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UNIVERSITY OF CALIFORNIA
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Optoelectronics at the Interface: Probing the Origins of Photocurrent in Complex
Heterostructure Materials

A Dissertation submitted in partial satisfaction
of the requirements for the degree of

Doctor of Philosophy

in

Physics

by

Farima Farahmand

September 2024

Dissertation Committee:

Dr. Nathaniel Gabor, Chairperson

Dr. Yantao Cui

Dr. Thomas Kolhman

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2024

The Dissertation of Farima Farahmand is approved:

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University of California, Riverside

Acknowledgments

Being in graduate school marked a challenging yet profoundly rewarding journey, characterized by substantial personal and intellectual growth, meaningful connections, and abundant learning opportunities. This experience is made significant by the invaluable support and guidance from key individuals who played a pivotal role in its realization. I wish to extend my sincere gratitude to those who have played pivotal roles in shaping these formative years.

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To mom, dad, Niloofar, and Radin

ABSTRACT OF THE DISSERTATION

Optoelectronics at the Interface: Probing the Origins of Photocurrent in Complex
Heterostructure Materials

by

Farima Farahmand

Doctor of Philosophy, Graduate Program in Physics
University of California, Riverside, September 2024
Dr. Nathaniel Gabor, Chairperson

In this dissertation, I discuss two comprehensive investigations that aim to advance our understanding of photocurrent generation in complex heterostructure material systems. I first introduce our novel microscopy method to map photocurrent streamlines in ultrathin bilayer magnetic devices made of platinum on yttrium iron garnet (YIG). Utilizing scanning photovoltage microscopy with a rotating magnetic field, we achieve the first-ever direct imaging of photocurrent flow patterns. These streamlines exhibit complex behaviors such as contortion, compression, and expansion, depending on the device geometry providing unprecedented insights into the charge flow in optoelectronic systems. Second, I describe measurements of photocurrent generation at the interface of two semimetals: WTe_2 (a Weyl semimetal) and graphene (a Dirac semimetal). By examining the detailed behavior of the photocurrent characteristics, we uncover how this hybrid semimetal system allows for an interlayer photovoltaic effect, reminiscent of semiconductor systems. Bridging these two investigations, we find that photocurrent generation is well described by the Shockley-Ramo

theorem for conductors, thus providing a foundational framework for future photogeneration studies in complex heterostructure optoelectronic devices.

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Chapter 1

Introduction

1.1 Motivation

Our quantum condensed matter research is concerned with the fundamental quantum processes of light-matter interaction, in quantum devices and observing them via optoelectronic techniques. These topics run in a wide field within the nanoscience area, including the complex electronic processes at the nanoscale. It attracts wide interest from various areas due to rich potential applications that can make a revolutionary differences in future device performance. One main area of focus is the development of ultra-efficient photovoltaics such as carbon nanotubes, that hold promise for significant improvements in energy conversion efficiency. In addition, researchers have discovered an unexpected regime for hot-carrier transport in graphene and other materials, which has opened up new paths for high-speed electronic and optoelectronic devices[?].

Beyond these applications, research in quantum condensed matter has pushed investigations into new materials and heterostructures, notably transition metal dichalco-

genides (TMDs) and their interfaces that occur when combined with other low-dimensional materials such as graphene. Ultimately, this research aims to uncover new methods of manipulating their unique properties in order to build innovative computing, sensing, and communications technologies. Quantum condensed matter research is currently at the cutting edge in the discovery and understanding of the new quantum phenomena that will revolutionize future technology.[1, 2, 3, 4, 5, 6, 7]

1.2 Ultrathin materials and their heterostructures

Two-dimensional (2D) materials such as graphene and Transition-Metal Dichalcogenides (TMDs) are two prominent classes of 2D materials that have garnered significant attention due to their exceptional properties and potential applications.[7, 8, 9]

Graphene, a single atomic layer of carbon atoms in a hexagonal lattice, has kept nanomaterials in the limelight since its isolation in 2004. Due to the very high electron mobility, mechanical strength, and thermal conductivity of this new material, it holds potential for applications ranging from transistors to sensors and even transparent conductive electrodes. One of the remarkable properties of graphene is its ability to support high carrier velocities because of its linear band structure near the Dirac points, making possible new quantum phenomena and prospects for ultrafast electronic devices.[8, 10, 11]

The other class of 2D materials, transition metal dichalcogenides, spans a wide range of electronic properties, from semiconducting to metallic behavior. Transition-metal Dichalcogenides (TMDs) such as MoS_2 , WS_2 , MoSe_2 , and WTe_2 , or in general MX_2 compounds, where M represents a transition metal atom from the IVB-VIB group (IVB: Ti

and Zr; V-B: Nb and Ta; VI-B: Mo and W) and X is a chalcogen (S, Se, or Te). For example TMDs such as MoS₂, WSe₂, and WTe₂ have shown immense potential for field-effect transistors, photodetectors, and valleytronic devices. The unique band structure of TMDs underpins their tunable electronic properties, which can be modulated by varying layer thickness, strain, and external electric fields. [12, 13, 14, 15, 16] The electronic structure of TMDs, influenced by d-orbitals, imparts a diverse range of electronic and optical properties. The unique band structure of TMDs underpins their tunable electronic properties, which can be modulated by varying layer thickness, strain, and external electric fields. These properties have contributed to advancements in practical devices such as photocells, photodetectors, and biosensors. [17]

Heterostructures of Pt on YIG with a GGG substrate are opening new avenues for major breakthroughs in photonic and electronic technologies. These heterostructures harness the unique properties of platinum and yttrium iron garnet.

Pt has been known to exhibit one of the strongest spin-orbit coupling effects, which is of essence in spintronics. Such properties usually endow Pt with the capability for producing large spincurrent from a normal charge current through the mechanism of the spin Hall effect. A thin film of Pt exhibited strong interaction between spincurrents and the magnetic properties of YIG due to this large value of spin-orbit coupling when deposited on a YIG substrate.

YIG is a ferrimagnetic insulator that exhibits perfect magnetic properties with ultra-low magnetic damping. These features make it a very good material for the study of spin dynamics and the development of spin-based devices. The ferromagnetic behavior of

YIG associated with the high spin-orbit coupling of Pt allows the study of the spin Hall effects and spin-orbit torques. The interaction of this pair is of particular significance in the functioning of spintronic devices where efficient manipulation of spin currents is required.

This can induce an out-of-plane temperature gradient across the Pt/YIG interface and create an in-plane, local photocurrent with a photoinduced Nernst-type effect. High spin orbit coupling in Pt combined with ferromagnetic behavior of YIG allows a host of new opportunities to engineer state-of-the-art photonic and electronic devices.

The unique photocurrent characteristics of heterojunctions composed of atomically thin materials remains a new frontier in optoelectronics. [18]

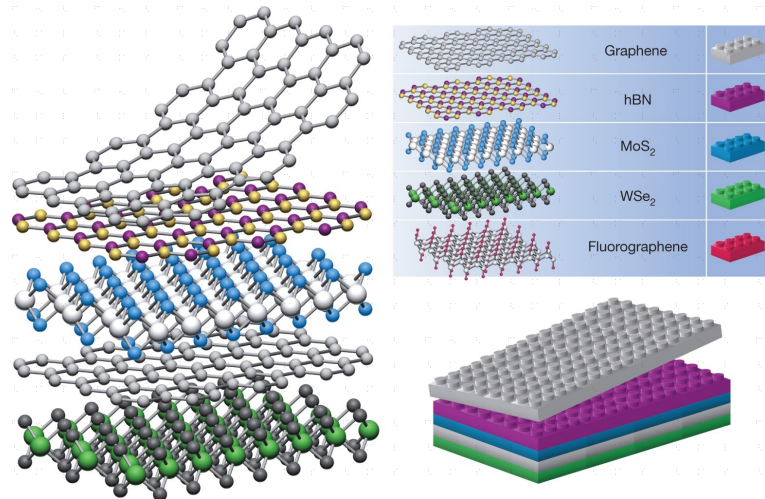


Figure 1.1: Van del Waals Materials[19]

1.3 Photoexcited Electrons in 2D Materials

The study of photocurrent in quantum materials presents a complex and intriguing challenge due to the nonlocal nature of the collection process and the intricate spatial patterns of photocurrent flow. This phenomenon is highly dependent on the configuration of current collecting contacts and the spatial nonuniformity and tensor structure of conductivity.

We employ near-infrared scanning photocurrent microscopy to investigate the dynamics of photoexcited electrons such as their emergence dynamics and directions and movement dynamics that control the electrical and optical conductivity at the interface in heterojunction.

In order to track the photoexcitation lifecycle, it is useful to consider the various stages of the photoexcitation cascade of an electronic system from initial photoexcitation to carrier relaxation and evolution back to equilibrium. There are four broad stages to the lifecycle (as illustrated in Fig. 1): (i) photoexcitation and absorption (ii) local current generation, (iii) relaxation and carrier dynamics, and (iv) propagation and collection. (i) and (ii) occur at the fastest timescales (several to hundreds of femtoseconds), whereas the timescales characterizing (iii) and (iv) often depend on material specific properties and span a wide set of timescales that range from hundreds of femtoseconds to tens of picoseconds, and, in some materials, even longer time scales. Photocurrent at each stage of this multi-step process can provide snapshots of the electronic quantum state, phase, as well as its symmetries.[20, 21]

We used photocurrent study to clarify the heterojunction band alignment in a stack of materials such as WTe_2 and graphene, which have charge compensated electron-hole bands and a Dirac point, respectively.

Our findings highlight the important role that the photo-thermoelectric effect plays in the photocurrent distribution, alongside other phenomena such as the photovoltaic effect that have been seen earlier in p-n junctions. Our measurements not only define the essential photocurrent and band alignment characteristics of this combined Dirac-Weyl system but also provide valuable insights for the design and enhancement of highly sensitive infrared sensors based on Van der Waals heterostructures.

On the other hand the photocurrent is able to exhibit a spatially nonlocal long-range nature. This feature provides "remote" sensing of the photoinduced currents that are generated far from the collection contacts, enabling in their turn innovative approaches to photocurrent diagnostics that "probe locally" and "sense remotely".

Directly visualizing photocurrent streamlines has been a challenging task, but recent advancements have led to the development of a microscopy method capable of imaging photocurrent streamlines within ultrathin heterostructure devices. This innovative approach involves the combination of scanning photovoltage microscopy with a uniform rotating magnetic field, enabling the mapping of photocurrent streamlines in various geometries, including unconventional wing-shaped electrofoil devices. The observations from this study revealed that the photocurrent streamlines in these devices exhibit behaviors such as con-tortion, compression, and expansion, reminiscent of the airflow around aerodynamic airfoils. This pioneering methodology provides a powerful tool for the visualization and characteri-

zation of charge flow in optoelectronic devices, shedding light on the intricate dynamics of photocurrent in quantum materials.[22, 23]

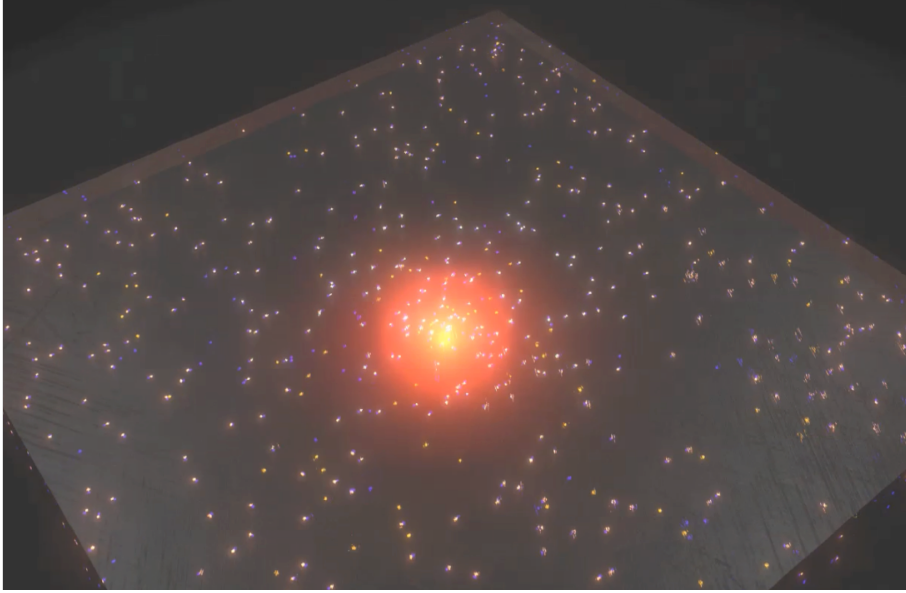


Figure 1.2: Incident of a Pulsed Laser Will Photo-excite Carriers in a Semiconducture

In the context of quantum optoelectronic devices, the visualization of photocurrent streamlines through direct imaging represents a significant challenge in comprehending the intricate behavior of these devices. In this study, we employ laser imaging in conjunction with light-sensitive devices to capture images depicting the trajectories of photocurrent within an operational device. Specifically, we introduce a novel device architecture termed an electrofoil, which facilitates the manipulation of photocurrent streamlines akin to the manipulation of airflow by wings, involving contortion, compression, and expansion. Through the utilization of imaging techniques with spatial resolution spanning over an order of magnitude, we demonstrate the efficacy of photocurrent streamline microscopy as a

robust experimental tool for the comprehensive visualization of photocurrent dynamics in quantum materials.[24]

1.4 Outline

This dissertation is structured as follows:

Chapter 1 An extensive discussion on the materials and nano-structures background that has been studied in this thesis as well as desired characteristics and properties including mechanical and electrical and optical properties .

