microscope can be used to do so.

The main challenge that exists with this method is the ability to synthesize a high yield of monolayer samples. This does not seem to be much of a challenge when it comes to graphene and some other TMDs, but with regard to MoS₂ this is not so simple. Based on recently published literature in an effort to increase the yield of monolayer MoS₂ various methods and techniques were tested and modified accordingly [62]. In this modified method an additional step to cleaning the substrate is added in which it undergoes oxygen plasma cleaning for 10 minutes to ensure the cleanliness of the substrate's surface. To promote more bonding between the substrate and the samples, the substrate is first heated at 300 °C for 10 minutes without any samples on it. During this process the normal cleaving of sample on tape from crystal taking place. Once the substrate is done heating the tape containing sample is immediately placed on the substrate and pressed firmly for several minutes. Then the substrate (with the tape still on it) is placed on a glass slide (microscope slide) and is heated at around 85 °C for five minutes. Next, the substrate (with tape) is removed from heat and the tape slowly peeled back from the substrate. The result should be a much higher yield of < 3 nm samples of larger surface area, and several trilayer, bilayer, and a few

monolayer samples.

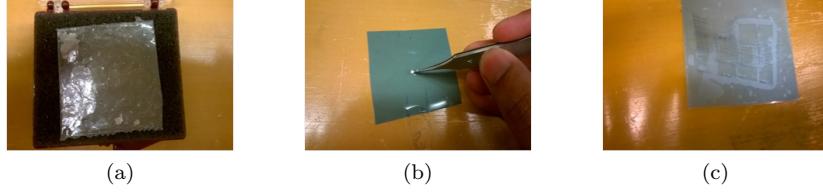


Figure 2.5: (a) Bulk MoS₂ crystal. (b) Single MoS₂ crystal on tape. (c) Tape with exfoliation MoS₂ crystals.

Most commonly SiO₂ substrates are the main items that are exfoliated onto. However, depending on the material being synthesized, this may not always be the case. In cases where samples of hBN or in the event that the thickness of the synthesized sample is not of great importance and can be tolerated up to 20 – 30 nm, polydimethylsiloxane (PDMS) is exfoliated onto instead of SiO₂ substrates. The resulting samples are of varying thickness, on average around 20 nm. Thin samples (usually trilayer and above) can be made using this method of PDMS, however, these samples tend to have small surface area and lack uniformity which poses problems as to their usability. As such, this remains an effective method for obtaining samples in which thickness is not the main concern. Once the samples have been optically characterized, the sample(s) on PDMS must be transferred to a SiO₂ substrate.

2.3 Device Synthesis

Once the sample or samples have been synthesized and characterized for their specific purpose these samples can begin to be synthesized into a device for measurement. Generally, this involves the technique of transfer. Transfer is usually done using the aforementioned PDMS or using polycarbonate (PC) known as the PC pickup method. Each method has its advantages and disadvantages depending on the type of sample, type of device, or any number of the factors.

2.3.1 PDMS Transfer

PDMS transfer is most useful for samples that were originally exfoliated onto PDMS, for example hBN. To manufacture PDMS a 10:1 ratio of silicone base and curing agent is mixed together and placed in vacuum for 30 minutes to ensure the removal of any remaining air bubbles. After this time the mixture is then spin coated on a plain SiO₂ wafer and heated at 80° C for 30 minutes then allowed to cool for 30 minutes. Once cooled, the surface of the wafer can be cut using a razor into small stamps that can be used for exfoliation and for transfer.

Once the samples that are to be transferred are on the PDMS stamp, then it is placed on a glass slide. Using the optical microscope to locate the sample on the stamp and using a razor to cut small excess pieces from the portions of the stamp where the desired sample is not located. This process is repeated until the size of the cut stamp is now reasonably small. The cut stamp is then placed at the edge of a new glass slide with sample area of the stamp as close to the edge as can be and the other side of the stamp is taped down using Scotch tape.

Next a substrate is placed and secured using glue (usually PMMA) to the stage of the transfer setup. The transfer stage setup is pictured in fig. 2.6. It consists of a microscope that has the capability of 10x or 20x magnification and a micro-manipulator. The micro-manipulator is where the glass slide with the PDMS stamp is placed. Using the manipulator the substrate on the stage is approached and the position of the stamp is checked and re-checked multiple times using the microscope to ensure correct overlap of

the desired portion of the sample(s). Upon reaching the desired position, the glass slide is lowered but this time there should be a contrast seen which is the overlapping of the glass slide and the substrate. Once the contrast has enveloped the entire sample that was to be transferred then the manipulator can be used to lift up the glass slide. Once the transfer is complete then the substrate should be annealed at 250°C for 30 minutes in order to remove any residue or organic matter that may have remained during the transfer process.

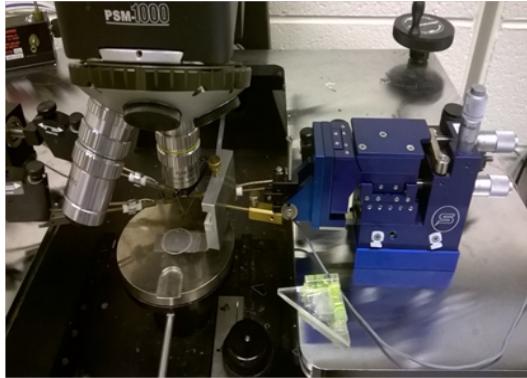


Figure 2.6: Transfer stage setup

2.3.2 Polycarbonate Pickup Method

The PC pickup method is used for samples that have been exfoliated onto a SiO₂ substrate. Generally these are thinner samples with larger surface area that are not as easily obtained by using the PDMS exfoliation method as described in sec. 2.2. To manufacture the PC, 3.0 g of chloroform and 0.18 g of polycarbonate resin are put on a plate shaker for about 60 minutes or until the polycarbonate resin have dissolved into the solution.

Next, the substrate that has the sample that is going to be transferred is taped using double-sided tape to a glass slide facing up. Then using a syringe the PC solution is placed in the substrate and evenly spread across it. Being sure to locate the area(s) on the substrate where the sample(s) are located, small pre-cut pieces of PDMS are placed over top of these areas. An outline of the PDMS stamps is cut using a razor and any excess PDMS is carefully torn away. Once only the PDMS strips are remaining on the substrate deionized water (DI) is put under the strips in order to create a hydrophobic surface and to ensure that the strip and PC that is trapped underneath it come off the substrate with relative ease. Each strip is placed on its own glass slide and is gently blown with N₂ gas to remove any excess DI from the surface.

Moving to the transfer stage setup and following the steps described in sec. 2.3.1 with regard to using the transfer stage setup. The only difference at this point to using this method as opposed to PDMS transfer is in the final step of the transfer. Instead of only lowering until the contrast change is shown between the region that is desired to be transferred, with PC transfer the entire PC must be lowered down. This is because since the PC will be heated before being lifted up. Lowering all the way ensures that all the PC will be melted. Once lowered all the way, the heating device, which is connected to the stage, should be turned up to 130°C. Once this temperature is reached, it should be maintained for approximately two minutes to fully melt the PC film. After heating the substrate and lifting up using micro-manipulator the substrate is placed in chloroform and covered for 30-60 minutes. The purpose of this is to remove any residue left over by the PC film or any other items that may have been introduced at any point in the transfer process. In practice the chloroform soaking generally needs to be repeated

several times over a few hours in order to ensure the least amount of remaining residue possible. To confirm the reduction of residue and also characterize the transferred samples an AFM is used.

2.4 Characterization

There are many ways used in modern academia and industry to characterize samples and devices. Some of these methods include scanning tunneling microscopy (STM), transmission electron microscopy (TEM), atomic force microscopy (AFM), and magnetic force microscopy (MFM) [57]. The primary characterization techniques used in this project are AFM and optical characterization.

2.4.1 Optical Characterization

The majority of the optical characterization is carried out using the optical microscope as shown in figs. 2.7(a) and 2.7(b). The microscope can magnify 5x, 10x, 20x, 50x, and 100x, in addition, it can show dark field images.

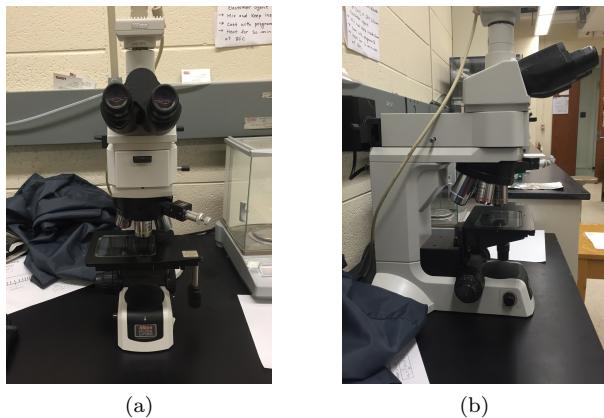


Figure 2.7: (a) Optical microscope front view. (b) Optical microscope side view.

2.4.2 AFM Characterization

In addition to the characterizing samples optically, AFM characterization is another important aspect of the device design and fabrication process. AFM characterization occurs several times throughout the process, after each transfer of a sample onto another, for example. This occurs for two reasons; to verify the thickness of the sample(s) that have been transferred, and also to verify the cleanliness of the surface of the sample (to ensure that any residue has been removed, especially during the course of PC transfer). Additionally, once the electrodes of the device have been fabricated a final AFM characterization is needed to determine the width of the device's channel which is needed to calculate various important electrical properties.

Fig. 2.8(a) shows a front view of the AFM used to characterize. For these purposes, the AFM is operating in “tapping” mode which is less invasive than “contact” mode [57]. In basic terms, an AFM works by measuring the force between the tip of a cantilever (see fig. 2.8(b)) and the sample being imaged.

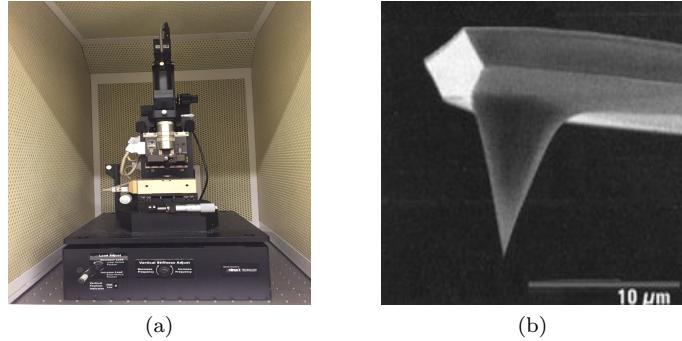


Figure 2.8: (a) Front view of AFM setup. (b) AFM cantilever tip [63].

2.5 Device Fabrication

In order to perform electrical measurements of devices one must fabricate devices. The devices are fabricated according to a specific process, though it is worth noting that under certain circumstances some steps of the process may be omitted or altered, however, the main idea remains the same regardless of the device type being fabricated. Once all transfer steps and samples have been placed in their correct locations the fabrication process begins. This process has three main steps: device design (sec. 2.5.1), electron beam lithography (EBL) (sec. 2.5.2), and metal deposition (sec. 2.5.3).

2.5.1 Device Design

The specifics of device electrode design depends largely on the type of measurement that is desired. For example, fig. 2.10(b) demonstrates a Hall bar device pattern. Despite the varying possibilities of patterns, the method for which the designs are generated remains largely the same. In order to generate these patterns a design software, Nanometer Pattern Generation System (NPGS), is used to draw the electrodes which in turn communicates the designed pattern to the SEM [64].

In preparation for EBL the substrate is spin-coated with two layers of polymethyl methacrylate (PMMA). The first layer of PMMA is 495-A4 is added then the substrate is baked on a hot plate for 5 minutes at 180° C. The first layer is followed by another layer of PMMA, 950-A2 (same polymer of different molecular weight). Again, the substrate is spin-coated with this PMMA layer and baked at the same temperature for 5 minutes.

2.5.2 Electron Beam Lithography

To generate the patterns described in sec. 2.5.1 a SEM is used to complete EBL. Fig. 2.9 shows the control panel and electron beam writer of the SEM. The SEM is optimized by adjusting the beam current to the saturation point. Using the alignment marks mentioned in sec. 2.1 the electron beam is aligned to correct position on the substrate. The NPGS system allows for adjustments in the concentration of the electron beam current at certain areas. For example, whether a line does or an area dose is necessary. In general, these values are adjusted as needed depending on the device design but are usually around $300 \mu\text{C cm}^{-2}$ for area doses and $15 \mu\text{C cm}^{-1}$ for line doses. Ultimately, the EBL process creates three separate patterns that are joined together. The first pattern is written at 1000x magnification on the SEM (see fig. 2.10(b)). The second and third patterns (written at 300x and 100x, respectively, see fig. 2.10(a)) are connected to the inner 1000x pattern and they connect this inner pattern to the “electrode pads” where the electrodes will eventually be connected for device measurement. After completing the device pattern, the substrate is developed in a solution of methyl isobutyl ketone (MIBK) and methyl ethyl ketone (MEK) for 70 seconds.

The MIBK is the main developer while the MEK acts to enhance the developing process, both fig. 2.10(a) and fig. 2.10(b) have undergone this development process.



Figure 2.9: Control panel and electron beam writer of scanning electron microscope.