Chapter 2 This section outlines the comprehensive fabrication process of these nanostructures, covering the entire process from inception to completion. The process transpired across different settings, including an ambient environment lab, a glove-box, and cutting-edge clean-room facilities.

Chapter 3 In this chapter, the measurement and characterization of these structures are thoroughly discussed. Various techniques were employed, including Raman spectroscopy and a comprehensive optoelectronic measurement known as Multiple Parameter Dynamic Photoresponse Microscopy (MPDPM) in the QMO laboratory.

Chapter 4 The initial and primary project in this dissertation, focusing on the dynamic behavior of laser-excited electrons and holes at the Dirac-Weyl interface composed of graphene and WTe₂, has been documented.

Chapter 5 This Chapter represents the collaborative project on Photocurrent streamline microscopy in micromagnetic heterostructure devices made of thin layer Pt on GGG/YIG.

Chapter 6 represents a summary of the dissertation.

Chapter 2

Device Fabrication

2.1 Introduction and motivation:

In this chapter, we discuss the fabrication procedures employed to create nanoscale devices for studying their optoelectronic properties including the photogeneration and transport of charge carriers. Our devices are classified into two categories: (1) van der Waals heterostructure devices, involving a layered stack of materials primarily composed of monolayer graphene and tungsten ditelluride, and (2) covalent heterostructure devices patterned into Hall bars formed by layering ultrathin Pt on GGG/YIG substrates. We offer a comprehensive overview of these fabrication techniques and their subsequent analysis to gain deeper insights into the devices' characteristics.

2.2 Heterostructure devices' fabrication:

2.2.1 Introduction

For the fabrication of heterostructure devices, our approach involves several key steps[25]. First, we carefully design the device structure, followed by preparing Silicon wafers to serve as the substrate for the fabrication process. The generation of ultra-thin layers of each material is achieved through mechanical exfoliation of bulk material, where we explore and compare various exfoliation techniques.

To ensure the quality and thickness of each material layer, we subject each flake to comprehensive characterization using state-of-the-art tools. Employing a modified dry transfer technique[26], we skillfully stack the exfoliated flakes together to form a completed heterostructure. Here, we present several strategies aimed at enhancing the efficiency of the assembly process.

Moving forward, we describe the methods utilized to fabricate devices from these heterostructures, where we employ cleanroom nanofabrication techniques. We outline details of the specific steps implemented during the fabrication process, and include alternative procedures designed to optimize device performance.

2.2.2 Device design

The process of device fabrication begins with the crucial step of designing the device structure. In our specific case, we utilized DesignCAD 2000 software to meticulously design the gold contacts intended for electron beam lithography as shown in Figure 2.1.

These gold contacts play a vital role in facilitating electrical connections within the device.

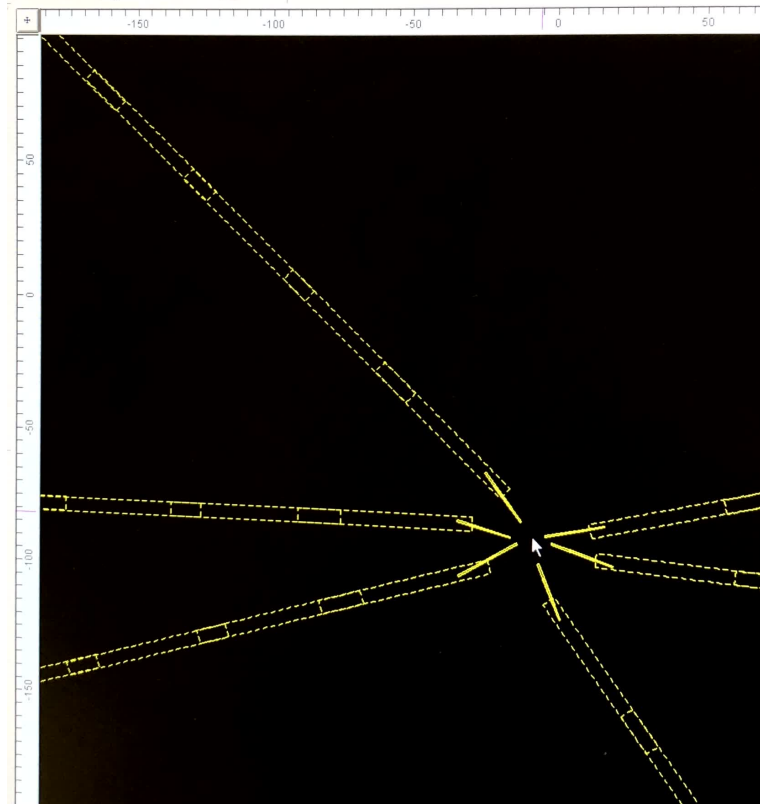


Figure 2.1: Gold Contacts CAD Design

In addition to the gold contacts, we designed the layered arrangement of materials used in our devices. The sequential layering includes from top, hexagonal Boron Nitride (hBN), followed by tungsten ditelluride (WTe_2), graphene, another layer of hBN, and finally graphite as a back-gate connection. The bottom hBN layer serves the crucial purpose of electrically isolating the bottom graphite layer from the rest of the device. The top hBN layer, on the other hand, acts as a protective element, shielding the underlying layers. These

topics will be further discussed in the section on "Building a heterostructure (Dry Transfer Technique)."

The strategic alignment of the WTe₂ and graphene layers on top of each other is critical. This arrangement ensures that each material makes individual contact with one or two of the gold contacts, thereby enabling efficient and specific electrical connections within the device. The precise alignment and layering of these materials are essential for achieving the desired functionality and performance of the fabricated device.

Accurate design and alignment ensures that the resulting heterostructure device exhibits the desired electrical properties and functionalities, making it a key determinant of success in our research and exploration of novel electronic devices.

2.2.3 Wafer preparation

The substrate serves as the fundamental building block for any heterostructure. The cleanliness and selection of the substrate material significantly influence the ultimate quality of the ensuing heterostructure. For our hBN-WTe₂-graphene-hBN heterostructures, we employ 2-inch diameter wafers with an oxide layer approximately 290nm $\pm$ 5% thick, featuring a (100) crystal orientation on the surface. These wafers play a dual role, serving both as a means to isolate individual layers of materials and as a platform to support the final heterostructures. The oxide layer's specific thickness ensures optimal optical contrast, enabling easy identification of ultra-thin to monolayer thicknesses in both graphene and WTe₂. Moreover, the (100) crystal orientation facilitates improved yield in terms of the number and size of individual flakes during the subsequent exfoliation process detailed in the following section.

To maintain the quality of our samples and facilitate straightforward identification, a cleaning process is undertaken. This step is crucial for ensuring the integrity and reliability of the resulting heterostructures. Our choice of Si/SiO₂ substrates, complemented by the precise oxide layer thickness and crystal orientation, forms the bedrock upon which we fabricate the hBN-WTe₂-graphene-hBN heterostructures. Our substrate cleaning process encompasses several steps to ensure optimal conditions for successful heterostructure fabrication. The procedure begins by carefully cutting the 2-inch substrates into smaller pieces using a diamond cutter. To eliminate any dust or particles generated during the cutting process, we utilize a controlled air flow to maintain a clean surface.

Following the cutting and cleaning of dust, the substrates undergo a thorough cleaning immersion in pure high-quality acetone for a duration of two hours. This acetone treatment effectively removes any residual contaminants and impurities from the substrate surface, enhancing its cleanliness. While the substrates are still soaked in acetone, they undergo an additional treatment involving isopropyl alcohol (IPA) and ultrasonic agitation for two minutes. The combination of IPA and ultrasound assists in dislodging and removing any remaining particles or contaminants that may have adhered to the substrate surface. The substrates are then left to soak in IPA overnight, completing the cleaning process.

When we are ready to use the cleaned substrates for the exfoliation process, we ensure they are thoroughly dried. This is achieved by employing a controlled blow-drying technique using a nitrogen gun to remove any traces of liquid from the surface. While this comprehensive cleaning process typically yields sufficiently clean substrates for exfoliation of most materials, certain materials, such as tungsten ditelluride (WTe₂), demand

additional preparation steps to ensure successful exfoliation. WTe_2 , for instance, requires enhanced stiction to the substrate surface, which can be achieved using Oxidation Plasma Etching. This additional step modifies the substrate surface, creating favorable conditions for improved adhesion of WTe_2 during the subsequent exfoliation process. By carefully customizing our cleaning procedures to accommodate the specific characteristics of different materials, we ensure the production of high-quality heterostructures, enabling us to explore and harness their unique properties for advanced applications in various fields of research and technology, see Figure 2.3.

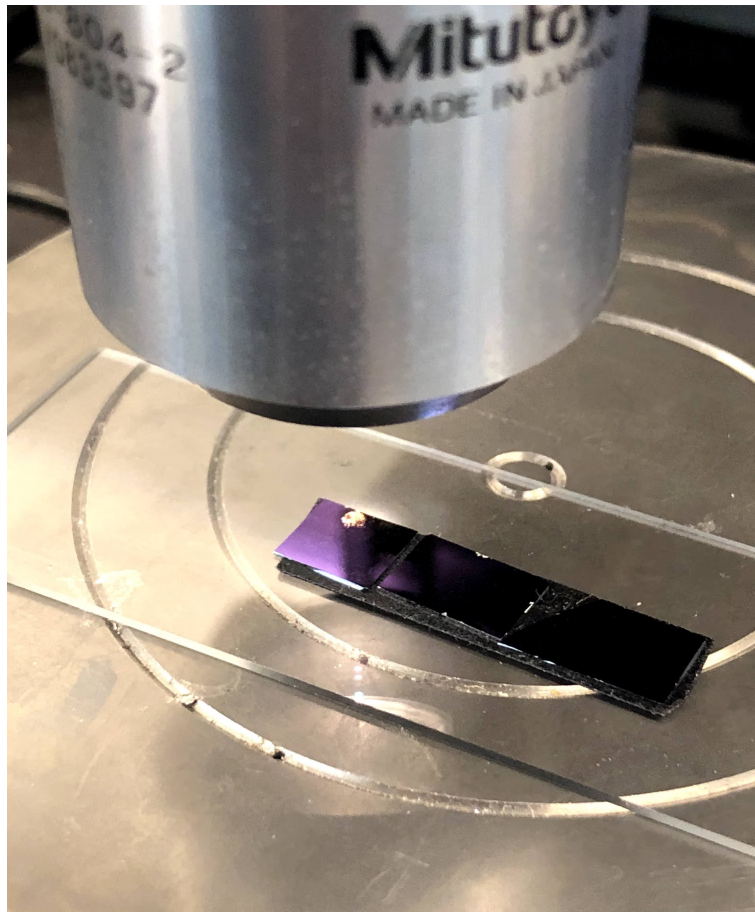


Figure 2.2: Si Substrates under Optical Inspection

2.2.4 Exfoliation and characterization of atomic layer materials

Our heterostructure devices comprise three key materials: hBN, WTe₂, and graphene. The primary technique employed for exfoliating these materials is mechanical exfoliation, which involves the following steps: dispersing the bulk material or its powder (in the case of hBN) onto a commercial scotch tape, and then repetitively pulling and peeling two sides of the tape together to achieve an even distribution of the material on the tape’s surface. The number of repetitions varies depending on the material, with graphene and hBN requiring around 10 to 15 repetitions, while WTe₂ being more fragile necessitates only 5 repetitions. Subsequently, we adhere the tape with the dispersed materials onto a meticulously prepared and clean SiO₂/Si substrate. To ensure proper bonding and flake formation, the sample is then heated to specific temperatures for each material for several minutes: 90 to 120 degrees Celsius for graphene, 60 to 80 degrees Celsius for hBN, and 110 to 130 degrees Celsius for WTe₂, with slight pressure applied using a weight between the tape and the substrate. The use of controlled pressure and temperature helps in achieving better flakes. After heating, the substrate is allowed to cool down, and the tape is peeled off at a small angle to facilitate the release of the flakes onto the substrate surface.

While the general exfoliation process is relatively straightforward, exfoliating WTe₂ demands additional care and a controlled environment to mitigate oxidation and preserve material quality effectively. WTe₂ oxidizes rapidly in ambient environments. Therefore, the entire exfoliation process is carried out in a glove box filled with argon gas as a nonreactive noble gas environment.

Next, we proceed with the material characterization process to assess the quality and desired thickness of the flakes. Employing an optical microscope, we inspect the substrate to identify suitable flakes that exhibit no cracks, possess the desired size, and exhibit a color that indicatives of their approximate thickness.