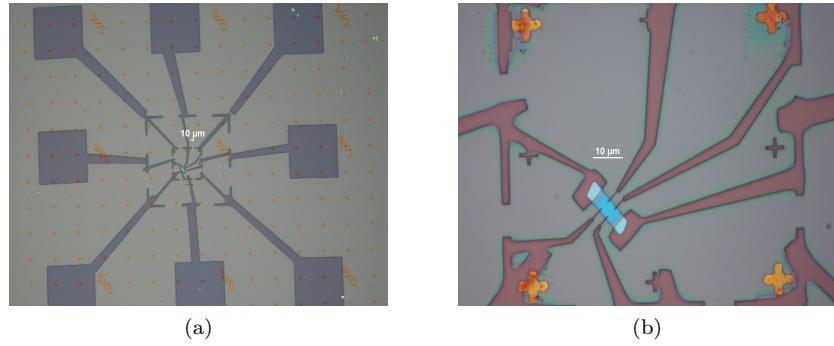


Figure 2.10: (a) Developed pattern at 10x magnification, (b) developed pattern at 100x magnification. Both developed using MIBK and MEK.

2.5.3 Metal Deposition

In order to make proper electrical contacts metal must be deposited on the pattern made by EBL. This process of metal deposition is done using a Bell Jar deposition (BJD) system which evaporates 10 nm of titanium (Ti) followed by 40 nm of Au on the substrate's surface at ultrahigh vacuum ($\sim 10^{-7}$ torr) at a rate of 1 \AA s^{-1} . After the metal deposition process is complete, a process called "lift-off" is performed. In this process the substrate is placed in acetone for approximately 5-10 minutes, until the Au has "lifted-off." Once this process is complete all that remains on the substrate is Au where the EBL was performed. Figs. 2.11(a) and 2.11(b) illustrates a substrate that has undergone the lift-off process, all that remains is the portion of the pattern that is necessary to perform electrical measurements on the device.

2.6 Electrical Measurements and Characterization

Upon completion of the fabrication process and physical characterization processes, the remaining process is electrical characterization. Many of the options available in electrical characterization are put to use, at least in some capacity regardless of the device. Then, depending on the device's quality, further measurements can be made in which a more comprehensive electrical data profile can be obtained.

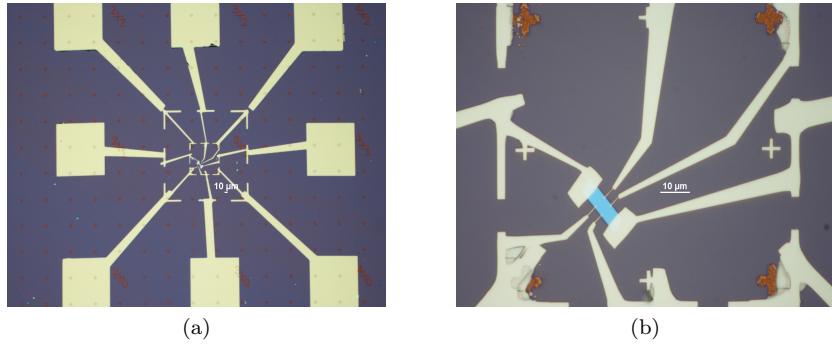


Figure 2.11: (a) Au/Ti deposited on developed sample after electron beam lithography at 10x. (b) Au/Ti deposited on developed sample after electron beam lithography at 100x.

2.6.1 Measurement Devices

Pictured in fig. 2.12(a) is a measurement device that allows for automated electrical data collection through pre-made programs. The user adjusts parameters and the desired device properties to be measured. Initially, the device is usually measured in non-vacuum conditions at room temperature in order to determine the device's quality. If it is determined that the device's quality is worth continuing with low temperature measurements then the substrate is placed in the vacuum measurement chamber pictured in fig. 2.12(b). This apparatus is used in conjunction with the measurement setup in fig. 2.12(a) to continue measurements. However, in this configuration the measurements take place under ultrahigh vacuum ($\sim 10^{-6}$ to 10^{-7} torr) and temperature control via liquid nitrogen (with the ability to cool to 70 K). In addition, there is another measurement device that allows for both temperature control and the application of a magnetic field. This device, known as a physical property measurement system (PPMS), is shown in fig. 2.12(c) and is used in situations when it is necessary to apply a magnetic field such as measuring a Hall device.

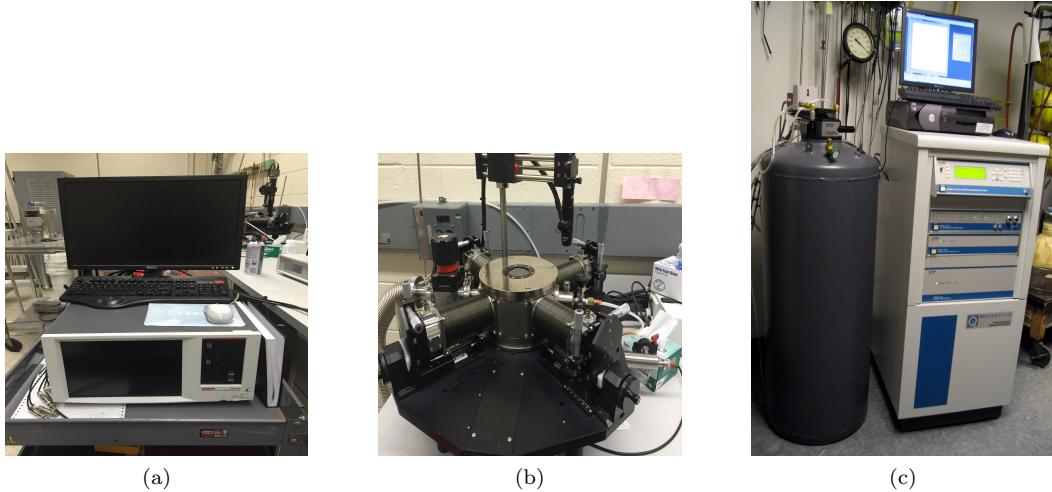


Figure 2.12: (a) Keithley semiconductor measurement system. (b) Low temperature, vacuum measurement chamber. (c) Physical property measurement system.

Chapter 3

Intrinsic Channel Properties and Scattering Mechanisms

Developing low resistance two-dimensional/two-dimensional (2D/2D) ohmic contacts opens up possibility to study the intrinsic properties of TMDs and quantum physics. In particular, quantum phenomena inherent to two-dimensional electron gass (2DEGs) and two-dimensional hole gass (2DHGs) such as the integer quantum Hall effect (IQHE) and Shubnikov-de Haas (SdH) oscillations can be explored in high mobility monolayer and few-layer TMDs [65]. In addition to quantum transport properties and quantum effects in monolayer and few-layer TMDs the study of mobility and its corresponding temperature dependence can be used to understand the multiple scattering mechanisms present [66]. These study of both electron and hole transport mechanisms is important due to the fact that high-performance *p*-type and *n*-type transistors are necessary for complimentary digital applications.

3.1 *p*-type WSe₂ Semiconductor Contact Resistance

One of the major challenges that still remains in fabricating devices to study intrinsic channel properties and scattering mechanisms is developing high quality *p*-type WSe₂ devices. This is due to the fact that the metal/WSe₂ (or MoS₂) interface is obstructed by a large Schottky barrier (SB) formed by the Fermi level pinning close to the conduction band of the WSe₂ [67, 68].

In order to fabricate high quality *p*-type WSe₂ devices, one aspect that must be addressed is how doping affects the Schottky barrier height (SBH). In particular, it is important to determine how doping can improve the 2D/2D contacts in devices. To address this issue of contact resistance several devices were fabricated and characterized in order to determine the contact resistance. Using the transmission line model (TLM) several WSe₂ devices were made with electrodes spaced at varying lengths from the source electrode. Fig. 3.1 illustrates an example of a transmission line device that was used to characterize the contact resistance. The device shown has a 0.05% Nb doped WSe₂ channel. The resistance in general is given by

$$R = \frac{\rho}{A} l, \quad (3.1)$$

where, in this case, it is assumed that the resistivity ρ and the area A are constant throughout the device [56]. The resistance R is then proportional to the length l . By determining the resistance as a function of length one can determine the contact resistance of the device. The total resistance of the device is given by

$$R = R_c + R_{ch}, \quad (3.2)$$

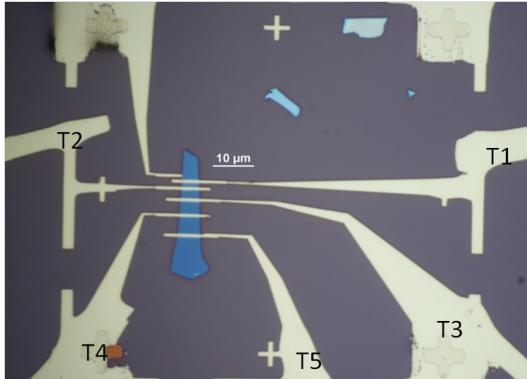


Figure 3.1: 0.05% Nb doped WSe₂ channel transmission lines with corresponding channel lengths and widths of $L_{12} = 1.04 \mu\text{m}$, $W_{12} = 4.42 \mu\text{m}$, $L_{23} = 2.04 \mu\text{m}$, $W_{23} = 4.47 \mu\text{m}$, $L_{34} = 3.09 \mu\text{m}$, $W_{34} = 4.90 \mu\text{m}$, $W_{45} = 5.15 \mu\text{m}$, and $L_{45} = 4.27 \mu\text{m}$.

where R_c and R_{ch} are the contact and channel resistances, respectively [56]. Thus by finding the resistance from the gradient of an I-V characteristic curve, one can apply the logic from eqs. 3.1 and 3.2 to find the contact resistance. Using this method the resistance as a function of length is shown in figs. 3.2(a),

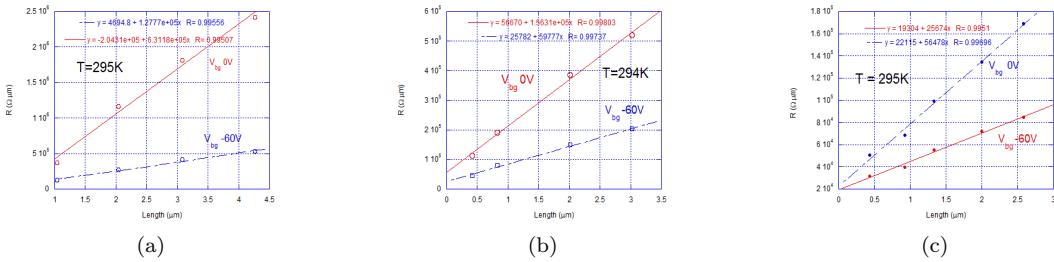


Figure 3.2: (a)-(c) show the resistance $R(\Omega \cdot \mu\text{m})$ as a function of L , where the contact resistance is found by using a linear fit function. The fits were performed at $T = 295 \text{ K}$ for V_{bg} of 0 V and -60 V . Note that (a) refers to the device shown in fig. 3.1.

3.2(b), and 3.2(c) for both $V_{bg} = 0 \text{ V}$ and $V_{bg} = -60 \text{ V}$. From these figures one can interpret the contact resistance of the device as the intercept value with the value, the resulting contact resistances are summarized in table 3.1. The results show linear behavior at room temperature for both backgate voltage of $V_{bg} = 0 \text{ V}$ and $V_{bg} = -60 \text{ V}$. The reported resistance values are larger for lower V_{bg} measurements, this can be explained by the energy mismatch of Fermi levels between the metal contacts and the lightly doped WSe₂. As the backgate voltage is increased from $V_{bg} = 0 \text{ V}$ to -60 V the SB that is present in the contacts has narrowed and as a result of the increased V_{bg} there is more transport across the barrier. The relatively small contact resistance of $2.35 \text{ k}\Omega \cdot \mu\text{m}$ indicates that low resistance contacts can be achieved at sufficiently high backgate voltage. The contact resistances are on the same order of magnitude measured by Cui *et al.* at $T = 300 \text{ K}$ and a relatively high backgate voltage ($10 \Omega \cdot \mu\text{m}$) [65].

In light of this result, the addition of degenerately doped contacts (like those fabricated in sec. 3.3) could further improve the contact resistance. By adding 0.5% Nb doped WSe₂ contacts, for example, the Fermi level energy mismatch between the metal and the degenerately doped WSe₂ contact would be decreased. Thereby lowering the SBH and coupled with sufficiently high backgate voltage, the ability to achieve

$< 2 \text{ k}\Omega \cdot \mu\text{m}$ contact resistance is plausible.

L_{\min}	L_{\max}	$R_c(\text{k}\Omega \cdot \mu\text{m})$ at $V_{bg} = 0 \text{ V}$	$R_c(\text{k}\Omega \cdot \mu\text{m})$ at $V_{bg} = -60 \text{ V}$
1.04 μm	4.27 μm	102 ^a	2.35 ^a
0.42 μm	3.02 μm	28.4 ^b	12.9 ^b
0.43 μm	2.58 μm	11.1 ^c	9.65 ^c

^a Length and resistance values from fig. 3.2(a).

^b Length and resistance values from fig. 3.2(b).

^c Length and resistance values from fig. 3.2(c).

Table 3.1: Summary of contact resistances for 0.05% Nb doped WSe₂ channel found using linear fit data from figs. 3.2(a), 3.2(b), and 3.2(c).