To verify material purity and thickness, Raman spectroscopy is utilized. The Raman spectra of WTe₂ from both literature and our experiment are depicted in the figure 1 showing excellent agreement between known spectral peaks and our own spectroscopic identifications. For graphene, the Raman spectrum exhibits two prominent peaks as G and 2D peaks. In monolayer graphene, the 2D peak intensity surpasses that of the G band peak, while in two or more layers, the situation is reversed as shown in Figure 2.3. [8, 27, 28]

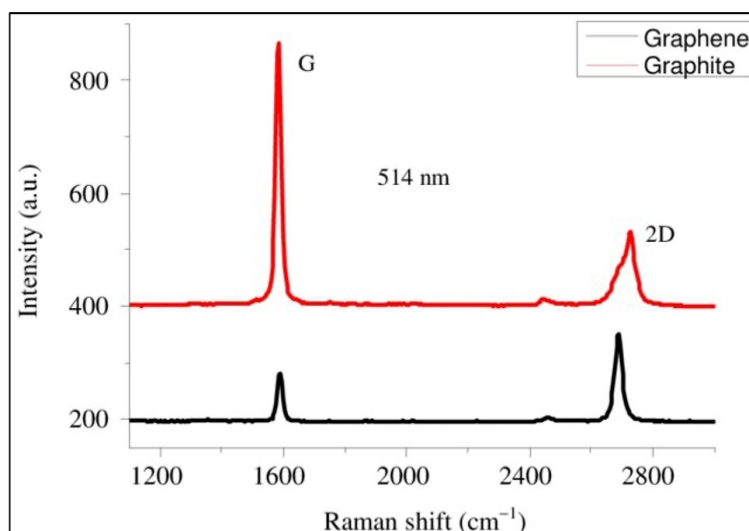


Figure 2.3: Raman Spectra of Graphene, Comparing Peaks' Intensity in Monolayer and Multilayer Graphene

2.2.5 Building a heterostructure (Dry Transfer Technique)

After successfully preparing high-quality flakes with the desired thickness for each material, the next step involves assembling them into a hBN-graphene-WTe₂-hBN-graphite heterostructure using a dry transfer technique adapted from [29]. For this purpose, we employ a custom-built transfer microscope, as illustrated in Figure. The entire transfer process is conducted within a glovebox as described previously to safeguard against any potential decay in the WTe₂ material.



Figure 2.4: Custom-built Transfer Microscope inside Glovebox

The transfer microscope consists of two key components: an upper cantilever (shown in the upper right image of Figure) and a bottom sample stage (shown in the lower right image of Figure). The bottom sample stage is composed of a machined aluminum surface to which aluminum tape and carbon tape are attached. The aluminum tape enhances the adhesion of the carbon tape to the stage while maintaining consistent heat transfer, and the carbon tape securely holds the sample in place. The cantilever, also constructed from machined aluminum, contains two slots designed to hold a stamp used for picking up

atomically thin flakes during the transfer process, see figure 2.4. The positions of the stage in the x, y, and Φ directions, as well as the temperature of the sample stage, lens, and camera, are conveniently controlled using a custom designed software from outside the glovebox, providing added convenience during the transferring process. Similarly, the positions of the cantilever in the x, y, and tilt directions are controlled through knobs located close to it, ensuring precise manipulation and alignment during the assembly process.

A small hole is drilled into the sample stage, providing access for a thermocouple wire to be positioned approximately 4 mm below the sample’s surface, visible in the Figure. This allows for accurate temperature readouts during the transfer process, facilitated by a multimeter connected to the software that reads the temperature, see figure 2.5.

This custom-built transfer microscope plays a crucial role in accurately assembling the heterostructure. By positioning and combining the flakes with precision, the microscope ensures the successful formation of high-quality hBN-graphene-WTe₂-hBN-graphite heterostructures. The controlled environment inside the glovebox safeguards the delicate WTe₂ material throughout the transfer process, ensuring the preservation of its integrity and properties.

Figure illustrates the construction of the stamp used in the dry transfer assembly of the hBN-graphene-WTe₂-hBN-graphite heterostructure. The stamp consists of a glass slide as the base, upon which a small square section of polydimethylsiloxane (PDMS) polymer, approximately 5 mm long on each side, is placed at the center. A thin layer of polypropylene carbonate (PPC) polymer is then spin-coated onto the stamp at 4000 rpm for 45 seconds with a 2000 rpm/s ramp. Subsequently, the stamp is heated at 180 for 2 minutes on a



Figure 2.5: Custom-built Transfer Microscope (Sample Stage)

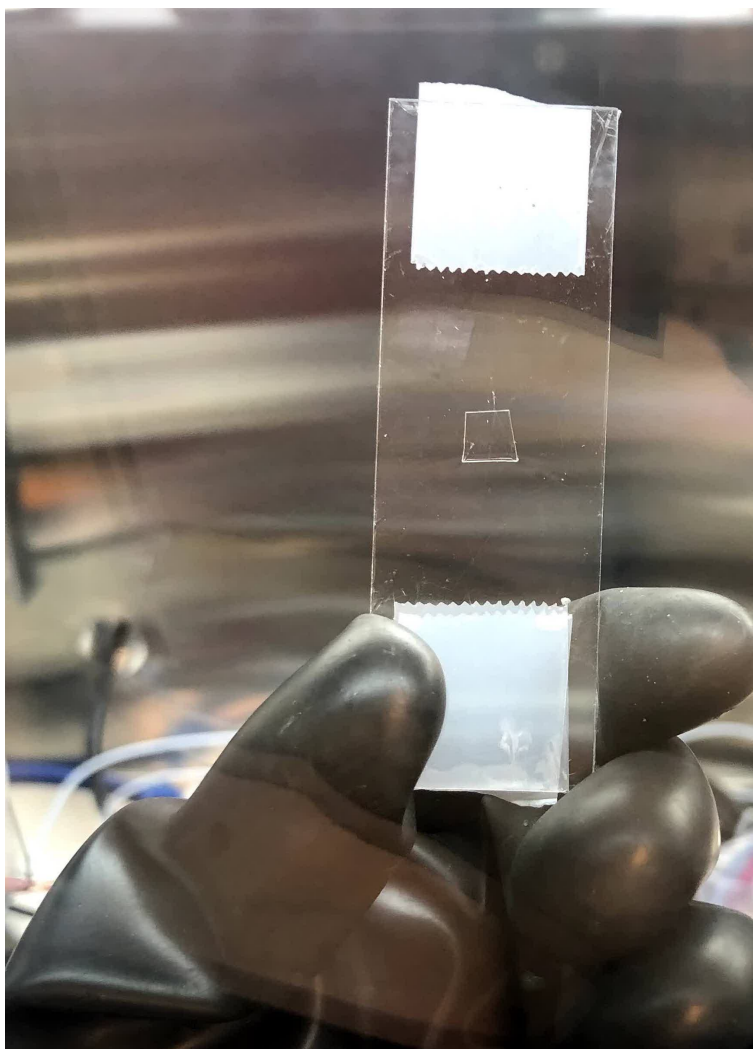


Figure 2.6: PPC-PDMS Layers on a Glass Slide

hotplate to allow the PPC to set. A glass slide with a layer, squared shape PDMS and PPC spin coated on top of it's shown in Figure 2.6.

The schematic of the standard transfer assembly method has been presented in detail in the figure. But the procedure is summarized as follows:

1. Initially, a hBN flake is placed on the sample stage, and a stamp is prepared using the glass slide, PDMS, and PPC. The hBN is picked up first because it acts as a transparent

and protective layer, especially for WTe_2 , and is crucial for successful monolayer graphene pick-up since picking up graphene with only PPC is challenging.

2. The stamp is then carefully lowered until it encounters the hBN on the sample stage.

3. After heating the stage to around 90 and allowing it to cool back to room temperature, the stamp is swiftly removed from the stage, thereby picking up the hBN flake, see Figure 2.7

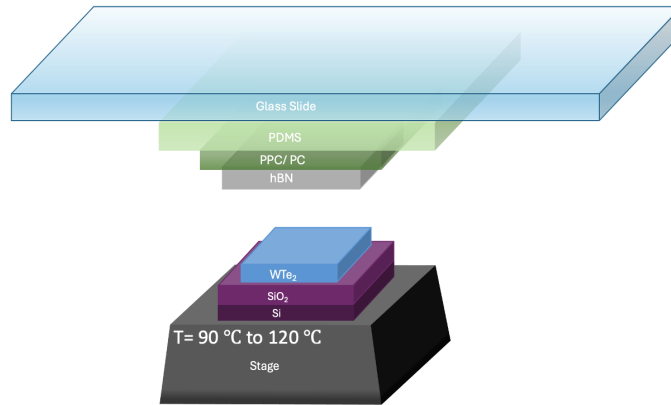


Figure 2.7: Dry Transferring Technique Steps, Picked Up hBN

4. The above steps are repeated to pick up subsequent layers, such as WTe_2 , Graphene, etc. the stage temperature need to go as high as 120 in order to pick up WTe_2 . During these steps, the flakes must be aligned at an angle to ensure no air bubbles are

trapped between the layers, and to minimize the likelihood of cracks, see Figures 2.8 and 2.9.

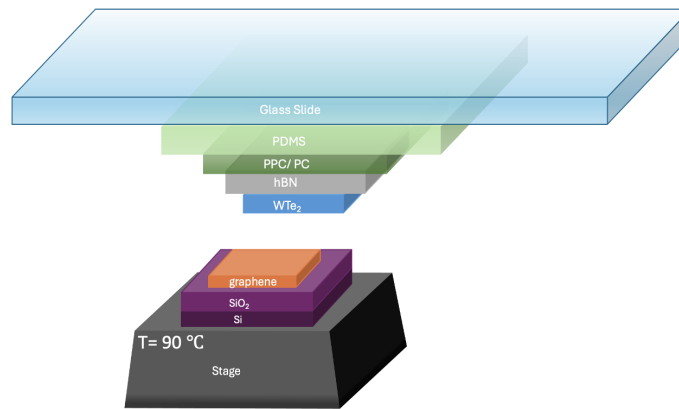


Figure 2.8: Dry Transferring Technique Steps, Picked Up hBN-WTe₂

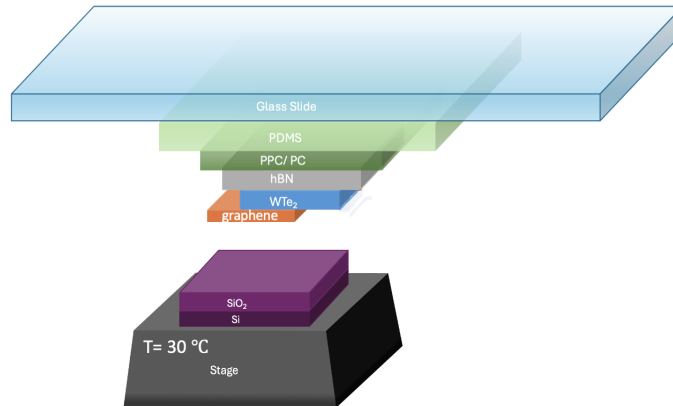


Figure 2.9: Dry Transferring Technique Steps, Picked Up hBN-WTe₂-graphene

5. Once the flakes are aligned on top of each other on the PPC, the stamp is lowered until it contacts the stage with the prepared Si substrate on it.

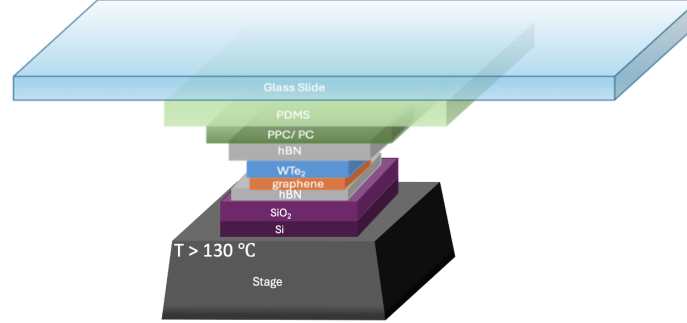


Figure 2.10: Dry Transferring Technique Steps, Dropping Stack of Materials on The Substrate

6. The stage is then heated to more than 130 passes the glass transition temperature of the PPC to melt the PPC layer, and the stamp is gradually raised to allow the PPC to separate from the PDMS, ultimately leaving the completed heterostructure on the desired substrate, see Figure 2.11. An optical image of hBN-WTe₂-graphene heterostructure shown in Figure 2.12.