3.2 *p*-type WSe₂ Hall Effect Measurements

In addition to knowing how doping can be used to lower the SBH, doping also affects the intrinsic channel properties. To study how these properties are effected several measurements were taken. First, Hall bar devices were fabricated. These devices were similar to those used in sec. 3.1, the channel was doped with the same amount of Nb. The only differing aspect of these devices was the electrode design. Fig. 3.4(a) shows an example of one such Hall bar device.

The Hall effect measurement is widely used for semiconductor characterization as it gives useful electrical properties such as the resistivity, carrier density, and mobility [56]. Consider the setup shown in fig. 3.3, where the length L is taken in the x -direction, width w in the y -direction, thickness t in the z direction, and e denotes a charge carrier which can be either an electron or a hole. The current I flows in the positive x -direction and is given by

$$I = Jwt = nev_x wt, \quad (3.3)$$

where J is the current density in the x -direction, n is the charge carrier number density, and v_x is the charge carrier drift velocity in the positive x -direction. The current I is a result of the application of an electric field E along the positive x -direction. In the presence of a magnetic field B in the positive z direction the charge carriers will experience a Lorentz force that deflects them toward one side of the device. As a result there is an accumulation of charges along one side of the device which in turn creates a transverse electric field E_y [59]. The transverse electric field E_y is given by

$$E_y = v_x B, \quad (3.4)$$

where B is the magnetic field in the z direction. This accumulation of charges along one side of the device creates a potential difference that is related to the transverse electric field E_y and can easily be used to find the Hall voltage V_H by

$$V_H = - \int_0^w E_y dy = -E_y w. \quad (3.5)$$

Finally, by combining eqs. 3.3, 3.4, and 3.5 a final expression for the Hall voltage V_H is given by

$$V_H = -\frac{IB}{t} \left(\frac{1}{ne} \right). \quad (3.6)$$

From eq. 3.6 two forms of the Hall coefficient become evident,

$$R_H = \frac{1}{ne} = \frac{V_H t}{IB}. \quad (3.7)$$

Since e refers to either holes or electrons, the sign of R_H would also vary in accord with the proper carrier being described in the circumstance. Furthermore the conductivity is given by

$$\sigma = ne\mu_H, \quad (3.8)$$

where σ is the conductivity and μ_H denotes the Hall mobility. Thus, an expression for the Hall mobility can be found by combining eqs. 3.7 and 3.8 to give

$$\mu_H = |R_H|\sigma = \frac{\sigma V_H t}{IB}. \quad (3.9)$$

Following this prescribed method to determine the Hall coefficient it was used to find the Hall mobility

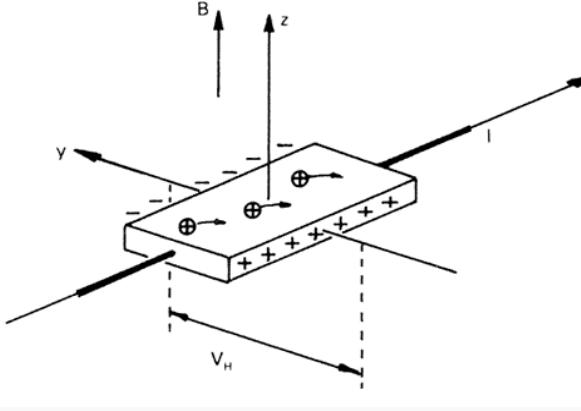


Figure 3.3: Geometry of Hall effect measurement. Current flows in the positive x -direction and magnetic field is applied in the positive z direction generating a Hall voltage [69]. Diagram originally appeared in ref. [70].

μ_H and charge carrier density for lightly doped channel WSe₂ devices.

Fig. 3.4(b) illustrates ohmic IV characteristics for the device pictured in fig. 3.4(a) for V_{bg} ranging from -70 V to 20 V at $T = 300$ K. Fig. 3.4(c) shows the conductivity σ as a function of V_{bg} for various temperatures. As the temperature increases one notices that the conductivity also increases (WHY IS THIS TRUE?). In addition to the conductivity increasing with temperature, so too, does the Hall mobility μ_H . This fact is shown in fig. 3.4(d). The Hall mobility as a function of temperature for charge carrier densities $n = -3.5 \times 10^{-7} \text{ C cm}^{-2}$ and $n = -2.5 \times 10^{-7} \text{ C cm}^{-2}$ is shown in fig. 3.4(e). Fig. 3.4(f) shows the charge carrier density n as a function of V_{bg} for several temperatures (DOES DATA MAKE SENSE?).

3.3 p -type WSe₂ Field-Effect Mobility

In an effort to improve on the approaches and results described in secs. 3.1 and 3.2 a new design of device was fabricated. These devices used a 0.01% Nb doped WSe₂ (lightly doped) channel with 0.5%Nb doped WSe₂ (degenerately doped) contacts. The device fabrication process involved transferring hBN to a Si/SiO₂ substrate then transferring the p -doped WSe₂ on top of the bottom hBN creating the channel (see fig. 3.5(a)). Once the p -doped WSe₂ is on the bottom hBN substrate then another layer of hBN is transferred to cover the channel (see fig. 3.5(b)). Next, the degenerately doped WSe₂ contacts are transferred onto the device (see fig. 3.5(c)). Finally, the electrodes are designed and the usual device fabrication steps ensue, fig. 3.5(d) shows an example of a measurement-ready device. The main quantity of interest here is the field-effect mobility as it allows for analysis of the device's quality and also allows

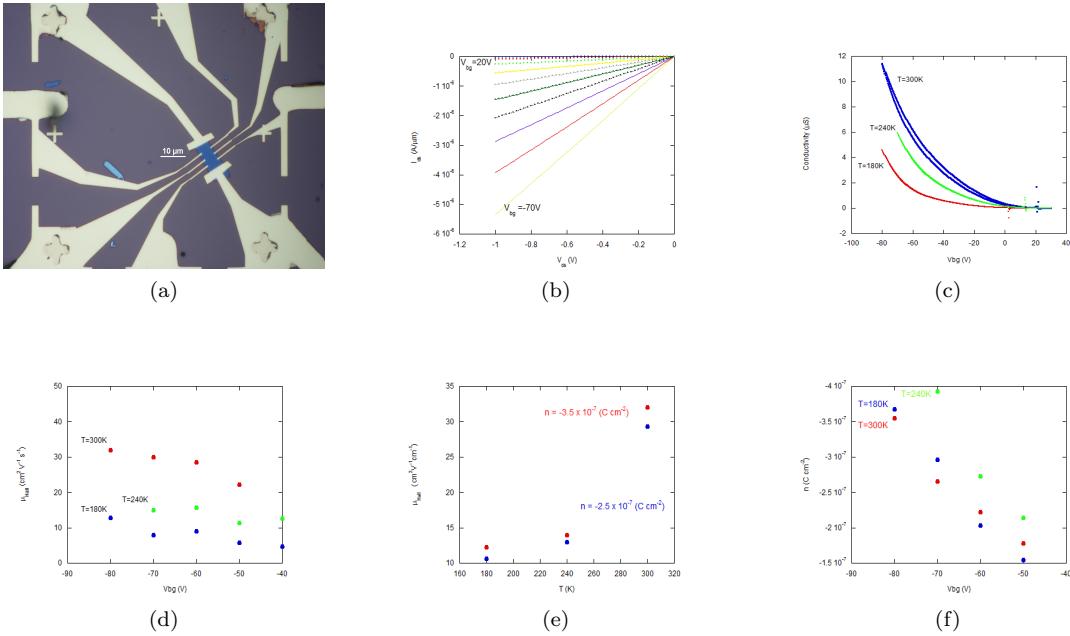


Figure 3.4: (a) Hall bar device with 0.05% Nb doped WSe₂ channel. Sample thickness is 7.74 nm with an average width $W_{avg} = 5.74 \mu\text{m}$. (b) IV characteristic curves at $T = 300K$ for V_{bg} ranging from -70 V to 20 V . (c) Conductivity as a function of V_{bg} for various temperatures. (d) Hall mobility as a function of V_{bg} for various temperatures. (e) Hall mobility as a function of temperature for different charge carrier densities. (f) Charge carrier density as a function of V_{bg} for various temperatures.

for a determination of the possible scattering mechanisms present. The two-probe field-effect mobility is given by

$$\mu_{FE} = \frac{L}{w} \frac{dI_{ds}}{dV_{bg}} \frac{1}{C} \frac{1}{V_{ds}}, \quad (3.10)$$

where L is the length of the channel, w is the width of the channel, I_{ds} is the drain current, V_{bg} is the backgate voltage, C is the capacitance, and V_{ds} is the drain voltage [71]. The capacitance is dependent largely on the substrate used, generally Si/SiO₂ and can vary from substrate to substrate as a result the field-effect mobility can vary slightly depending on the capacitance of the substrate.

Initially, the IV characteristics for the device in fig. 3.6(d) show ohmic contacts at $T = 300 \text{ K}$, however, at lower temperatures the contacts are less ohmic. Fig. 3.6(e) shows the linearity expected for various V_{bg} at $T = 300 \text{ K}$, but one can see that in fig. 3.6(f) there is a deviation in the linearity that is expected of ohmic contacts. This deviation from ohmic contacts at lower temperatures explains the behavior shown in fig. (g) where the field-effect mobility is degraded at lower temperatures ($T < 80 \text{ K}$). In attempt to improve the contacts at lower temperatures the device was annealed at 250° C for 30 minutes. The result is improved contacts at lower temperatures, fig. 3.6(i) exhibits the linearity expected of ohmic contacts for $T = 10 \text{ K}$. Annealing removed any residue or absorbant that may have degraded the contact interface. This result is supported by Britton *et al.* in which annealing improved the mobility, particularly at lower temperatures [72]. The PDMS transfer method, while relatively clean as compared to other available transfer methods, still has the potential to introduce surface residues that can affect the overall device performance, however, it has been shown that while they cannot be eliminated completely, annealing can remove them to some degree and improve device mobility. This change improved the field-effect mobility

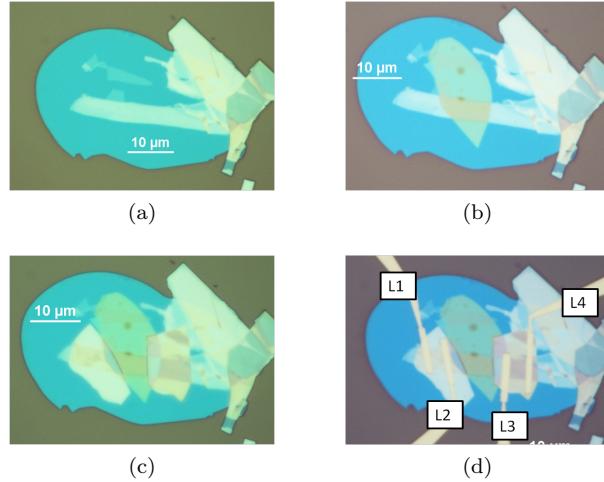


Figure 3.5: (a) 0.01% Nb doped WSe₂ transferred to hBN substrate. (b) Top hBN transferred on WSe₂ channel. (c) Degenerately doped (0.5% Nb) contacts transferred. (d) Device with 0.01% Nb doped WSe₂ channel and 0.5% Nb doped WSe₂ contacts with electrodes fabricated. Channel dimensions: $L = 9.0 \mu\text{m}$ and $W = 4.02 \mu\text{m}$ with a device thickness of 9 nm.

shown in fig. 3.6(j) where the mobility is not degraded for either V_{ds} and the overall values for mobility improved after annealing. The field-effect mobility shown here is comparable to the results found in Pradhan *et al.* where they observed field-effect mobility values as high as $350 \text{ cm}^{-2}\text{V}^{-1}\text{s}^{-1}$ at $T = 300 \text{ K}$ and a maximum field-effect mobility of $650 \text{ cm}^{-2}\text{V}^{-1}\text{s}^{-1}$ for temperature values below $T = 150 \text{ K}$ for a *p*-doped WSe₂ device on a Si/SiO₂ substrate [73]. The variation among data sets can be attributed to a number of factors; mainly doping concentration and the device fabrication method.

The field-effect mobility in figs. (g) and 3.6(j) increases with decreasing temperature which is expected behavior. The At high temperatures the mobility is largely dominated by phonon scattering which is decreased with temperature. At lower temperatures phonon scattering is decreased compared to room temperature measurements and thus long-range Coulomb scattering and short-range atomic defects play a large role [74]. Another possible scattering mechanism is interface scattering or surface roughness scattering. However, the use of a hBN substrate as opposed to a Si/SiO₂ can minimize this scattering mechanism significantly. Channel defects are another source of scattering present in this device. This is largely due to the doping of the WSe₂ channel which introduces impurities that degrades the mobility across all temperatures.

In short, the improved mobility due to the fabrication of a lightly doped channel and using degenerately doped contacts provides stepping-stone toward further refined devices. However, this technique is challenging because as doping decreases the contact resistance, it increases the channel scattering. In order to fully understand the balance and relationship between doping and the trade-off with scattering mechanisms further study of various doping strengths is needed. With such devices it will become possible to explore the intrinsic property limits of TMDs.