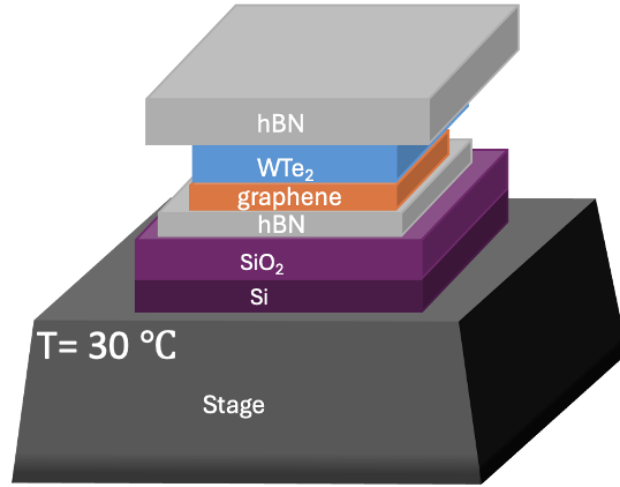


Figure 2.11: Materials Stacks

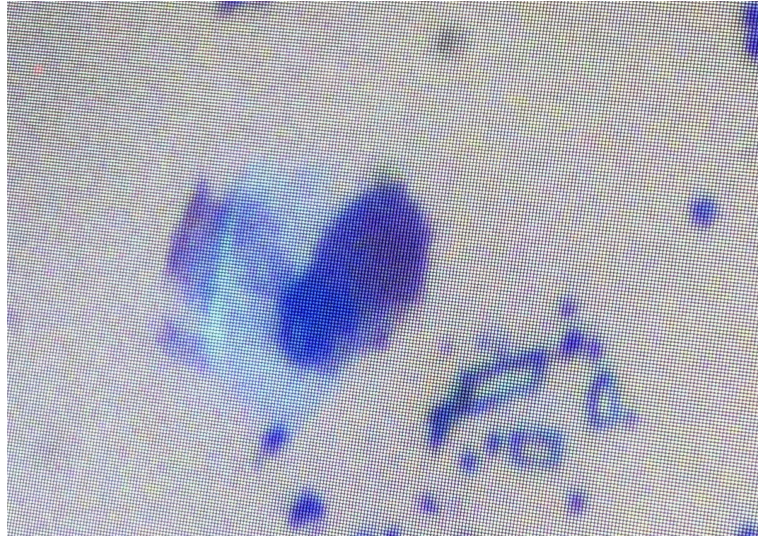


Figure 2.12: Optical Image of Materials Stacks

As mentioned before, the lattice mismatch between W and Te atoms may lead to some structural instability, and we observe that WTe_2 easily breaks during the transfer

process. For this reason, we employ PC instead of PPC, which has a higher melting point, making the transferring process more efficient, especially for WTe₂, that can be picked up more easily at higher temperatures. This variation in material choice contributes to the successful fabrication of the hBN-Graphene-WTe₂-hBN-Graphite heterostructure, tailored to the specific requirements of the devices.

It is noteworthy to highlight that incorporating two graphene layers at the end of WTe₂ and a monolayer graphene to make electrical contact with gold contacts is a more favorable choice. This approach is preferred due to the significantly lower work function difference between graphene and gold compared to that of monolayer graphene or WTe₂ with gold. This design has been used in some of our devices.

The sample is then washed in an acetone bath for 30 min to 1 hour in order to ensure complete removal of the PPC, leaving behind the completed hBN-Graphene-WTe₂-hBN-Graphite heterostructure.

2.2.6 Electrical contacts fabrication

There are two distinct approaches to fabricate electrical contacts. The first approach, known as the contact pre-write fabrication method, begins by pre-fabricating the electrical contacts. Subsequently, the materials in the heterostructure are aligned based on these pre-existing contacts to ensure reliable electrical conductivity. On the other hand, the second approach, which is the standard fabrication method, involves creating the heterostructure first and then designing and fabricating electrical contacts based on the alignment of the heterostructure.

Contact pre-write fabrication method:

The majority of devices investigated in this research were fabricated using the following method. Electrical contacts were established through electron beam lithography (EBL). In the pre-write fabrication approach for contacts, the heterostructure is initially constructed, and electrical contacts are precisely aligned with desired layers of materials to create a predetermined pattern. This process minimizes the exposure of materials to electron beams and streamlines the alignment, ensuring that contacts align precisely with different material layers.

The procedure commences with spin coating a layer of MMA polymer onto the surface at 4000 rpm for 45 seconds, with ramp-up and ramp-down speeds of 1000 rpm/s. The layer is then cured at 180°C for 10 minutes. Subsequently, a layer of PMMA EL 4 Electron Beam Resist is spin-coated and cured at 180°C for another 10 minutes. The pattern for electrical contacts is generated using the Nanometer Pattern Generation System (NPGS) and loaded onto a Leo XB1540 focused ion beam milling system, which features EBL capability, or an EBL system itself. This system exposes the substrate, causing the predetermined pattern to be written on the resist based on a pre-fed CAD file. Some of the devices were patterned using MLA 150 Advanced Maskless Aligner Lithography system which is way faster and accurate in our device precision range. Following exposure to the electron beam, the pattern is developed in a 1:3 MIBK:IPA solution, see Figure 2.13.

After development, a 4nm layer of Titanium and 25 to 75 nm of gold are deposited onto the surface using an electron beam evaporator. Titanium is deposited for stiction and device durability, while gold serves as the conductor. Alternatively, in some cases, the

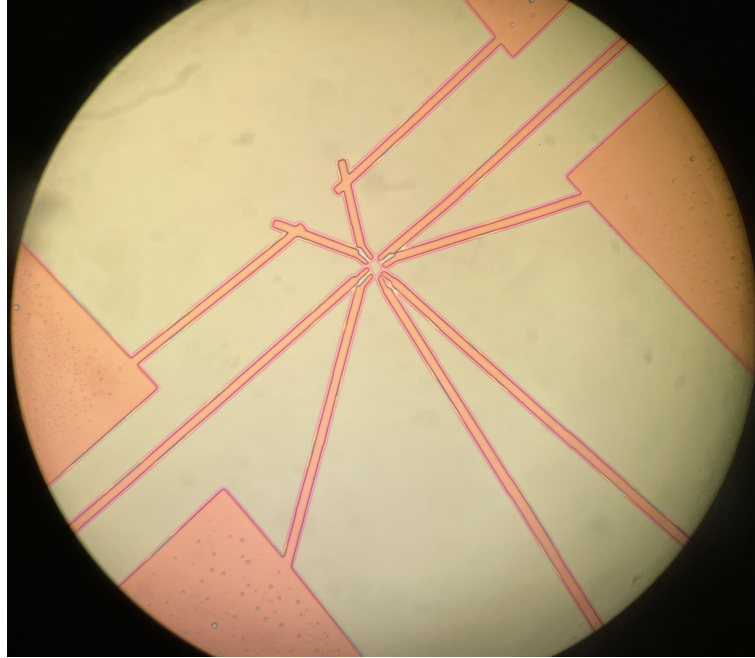


Figure 2.13: Developed Electrical Contacts

AJA Sputtering system was utilized to deposit Pt as the conductor layer, demonstrating enhanced durability and conductance. The polymer resist is subsequently removed by soaking in acetone for over 20 minutes, leaving behind the desired pattern. In certain instances, a Sonicator has been employed for thorough resist removal. The smaller ends of the contacts are arranged in a rectangle to establish contact with individual material layers, see Figure 2.14. Conversely, the larger contact rectangular pads are arranged in a half-circle, facilitating wire bonding to a larger sapphire substrate, enabling integration into the measurement system.

Post-Write fabrication method:

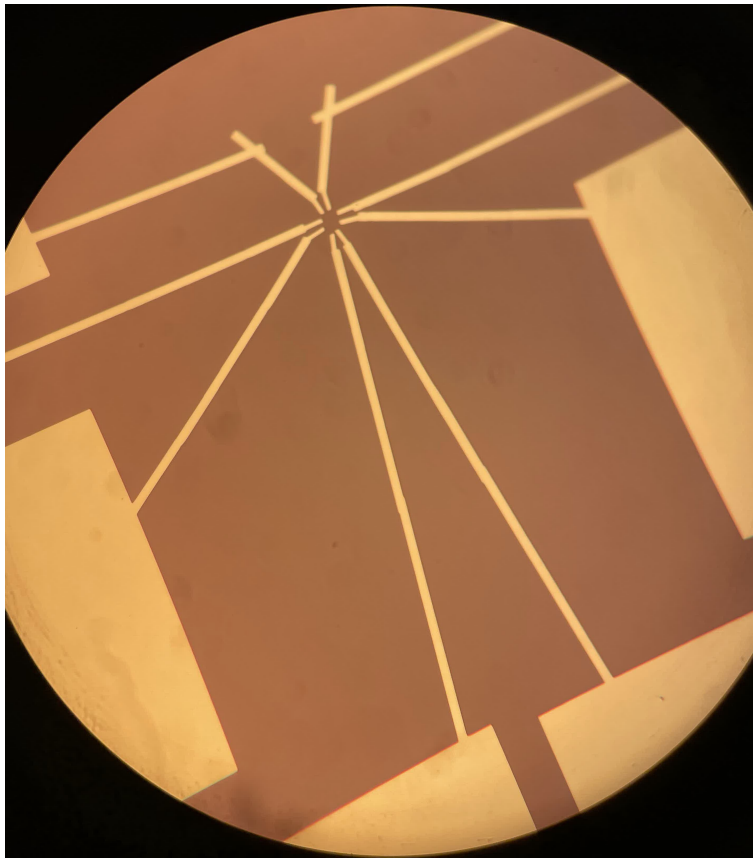


Figure 2.14: Pre-fabricated Electrical Contacts After Ti and Gold Deposition

Initially, the material's heterostructure is transferred onto the substrate, and subsequently, contacts are precisely inscribed at locations corresponding to the material's points of contact.

In this fabrication method for heterostructure devices, two key steps are involved. First, alignment markers are meticulously inscribed near the heterostructure on the sample, facilitating accurate writing of electrical contacts in the subsequent phase. Optical images of the heterostructure are captured and reconstructed to create a visual map, determining the exact distance from a chosen corner to the heterostructure. A grid of alignment markers, featuring both large and small crosses, is generated for coarse and fine alignment. This alignment pattern is overlaid onto the optical image of the heterostructure in a CAD file. Subsequently, the alignment file is loaded into the NPGS software on the FIB machine.

To inscribe the alignment markers, a suitable resist is applied to the sample in two layers through spin-coating. The FIB is then utilized to inscribe the markers by creating a small incision along one edge of the substrate. Following inscription, the sample undergoes development using a combination of solutions. The final step involves metal deposition using an e-beam evaporator, depositing 4 nm of Ti followed by 25 to 75 nm of Au sequentially. The Ti acts as an adhesion layer, and Au is chosen for its visibility under an electron beam. After the evaporation process, the sample is immersed in an acetone bath, followed by rinsing to remove excess Au, confirming the completion of alignment markers observed under an optical microscope. With the successful creation of alignment markers, the focus then shifts to fabricating the actual contacts of the device.

Similar to the alignment procedure, the process begins with capturing optical images of the

sample, including a large-area, low-resolution image at 5x magnification that shows both the heterostructure and alignment markers. Four large markers are chosen for rough alignment, captured at 50x magnification, while four small markers for fine alignment are imaged at 100x magnification, encompassing both the heterostructure and the selected small crosses. To ensure uniform orientation, images are aligned by connecting the central large cross with a small cross on its right or left before capturing them.

These optical images are then imported into another DesignCAD 2000 workspace and aligned. Designs of large and small crosses from the alignment CAD file are carefully superimposed onto the selected crosses in the optical images. The design for the actual contacts on the heterostructure is formulated, featuring four contacts on both the top and bottom layers of graphene for interlayer photocurrent measurement and redundancy. The narrow, tapered contacts closest to the heterostructure measure only 200 nm in width at their narrowest section, concluding at substantial square contact pads for wire-bonding to a chip carrier. After completing the contact design, an origin point for the contact file is designated, typically the central large cross or one of the other large crosses. The CAD file is segmented into several files for writing at different resolutions, including three files for the contact design and a distinct file for the alignment markers. Subsequently, all these files are loaded into the FIB machine.

Following the application of resist to the sample for a second time using the same procedures, contacts are inscribed using the FIB. The subsequent steps maintain consistency with the pre-write contact fabrication process as previously elucidated.

Upon deposition of the gold contacts onto the substrate, we proceed to the final stage of the dry transfer technique. This involves aligning the heterostructure, which comprises a series of stacked materials, onto the contacts. Each material in the heterostructure is electrically connected to a specific contact. The connection process is facilitated by increasing the temperature, which causes the polymer to melt and temporarily bond the heterostructure to the gold contacts. Following this, the polymer is removed using acetone, leaving only the heterostructure and the contacts intact, as illustrated in the figure 2.15. The device depicted on the right was fabricated in collaboration with Ao Shi from Prof. Lui's group at University of California, Riverside.

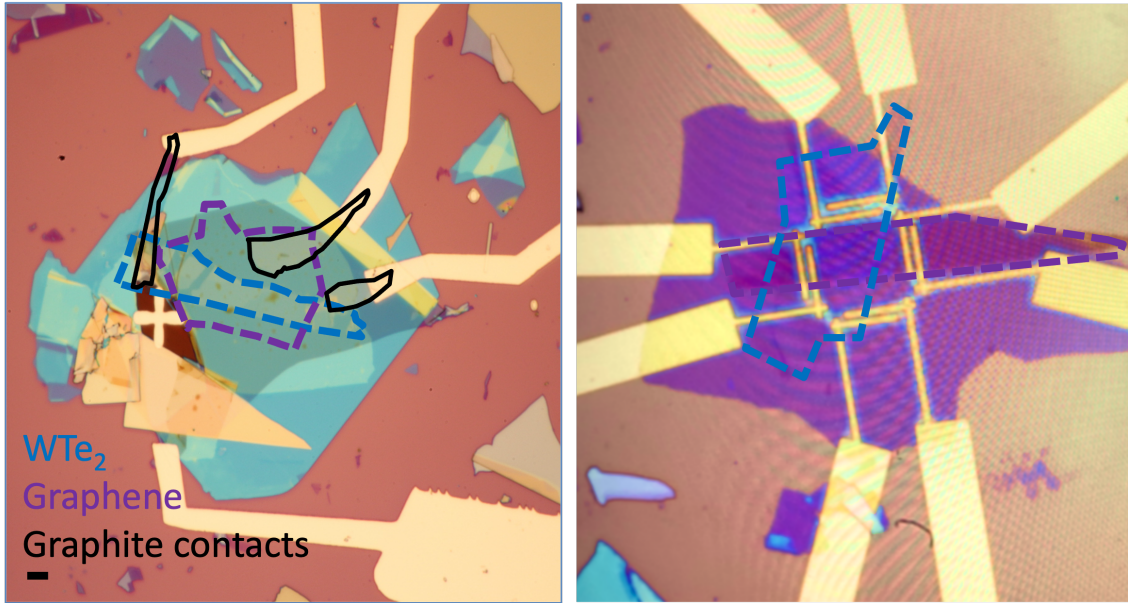


Figure 2.15: Optical Images of hBN-WTe₂-graphene-hBN Devices with Gold Contacts

2.2.7 Chip carrier fabrication

Our measurement system employs a custom-built Chip Carrier to integrate the devices, facilitating electrical connections for current measurement and voltage application. Sapphire was a deliberate choice for the chip carriers due to its advantageous thermal properties and insulating behavior. This ensures efficient thermal contact with the silicon substrate, particularly during cooling to cryogenic temperatures for low-temperature measurements. Moreover, the insulating nature of sapphire allows for the creation of separate metal patterns on top, which can be wire bonded to the small electrical contacts on the silicon substrate.

To fabricate the chip carriers' patterns, we utilize a photo-lithography procedure and deposit a 4nm/200nm layer of Ti/Au using an electron beam evaporator.

After the chip carrier is prepared, the device is mounted on top, and suitable electrical connections are established through wire bonding. For devices without Graphite as a backgate, a diamond scribe is utilized to selectively remove some SiO₂ from the bottom of the silicon substrate, revealing the conductive silicon underneath.

The device is then securely attached to the large center gold area using a small amount of silver paint, forming an electrical connection between the center gold area and the silicon substrate. This connection may function as an electrical gate for the entire device. Following this, the small electrical contacts on the device are individually wire bonded to the outer gold pads of the chip carrier, see Figure 2.16 and Figure 2.17.

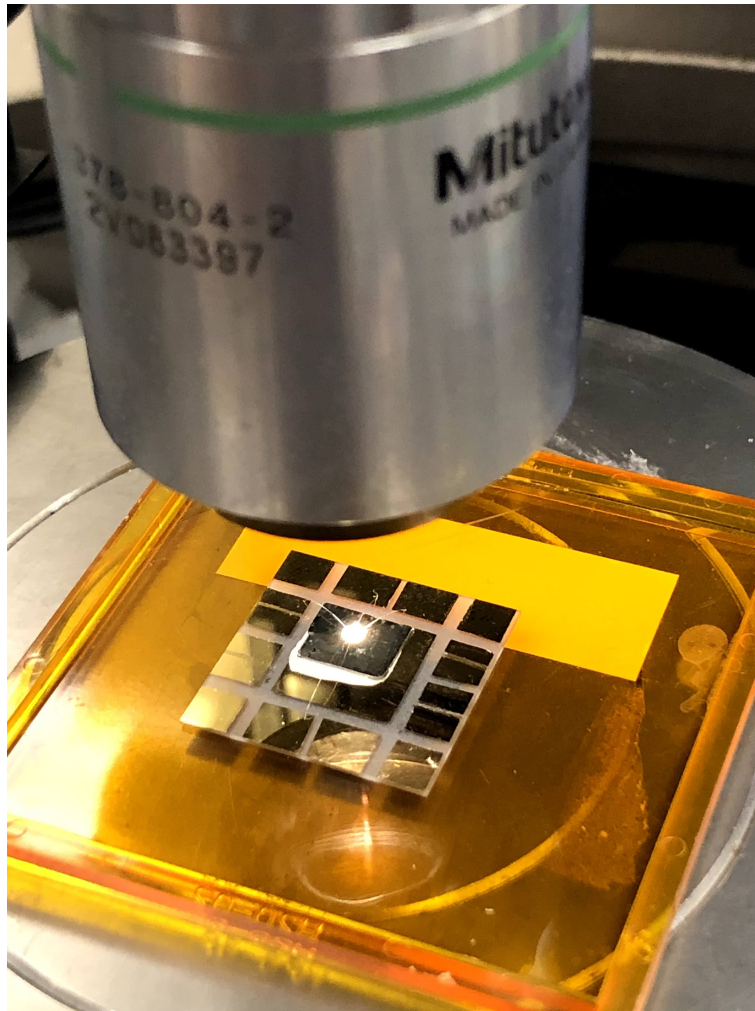


Figure 2.16: Device Wire-Bonded on a Sapphire Substrate

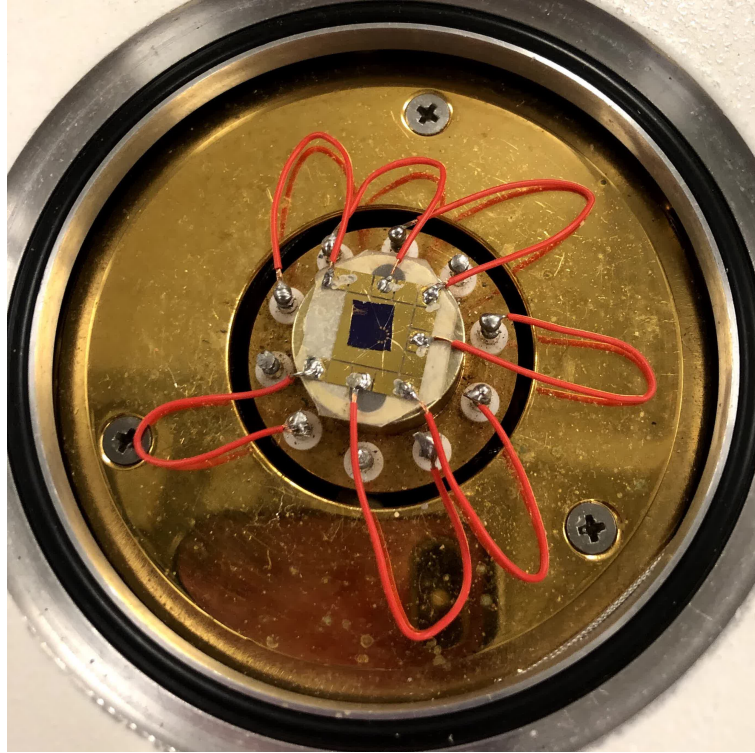


Figure 2.17: Device Wire-Bonded on a Sapphire Soldered to the Cryostat Probes

By implementing this setup, the probe needles in our measurement system gain the capability to apply voltage selectively and measure current from different contacts on the chip carrier, facilitating precise and thorough device characterization.

2.3 Hallbar Devices:

2.3.1 Device Design

The devices employed in this study were crafted utilizing meticulously characterized micromagnetic heterostructures. A comprehensive magnetic analysis of these metal/-

magnetic insulator devices has been previously documented in various publications by both our research group and others.

Our samples are composed of thin platinum films (5 nm) intricately patterned on a thin layer of yttrium iron garnet (YIG) positioned atop gallium gadolinium garnet (GGG). The 80 nm YIG films are grown through pulsed laser deposition on GGG substrates with a (110) crystal orientation, and the growth details for these YIG thin films are outlined in an earlier report. Following the confirmation of the atomic flatness and room temperature magnetic properties of the films, we utilized e-beam lithography with PMMA resist topped with Elektra 92 (SX AR-PC 5000/90.2) conductive resist to generate distinct device patterns on the surface of the thin film. The pattern employed for the electrofoil was crafted using designCAD 2000 to replicate the Clark Y airfoil design, with each pattern rotated at different angles relative to the 0 (horizontal) design.

2.3.2 Device fabrication

Hallbar and electrofoil devices were crafted employing PMMA resist covered with Elektra 92 (SX AR-PC 5000/90.2) conductive resist. This choice was made due to the insulating nature of GGG/YIG, making it undetectable for EBL. Subsequent to pattern writing, we cleaned the coating using DI water for 10 seconds, followed by resist development using IMBK for approximately 45 seconds, IPA for 45 seconds, and then water for an additional 45 seconds.

Following the development of patterns, we subjected the film to deposition in a sputtering chamber with a base pressure of 5×10^{-8} Torr, resulting in a 5nm Pt deposition. This step was succeeded by the liftoff of negative exposure features in acetone. Optical

images were captured for several of the resulting device geometries. Using this technique we were able to make devices as small as 20 microns, see Figure 2.18

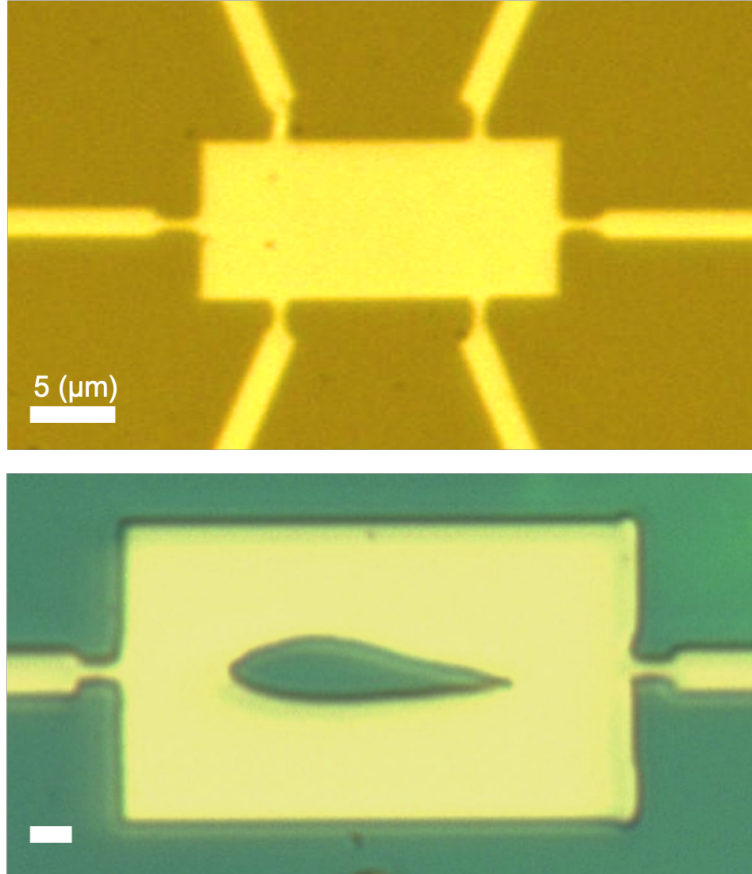


Figure 2.18: Hallbar and Electrofoil Devices

2.3.3 Chip carrier fabrication

The chip carrier designed for this measurement exhibits a unique configuration tailored to facilitate its placement between two magnets. This specific design allows for the rotational movement of the magnets at various angles around the device. The objective is to ensure the application of a uniform magnetic field from multiple orientations.

After the chip carrier is prepared, the device is mounted using carbon tape to secure it on top of the chip carrier, and suitable electrical connections are established through wire bonding, shown in Figure 2.19.

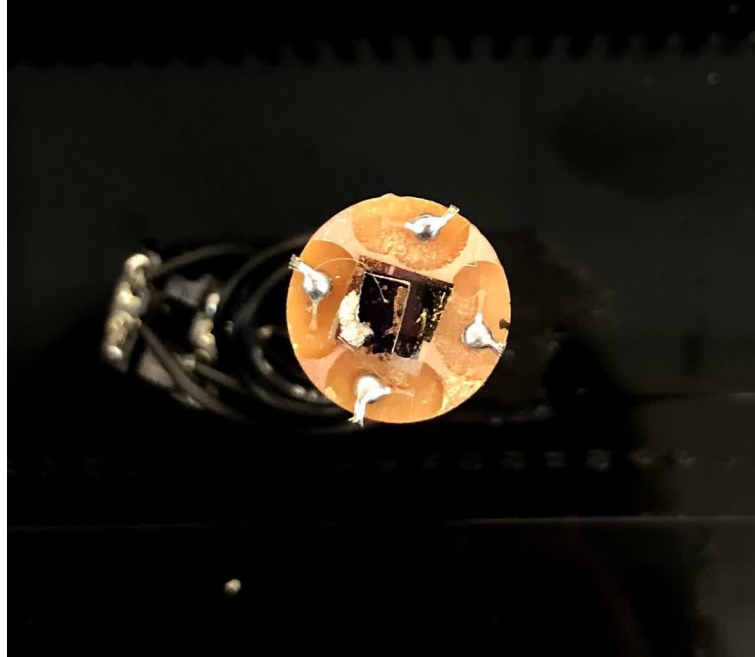


Figure 2.19: Chip Carrier

2.4 Summary:

The fabrication of nano-scale devices utilizing 2D materials presents considerable challenges, particularly when these materials exhibit air sensitivity or extreme fragility. In such instances, the precise conditions of the fabrication process become paramount. Variables such as temperature, pressure, and environmental humidity levels assume critical significance. In summary, our efforts have focused on the fabrication of devices featuring a graphene/WTe₂ interface to investigate photo-generated carriers at this heterojunction.