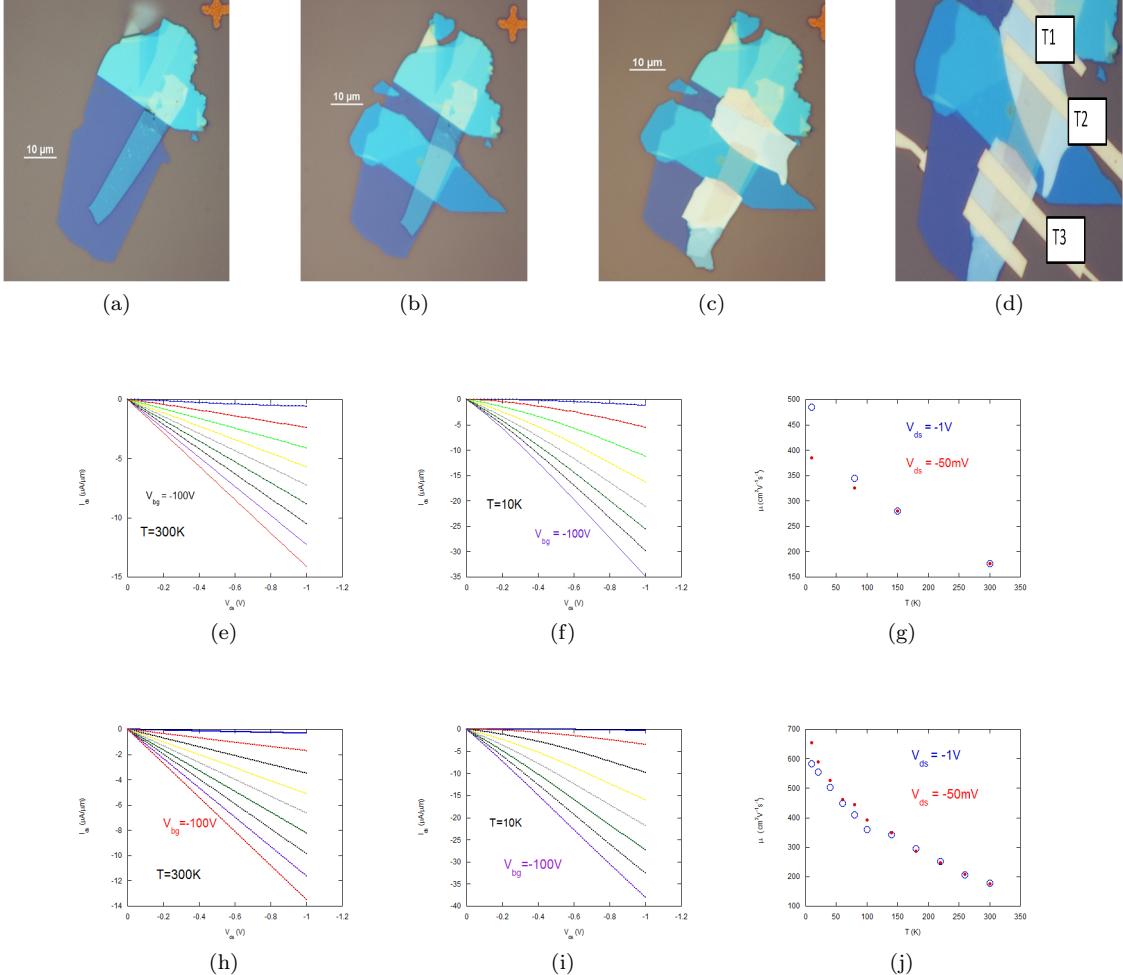


Figure 3.6: (a) 0.01% Nb doped WSe₂ transferred to hBN substrate. (b) Top hBN transferred on WSe₂ channel. (c) Degenerately doped (0.5% Nb doped WSe₂) contacts transferred. (d) Device with 0.01% Nb doped WSe₂ channel and 0.5% Nb doped WSe₂ contacts with electrodes fabricated. Channel dimensions: $L = 12.9\text{ }\mu\text{m}$ and $W = 7.5\text{ }\mu\text{m}$ with a device thickness of 5.6 nm. (e) IV characteristic curves for V_{bg} ranging from -100 V to -20 V at $T = 300\text{ K}$ before annealing the device. (f) IV characteristic curves for V_{bg} ranging from -100 V to -20 V at $T = 10\text{ K}$ before annealing the device. (g) Two-probe field-effect mobility as a function of temperature for $V_{ds} = -50\text{ mV}$ and $V_{ds} = -1\text{ V}$ before annealing the device. (h) IV characteristic curves for V_{bg} ranging from -100 V to -20 V at $T = 300\text{ K}$ after annealing the device for 30 minutes at 250° C . (i) IV characteristic curves for V_{bg} ranging from -100 V to -20 V at $T = 10\text{ K}$ after annealing the device for 30 minutes at 250° C . (j) Two-probe field-effect mobility as a function of temperature for $V_{ds} = -50\text{ mV}$ and $V_{ds} = -1\text{ V}$ after annealing the device for 30 minutes at 250° C .

Chapter 4

Future Works and Conclusion

Certain measurable quantities are valued because of the information they reveal about the geometry of the Fermi surface. Additionally, these quantities are attractive because they depend only on universal constants, experimentally controlled variables, and information about the electronic band structure which is entirely determined by the shape of the Fermi surface [58]. These measurable quantities commonly arise in the presence of a strong magnetic field and low temperature system. Determining the Fermi surface geometry provides insight into the transport and scattering properties of the material.

4.1 Performance Limits in TMDs

4.2 Integer Quantum Hall Effects

In a two-dimensional electron system (2DES) there are a number of interesting phenomena that occurs at low temperatures in the presence of strong magnetic fields. One such effect is the IQHE. The IQHE was discovered in 1980 by Klitzing *et al.* [75]. They showed that under a quantum regime of temperature and magnetic field there is a quantization of the Hall resistance, which deviates from its linearity in the magnetic field seen in the classical Hall effect, displaying plateaus at particular values of the magnetic field where the Hall resistance is given purely in terms of universal constants. In addition, the plateaus observed in the Hall resistance are accompanied by a vanishing longitudinal resistance [75, 74, 76, 77].