Additionally, we have developed Pt-based Hall bar devices to introduce a novel approach for mapping the photocurrent streamlines within these devices.

Chapter 3

Material characterization and measurement

3.1 Optical Microscopy Inspection

Optical microscopy inspection of 2D materials involves the use of light microscopy techniques to examine the surface and structural characteristics of these materials. Unlike traditional microscopy, which has limitations due to the diffraction limit of light, optical microscopy for 2D materials often employs advanced methods to enhance resolution. Techniques such as super-resolution microscopy, total internal reflection microscopy, and confocal microscopy can be utilized to achieve higher spatial resolution and provide detailed insights into the morphology, layer thickness, and defects of 2D materials.

Optical microscopy is particularly valuable for its non-invasive nature, allowing for real-time observations and the examination of large areas. Additionally, it enables the

visualization of optical properties, such as contrast variations related to different layers and electronic band structures. While not as high-resolution as electron microscopy methods, optical microscopy remains a powerful tool for preliminary assessments and dynamic observations of 2D materials.

In the course of this investigation, optical microscopy was employed to identify material flakes, representing a few layers of the material, on the substrate. The analysis aimed to assess the dimensions, surface characteristics, and overall quality of these flakes, with a particular emphasis on identifying any potential evidence of breaks or cracks.

Given that each material possesses a distinct optical index, the introduction of additional layers to the flake alters this property, resulting in changes in the coloration of the flakes. By acquiring images of the material under ideal or proposed conditions, we can effectively determine whether a particular flake is a suitable candidate for utilization in a device. This assessment is crucial for ensuring the optimal performance and reliability of the materials selected for device applications.

In the Figure 3.1 and Figure 3.2, it is evident that the identification of a single-layer graphene flake is discernible solely through color contrast, we verified this inspection using Raman spectroscopy and AFM later in the fabrication process. Additionally, in Figure (), a clear distinction is presented between the selected WTe₂ flake suitable for device applications and one that is deemed unsuitable.

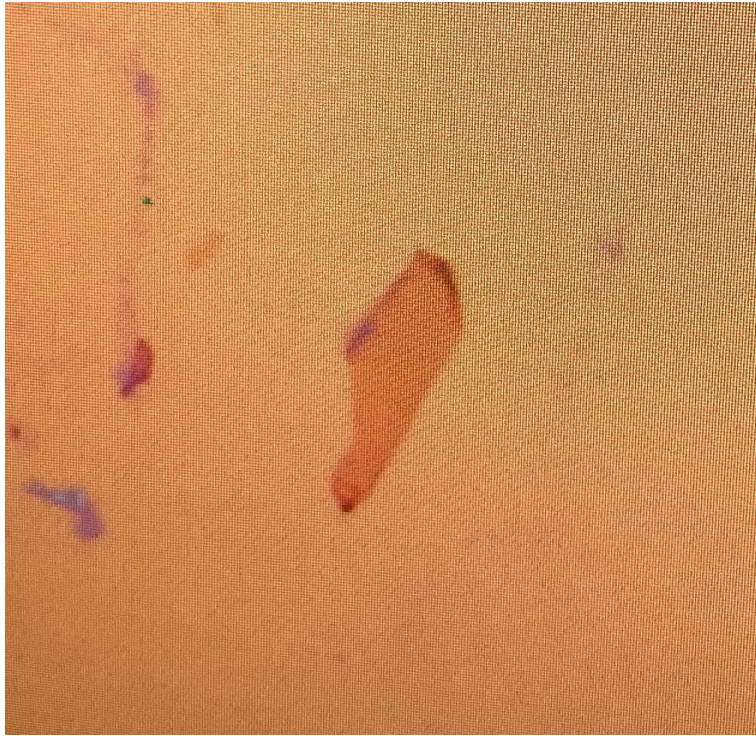


Figure 3.1: Monolayer Graphene



Figure 3.2: Multilayer Graphite

The meticulous selection of the appropriate flake for device integration holds paramount significance. This choice not only influences the quality of the device but also significantly facilitates transferring steps in the device fabrication process.

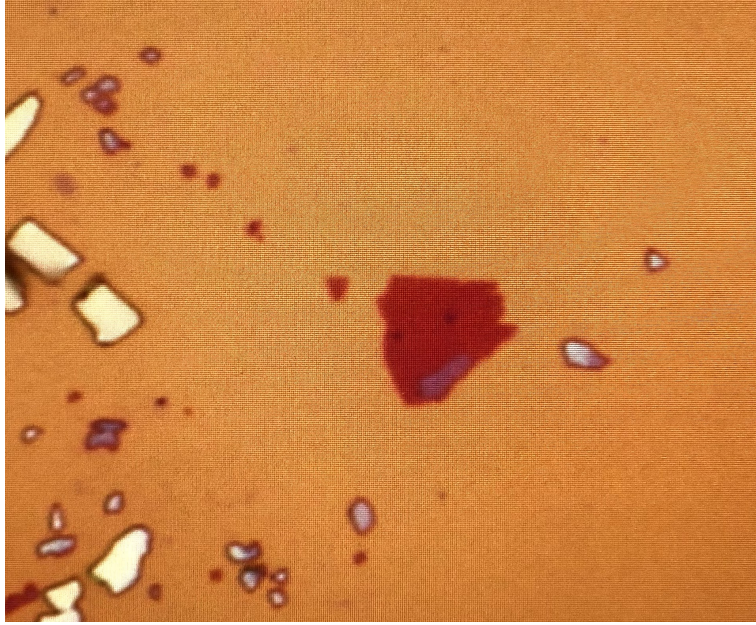


Figure 3.3: WTe₂ Flakes on SiO₂/Si Substrate

3.2 Raman Spectroscopy

Another integral characterization technique employed in the device fabrication process is Raman spectroscopy. This technique proves instrumental in distinguishing graphene flakes based on their number of layers, as illustrated by the Raman peaks depicted in Figure 3.4. Furthermore, the quality and characterization of WTe₂ flakes were thoroughly examined through Raman spectroscopy, as evidenced in Figure 3.5[30].

This comprehensive analysis aids in ensuring the structural integrity and desired properties of the materials crucial for the successful development of optoelectronic devices.

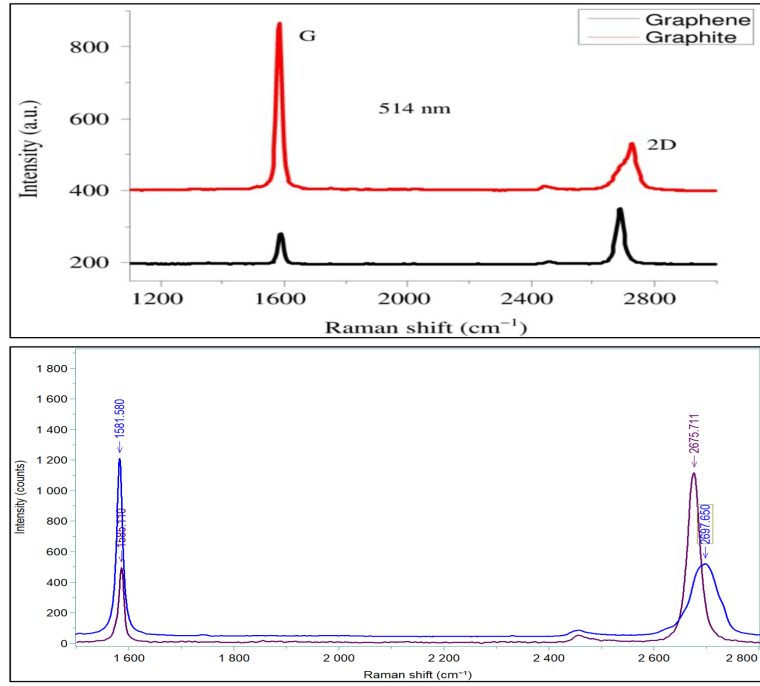


Figure 3.4: Monolayer Vs Multilayer Graphene Raman Spectra, top is borrowed from a previous study, bottom has been measured

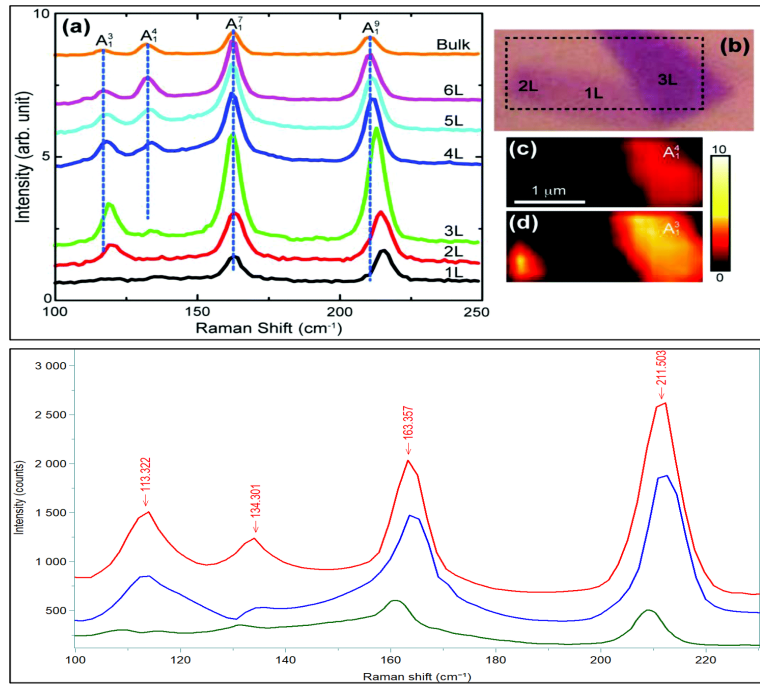


Figure 3.5: WTe₂ Raman Spectra, top is borrowed from a previous study, bottom has been measured

3.3 Atomic Force Microscopy, AFM

We used AFM to characterize the thickness of the materials' flakes. As shown in the figure the height of the substrate is () and the height of the flake is (). Based on this study () the thickness of a single layer WTe₂ is () so mean that the flakes shown is the figure contains () number of layers.[31]

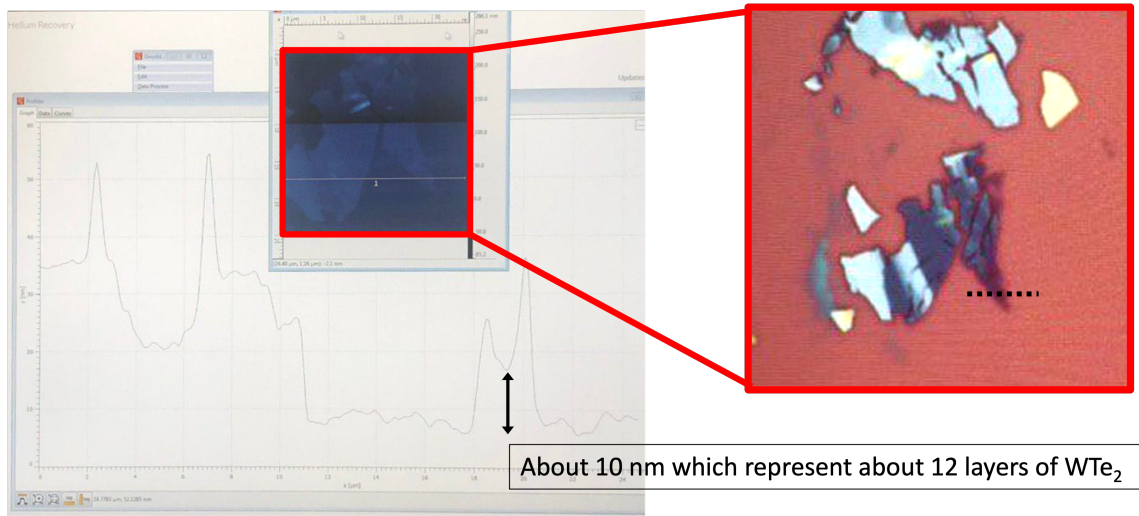


Figure 3.6: WTe₂ AFM Height Characterization

3.4 Multiple Parameter Dynamic Photoresponse Microscopy, MPDPM

We conducted a comprehensive investigation into the photoresponse characteristics and the influence of temperature gradients on the device utilizing advanced instrumentation. Specifically, we employed a scanning photocurrent microscope intricately integrated with a Multi-Parameter Dynamic Photoresponse Measurement system. This sophisticated setup allowed for meticulous mapping of photocurrent distribution across the device.[32]

Central to our methodology was the utilization of an NIR laser, seamlessly integrated with our electronic infrastructure. This integration facilitated the precise tuning of experimental parameters, including backgate voltage, source voltage, laser power, and wavelength. By systematically adjusting these parameters, we were able to adjust both the chemical potential and electric potential within the device, thereby understanding their impact on its optoelectronic properties, the setup shown in Figure 3.8, the red lines are indicating the laser path from the source to the sample.

It is important to note that each measurement required tailored adjustments to both the optical and electronic setups to effectively address the specific objectives of the investigation. Flexibility in our experimental approach was crucial, allowing us to adapt the setup according to the details of each measurement scenario.

In the pursuit of scientific thoroughness, I herein present one of the most frequently employed optoelectronic configurations utilized in our research endeavors. This illustration underscores the delicate interplay between advanced instrumentation and experimental methodology in revealing the complexities of optoelectronic phenomena within nanoscale devices.

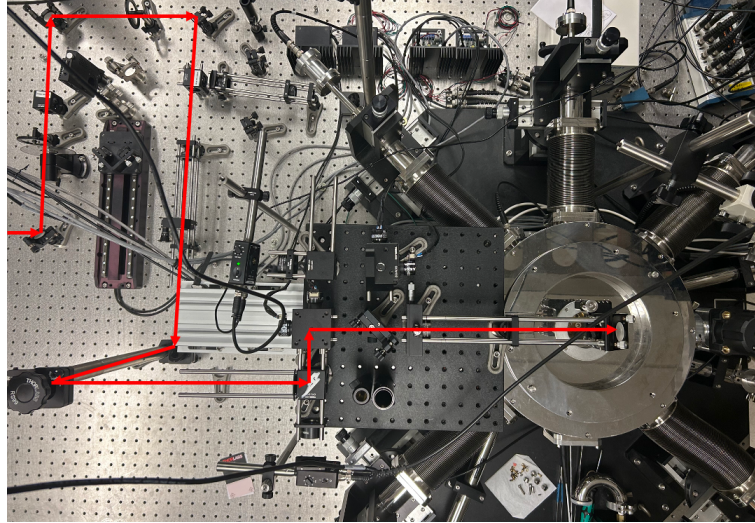


Figure 3.7: MPDPM

Figure 3.8: Optoelectronic Measurement System

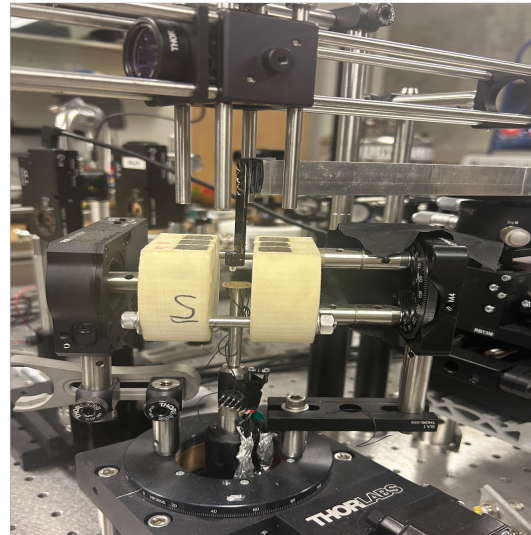
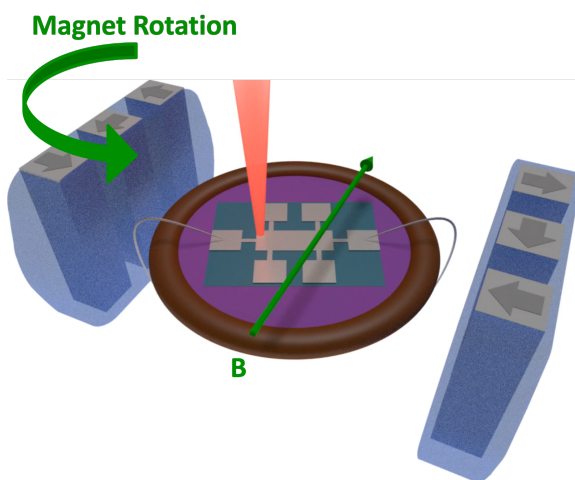


Figure 3.9: Rotational Magneto-Optoelectronic Measurement System

Chapter 4

Photocurrent streamline microscopy in micromagnetic heterostructure devices

4.1 Introduction

Photocurrent in quantum materials is often collected at global contacts far away from the initial photoexcitation. This collection process is highly nonlocal. It involves an intricate spatial pattern of photocurrent flow (streamlines) away from its primary photoexcitation that depends sensitively on the configuration of current collecting contacts as well as the spatial nonuniformity and tensor structure of conductivity. Direct imaging to track photocurrent streamlines is challenging. Here, we demonstrate a microscopy method to image photocurrent streamlines through ultrathin heterostructure devices comprising

platinum on yttrium iron garnet (YIG). We accomplish this by combining scanning photovoltage microscopy with a uniform rotating magnetic field. Here, local photocurrent is generated through a photo-Nernst type effect with its direction controlled by the external magnetic field. This enables the mapping of photocurrent streamlines in a variety of geometries that include conventional Hall bar-type devices, but also unconventional wing-shaped devices called electrofoils. In these, we find that photocurrent streamlines display contortion, compression, and expansion behavior depending on the shape and angle of attack of the electrofoil devices, much in the same way as tracers in a wind tunnel map the flow of air around an aerodynamic airfoil. This affords a powerful tool to visualize and characterize charge flow in optoelectronic devices.

Photocurrent in optoelectronic devices often adopts a long-range character: Photocurrent generation at a spatially localized laser spot can induce voltage/current signals at contacts far away, even in nonballistic electronic materials. This remote sensing of photocurrent yields a wealth of complex spatial photocurrent maps that enable the identification of local symmetry breaking[33], chirality[34], and electronic behavior in topological surface states [35, 36]. The patterns are predicted to obey the Shockley–Ramo [37]theorem for conductors[38], wherein a local electromotive force creates a global diffusion current that flows into the device contacts. For local charge photocurrent density J_c generated at a local laser spot, the global photovoltage V_{ph} is highly sensitive to the smoothly varying streamline pattern $S(r)$, which depends on both local properties (conductivity) and global boundaries of the system.

$$V_{ph} = R_G \int \vec{J}_c(\vec{r}) \cdot \vec{S}(\vec{r}) d^2\vec{r} \quad Eq.1$$

In Eq. 1, the photocurrent streamline $S(r)$ describes a spatially varying directional pattern along which spatially local photocurrent generates maximum photoinduced voltage when collected at faraway contacts; R_G is the configuration specific resistance determined by the device geometry. Eq. 1 is agnostic to the precise mechanism for photocurrent generation and has been applied to diverse settings that range from bulk quantum geometric photo-current generation to those of the thermoelectric/Nernst origin.[34, 39]

$S(r)$ values are typically derived through numerical methods, by solving an appropriate Laplace boundary value problem, and serve to phenomenologically elucidate the spatial distributions of photocurrents. The direct empirical observation of $S(r)$ poses significant challenges, necessitating precise and continuous modulation of the local photocurrent density, denoted as $j_c(r)$. Generating a localized photocurrent often entails the creation of in-plane p-n junctions, which inherently affect the photocurrent streamline field, $S(r)$. Moreover, the orientation of photocurrents at these junctions is usually fixed during the device's manufacturing process, making in situ directional adjustments impossible and thus restricting the examination of the complete vector field $S(r)$. [33, 34, 35, 36, 40] Additionally, although various methods are available for probing electronic currents at the nanoscale, such as scanning single electron transistors, magnetometers, and quantum gases, these localized measurement approaches fail to connect local photocurrent generation with overall device behavior.[41, 42, 43, 44, 45, 46, 47, 48] Despite widespread use of theoretical models, there is a pressing need for experimental methodologies that allow for the active management of current direction and provide simultaneous visualization of both global device responses

and photocurrent streamlines. Developing such techniques is crucial for deepening our comprehension of the intricate behaviors of optoelectronic devices.[41, 42, 43, 44]

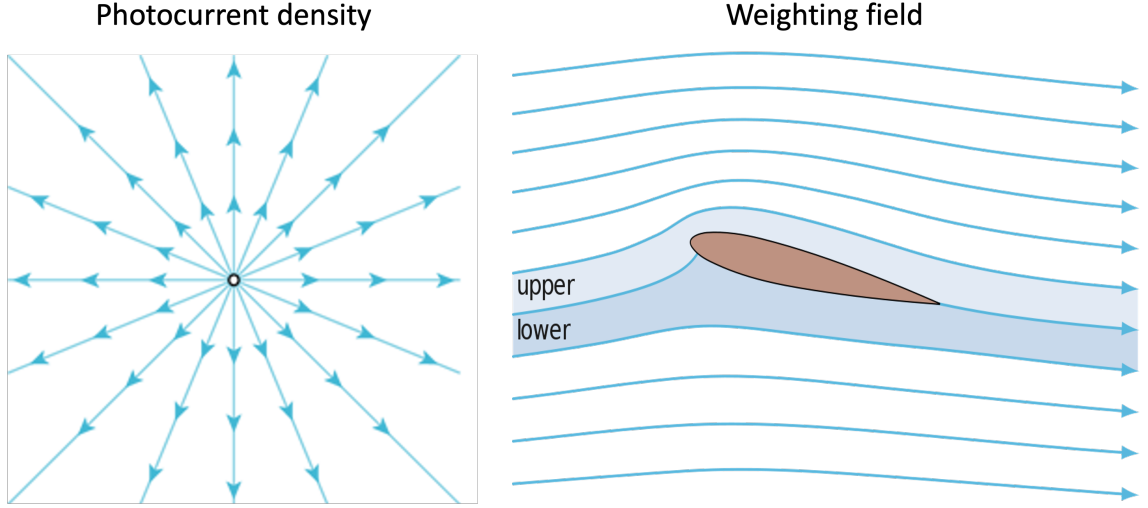


Figure 4.1: Photocurrent Density and Weighting Field

In our study, we present the direct empirical visualization of photocurrent streamlines, $S(r)$, achieved through in situ manipulation of locally induced photocurrent density within a micromagnetic heterostructure comprising a platinum (Pt) thin film on a yttrium iron garnet (YIG) substrate (Pt/YIG). The methodology for photocurrent streamline imaging introduced in experimental method section, leverages a photoinduced Nernst-type phenomenon within these micromagnetic heterostructures.[49, 50] Specifically, by employing a focused laser beam, we create an out-of-plane thermal gradient at the Pt/YIG boundary; this gradient, via a Nernst-type effect at the interface, facilitates the generation of an in-plane local photocurrent. This current's direction can be modulated by an applied in-plane magnetic field, while its spatial precision is governed by the laser's focal point. This approach circumvents the challenges associated with creating in-plane p-n junctions

by utilizing the vertical Pt/YIG interface and a lateral Nernst-type current. Utilizing this technique, we perform comprehensive empirical mappings of photocurrent streamlines across various device configurations, showcasing how the streamlines are highly sensitive to the device’s geometric design and can exhibit diverse patterns of twisting, compressing, and expanding.

4.2 Device fabrication

In this research, the devices under examination, consist of ultrathin platinum layers deposited onto a ferrimagnetic insulating substrate of yttrium iron garnet (YIG), which is epitaxially grown on gadolinium gallium garnet (GGG) substrates. Our specimens are made up of thin platinum films (5 nm) patterned atop YIG thin films, which are themselves mounted on GGG substrates. The YIG layers, measuring 80 nm in thickness, are fabricated using pulsed laser deposition techniques on GGG bases with a (110) crystal orientation. These GGG/YIG substrates were supplied by the team led by Prof. Shi at UCR. Following the verification of the atomic-level smoothness and the magnetic characteristics at room temperature of these films, we employed electron beam lithography with PMMA resist, further coated with Elektra 92 (SX AR-PC 5000/90.2) conductive resist, to etch various device configurations onto the surface of the thin film. The design for the electrofoil patterns was conceived using DesignCAD 2000, inspired by the Clark Y airfoil configuration, with each pattern adjusted at various angles from the horizontal (θ) orientation. Subsequent to the patterning phase, we conducted the deposition of 5 nm of platinum in a sputtering chamber under a base pressure of 5×10^{-8} Torr. The process concluded with

the removal of the unexposed areas in acetone, facilitating the lift-off technique. Optical imagery showcasing a selection of the resultant.

4.3 Experimental methods

Our research employed Scanning Magnetophotovoltage Microscopy (SMPM), a powerful technique built upon existing thermomagnetic microscopes. SMPM combines a strong, adjustable magnetic field with a scanning laser to study the thermoelectric response in novel micromagnetic materials at room temperature.

In this setup, the laser creates a temperature gradient within a small region of the micromagnetic device, inducing the Longitudinal Spin Seebeck Effect (LSSE). Simultaneously, we measured the resulting voltage across the device while exploring various parameters.

SMPM is fully automated, allowing control over several key aspects. Laser power is adjusted using a neutral density filter. Beam position is controlled by a galvanometer for precise 2D scanning. Beam polarization can be tuned to explore its impact. Magnetic field orientation is rotatable in both in-plane (x-y) and out-of-plane (x-z) directions.

For photovoltage generation, SMPM utilizes a high-frequency, mode-locked titanium-sapphire pulse laser (830 nm, 200 fs pulses, 76 MHz repetition rate). A rotatable Halbach array generates the magnetic field ($B = 0.52$ T at the sample). A lock-in amplifier, along with a custom DAQ system, records the photovoltage response.

Beam steering and focusing are achieved using a combination of galvo-controlled mirrors and optics. This allows for a tightly focused, scanning beam spot on the sample,

delivered either through a microscope objective or a GRIN lens. Additionally, a 50/50 beam splitter and a photodiode record the back-reflection amplitude for each measurement. The lock-in amplifier, synchronized to a 40 Hz optical chopper, measures the voltage between two terminals with a dwell time of 3.18 seconds per point.