In order to fully mathematically describe the theory behind the IQHE one must first introduce the concept of Landau levels. Here we assume a quantum regime in which there is a low temperature and high magnetic field such that $\hbar\omega \gg k_B T$. The Hamiltonian of a particle in a uniform magnetic field is given by

$$\hat{H} = \frac{1}{2m} (\hat{p}_x + eBy/c)^2 + \frac{\hat{p}_y^2}{2m} + \frac{\hat{p}_z^2}{2m} - (\mu/s) \hat{s}_z B, \quad (4.1)$$

where \hat{p}_i is the momentum operator in the specified coordinate direction, B is the magnetic field, e is the charge of an electron, $(\mu/s) \hat{s}_z$ is the intrinsic magnetic moment operator [78]. It is worth noting that the vector potential chosen in eq. 4.1 is known as the Landau gauge, $\vec{A} = (-By, 0, 0)$, which implies the magnetic field B is directed in the positive z -direction [79, 80]. In this case the eigenfunctions of the Hamiltonian must take the form,

$$\psi(\vec{r}) = e^{(i/\hbar)(p_x x + p_z z)} \chi(y), \quad (4.2)$$

where $\chi(y)$ is defined by solutions to

$$\frac{\partial^2 \chi}{\partial y^2} + \frac{2m}{\hbar^2} \left[E + (\mu/s) \sigma B - \frac{p_z^2}{2m} - \frac{1}{2} m \left(\frac{eB}{mc} \right)^2 (y - y_0)^2 \right] \chi = 0, \quad (4.3)$$

where $y_0 = -cp_x/eB$ and $\omega = |e|B/mc$. Additionally, since the Hamiltonian does not explicitly depend on x and z this implies that both the x and z components of the generalized momentum are conserved. Eq. 4.3 is formally identical to that of the linear oscillator, thus the expression for the energy levels of a particle in a uniform magnetic field is

$$E = \left(n + \frac{1}{2}\right) \frac{|e|\hbar B}{mc} + \frac{p_z^2}{2m} - (\mu/s)\sigma B, \quad (4.4)$$

where n is any integer [78]. These quantum numbers n specify states known as Landau levels. For the case in which the motion of particles is restricted to a rectangular geometry of $L_x \times L_y$, also let $p_z = 0$ as the motion of particles is restricted in this case to only the $x - y$ plane. In this case the energy of each Landau level is given by

$$E = \left(n + \frac{1}{2}\right) \frac{|e|\hbar B}{mc} - (\mu/s)\sigma B. \quad (4.5)$$

Due to the restriction that $\hbar\omega \gg k_B T$, thermal excitations can be neglected because the interval between Landau levels is much greater than thermal excitation energy. As a result, the probability that electrons will be thermally excited to higher energy levels can be neglected. In order to fill higher energy levels the density of states must be increased. When the Landau level is fully occupied from the lowest to the i th energy level the transverse resistivity becomes

$$\rho_{xy} = \frac{h}{ie^2}, \quad (4.6)$$

where i is any integer corresponding to a specific filled Landau level, e is the charge of an electron, and h is Planck's constant [75, 76]. Eq. 4.6 shows that at critical values of the field, the Hall resistivity (or conductivity) is quantized in units of h/e^2 [57, 77]. Fig. 4.1 demonstrates an example of the quantized

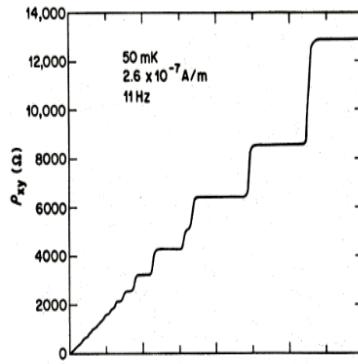


Figure 4.1: ρ_{xy} as a function of magnetic field B at low temperature ($T = 50$ mK). Figure originally appeared in ref. [81].

nature of the transverse resistivity. The distance between each plateau (step height) is given by h/e^2 divided by an integer i . These steplike increases with plateaus in the magnetic field region where the longitudinal resistivity ρ_{xx} vanished [82]. Thus, when $\rho_{xx} = 0$ then $\rho_{xy} = h/ie^2$ and is at a plateau. Note that the temperatures needed to observe the IQHE ($\lesssim 4$ K) is a likely reason why it was not discovered until 1980. It is also important to note that the value of resistivity only depends on fundamental constants of physics and can be used as a primary resistance standard known as the von Klitzing constant, $R_{K-90} = 25812.807 \Omega$ [75, 83, 84].

The measurement of the IQHE in two-dimensional materials

4.3 Shubnikov-de Haas Oscillations

There are several techniques and measurements that can determine the geometry of the Fermi surface, many of these are closely related to one another and are based on the same underlying mechanism. One such effect is the SdH effect [85]. The SdH effect is an oscillatory dependence of the resistivity on the magnetic field, there it is related to the quantum hall effect (QHE) [86]. This effect is produced by the oscillation of the density of states at the Fermi level which is caused by the quantization of electron energy levels in the presence of a magnetic field (see sec. 4.2, Landau levels) [87, 88]. (ADD MORE HERE)

SdH oscillations can provide a wealth of information about the effective 2DEG (2DHG). For example, one can extract the carrier lifetimes, effective cyclotron mass, information about the Fermi surface [89]. In order to observe the QHE and therefore observe SdH oscillations, high device mobility is required [90]. Recently the Cui *et al.* observed the first SdH oscillations in MoS₂ and Li *et al.* observed this same effect in black phosphorus [65, 89]. Cui *et al.* were able to determine geometry of the Fermi surface from the oscillation frequency and prove the two-dimensional nature of their system. Furthermore, from the analysis of the oscillation amplitude the cyclotron effective masses of both electrons and holes could be determined. From this method the carrier lifetime could also be determined, they reported lifetimes of $\tau = 0.11$ ps and $\tau = 0.12$ ps for holes and electrons, respectively [89, 91]. Cui *et al.* was able to provide insight about the quantum lifetimes and the underlying interactions between short-range and long-range scattering limits [65]. The results provide promise that with the implementations of improvements in contact resistance reduction and further mobility enhancement that the world of TMDs is closer to the routine study of novel quantum physics in two-dimensional systems.

4.4 Limitations

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Appendices

2DEG two-dimensional electron gas

2DES two-dimensional electron system

2DHG two-dimensional hole gas

AFM atomic force microscopy

Au gold

BJD Bell Jar deposition

DI deionized water

DFT density functional theory

EBL electron beam lithography

IC integrated circuit

IPA isopropanol

IQHE integer quantum Hall effect

MEK methyl ethyl ketone

MFM magnetic force microscopy

MIBK methyl isobutyl ketone

Mo molybdenum

MoS₂ molybdenum disulfide

N₂ nitrogen

Nb niobium

Ni nickel

NPGS Nanometer Pattern Generation System

PC polycarbonate

PDMS polydimethylsiloxane

PMMA polymethyl methacrylate

PPMS physical property measurement system

QHE quantum hall effect

Re rhenium

S sulfur

SB Schottky barrier

SBH Schottky barrier height

Se selenium

SEM scanning electron microscope

SdH Shubnikov-de Haas

SiO₂ silicon dioxide

STM scanning tunneling microscopy

Te tellurium

TEM transmission electron microscopy

TLM transmission line model

Ti titanium

TMD transition metal dichalcogenides

V vanadium

W tungsten

WS₂ tungsten disulfide

WSe₂ tungsten diselenide