Micromagnetic devices are mounted within the uniform magnetic field region of the Halbach array. This array, composed of precisely arranged neodymium magnets, provides high uniformity (over 9 cm²) for accurate measurements. Rotation stages allow for precise control of the magnetic field direction (90° out-of-plane and 360° in-plane). Detailed characterization ensures proper data analysis and interpretation, including verification of field and current signs within the system.

SMPM data is collected by a custom-built Data Acquisition, DAQ system and analyzed automatically using Python scripts previously developed by our group. These scripts are readily available upon request. For this study, we primarily focused on the spatial beam location and in-plane magnetic field angle as key parameters.

4.4 Results and discussion

Our investigation began with a characterization of the Pt/YIG Hall bar device. We measured the photovoltage response as a function of both laser position and the in-plane magnetic field angle denoted as θ_B . At a fixed θ_B value, the laser was scanned across the device to generate spatial maps of the reflected intensity shown in Figure 4.2 c, and the magnetic field dependence of the photovoltage, V_{ph} , see Figure 4.2 d.

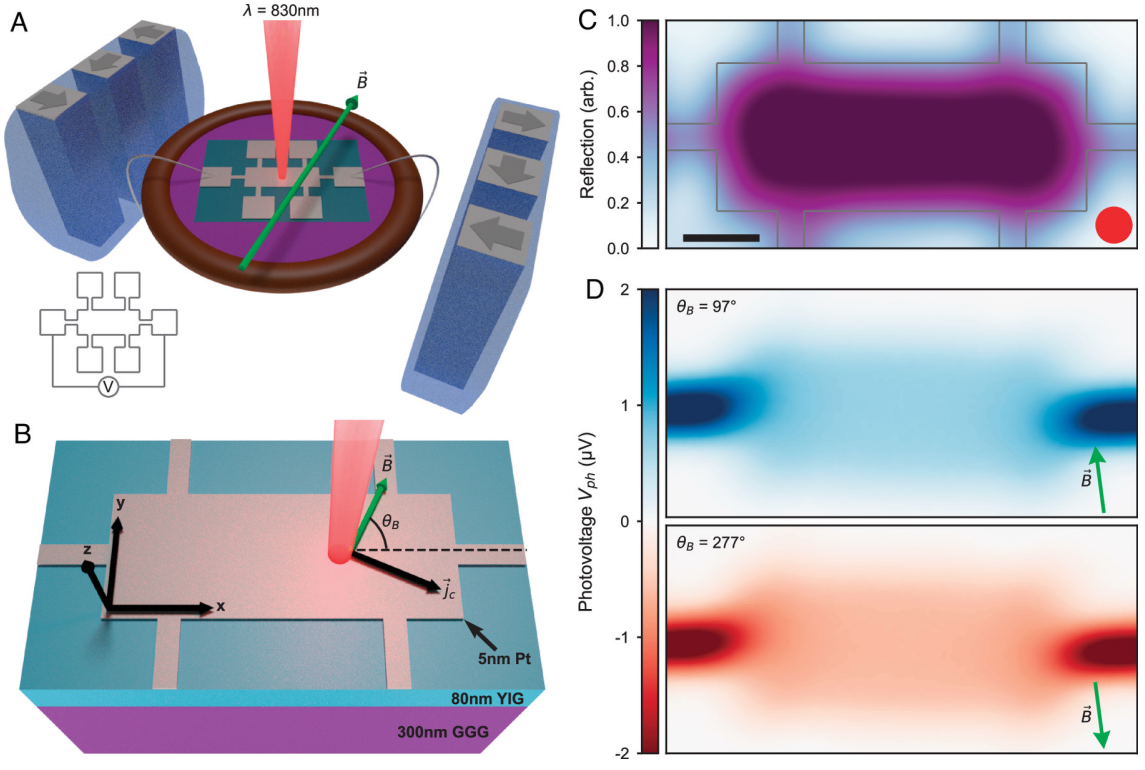


Figure 4.2: Photocurrent Streamline Microscopy of Micro magnetic Heterostructure Devices

Figure 4.2 d (Top) demonstrates the key observation. When the magnetic field (B) was oriented perpendicular to the device ($\theta_B = 97^\circ$) relative to the voltage probes positioned along the x-axis (Fig. 1B), a spatially uniform and positive photovoltage was observed. This photovoltage was further enhanced near the electrical contacts. Interestingly, rotating the magnetic field by 180° resulted in a reversal of the V_{ph} polarity (negative) while maintaining similar spatial features Figure 4.2 d (Bottom).

The photovoltage images in Figure 4.2 d can be effectively explained by a thermomagnetic photoresponse. Laser illumination generates an in-plane, local charge current density denoted by $j_c(r)$ (where r represents position). This current density is related to

the out-of-plane temperature gradient (∇T) via the Nernst coefficient (N) as described by

$$\vec{J}_c(\vec{r}) = -N \nabla T_z(\vec{r}, \vec{z}) \times \vec{B} \quad Eq.2$$

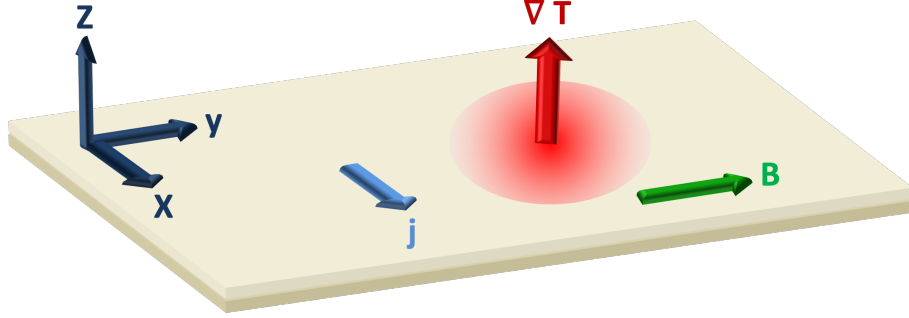


Figure 4.3: Schematic of Photo Induced Nernst Effect in Magnetic Bilayer Systems

This response is analogous to the Photo-Nernst effect reported in previously. The local charge current $j_c(r)$ flows in a direction orthogonal to both the out-of-plane temperature gradient (established across the Pt/YIG interface) and the applied magnetic field (B). Both ∇T and B are controllable parameters in our experiment. This local in-plane charge current, along with the photocurrent streamlines defined in Eq. 1, subsequently generates a photoinduced voltage across the device at the current-carrying contacts. Notably, reversing the B direction flips the direction of $j_c(r)$, leading to opposite signs of laser-induced photovoltage at the global contacts Figure 4.2. It's important to note that B in Figure 4.2 is perpendicular to the direction connecting the photovoltage contacts, as predicted by Equation 2.

The local in-plane photocurrent term in Eq. 2 presents a valuable experimental tool. By utilizing a laser beam to generate a localized interfacial temperature gradient, we can manipulate the direction of the local charge current (j_c) by rotating the magnetic field

angle (θ_B) as illustrated in the schematic of Figure 4.2 b. Figure 4.3 confirms this concept. Optoelectronic measurements captured while varying θ_B exhibit remarkable spatial patterns, with prominent photovoltage features appearing at corners and narrow constrictions of the device. These patterns are distinctly different from measurements taken with the in-plane magnetic field oriented across the device Figure 4.2 d. When the B-field is aligned parallel to the voltage probe direction ($\theta_B = 177^\circ$, Figure 4.4, Middle image), V_{ph} is suppressed in the central region, as expected from a conventional thermomagnetic response. However, novel spatial features emerge at sharp corners and along transverse contacts. As the magnetic field undergoes a 180° rotation, the polarity of these anomalous features reverses sign.

From the scanning magneto-photovoltage images, we can closely study how certain photovoltage features change as the magnetic field angle (θ_B) varies. In Figure 4.4 b, we see a scaled V_{ph} map taken at $\theta_B = 357^\circ$. For a point in the center of the device (marked by the yellow circle in Fig. 2B), we plot the photovoltage versus θ_B in Figure 4.4 c. The data fit well to a sinusoidal function

$$V_{ph} = V_m \cos(\theta_B - \theta_B^{max})$$

characterized by two parameters: the photovoltage amplitude V_m and the angle at which the photovoltage reaches its maximum θ_B^{max} . As per Eq. 2, the photovoltage at the center (yellow data in Figure 4.4 c) peaks at $V_m = 0.6 \mu\text{V}$ when θ_B is at a right angle to the x-axis ($\theta_B^{max} = 90^\circ$).

Different positions within the device show that maximum photovoltage occurs at different magnetic field alignments $\theta_B^{max}(x, y)$. For instance, if the laser is fixed at the red

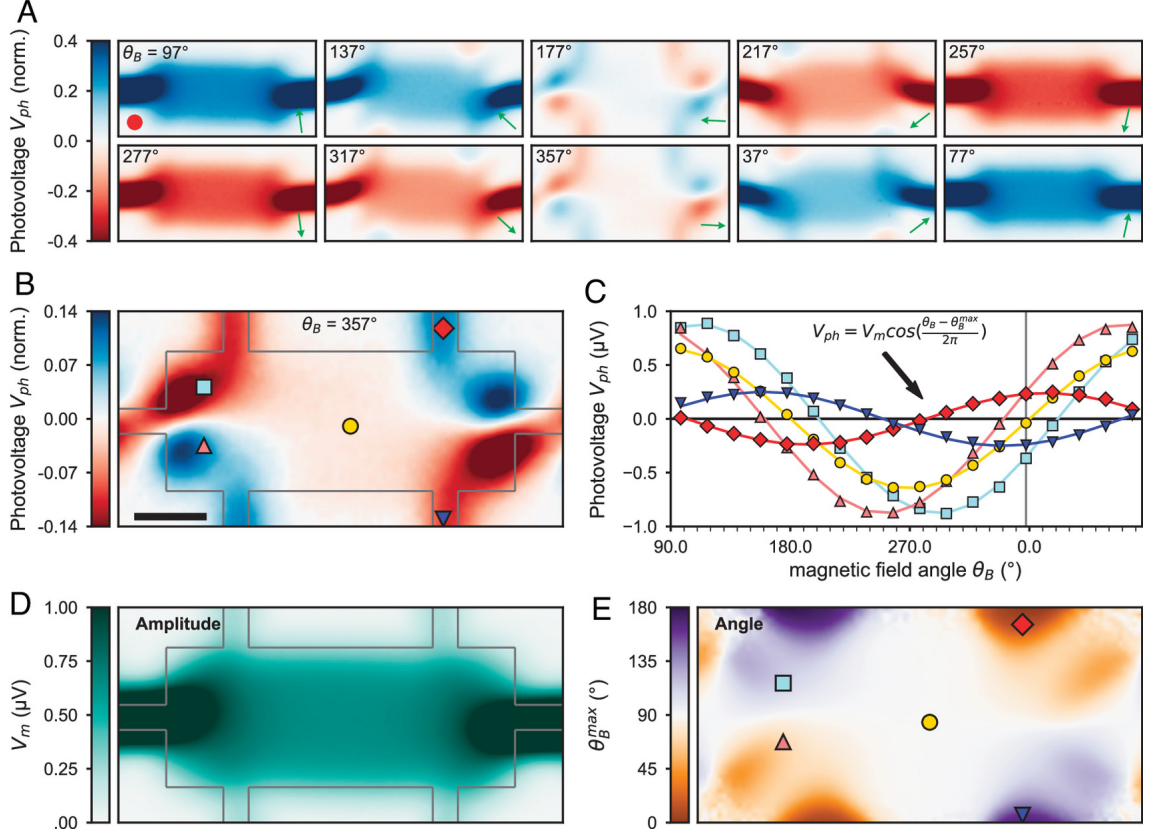


Figure 4.4: Imaging The Magnetic Field-dependent Photovoltage in a Micromagnetic Heterostructure Device

diamond at the top right in Figure 4.4 b, the photovoltage peaks when $\theta_B = 0^\circ$. Conversely, at the blue triangle at the bottom right in Figure 4.4 b, the photovoltage peaks when $\theta_B = 180^\circ$. Both spatial features exhibit sinusoidal behavior offset by a 90° phase compared to the central region (Figure 4.4 c). To visualize the amplitude and phase patterns better, we generate images of the photovoltage amplitude $V_m(x, y)$ and the angular offset $\theta_B^{max}(x, y)$ using the sinusoidal fits in Figure 4.4 c. While the amplitude map (Figure 4.4 d) looks similar to the ordinary photoresponse, the angular offset map (Figure 4.4 e) shows the magnetic field angle needed to maximize the photovoltage.

This phase map reveals a complex pattern of streamlines through the device. In Figure 4.5 a, we overlay a two-dimensional vector field on the photovoltage amplitude image $\theta_B^{max}(x, y)$. Green arrows show the direction of the magnetic field that gives the maximum photovoltage amplitude. Since the magnetic field controls the direction of the local charge current (Eq. 2), we show black arrows indicating the direction of the current j_c that yields the maximum global photovoltage. By interpolating the local current density arrows in Figure 4.5 a, we obtain a flow field through all points in the device, shown in Figure 4.5 b.

The streamlines observed in Figure 4.5 b meander smoothly within the Pt film conforming to the device boundaries. These flow lines indicate the direction of local (photoinduced) charge flow that maximizes the global photovoltage response measured at current-drawing contacts. As such, they naturally explain the photovoltage polarity changes observed at corners and near the additional side contacts in Figure 4.4 b: Local charge current that is aligned to the flow lines generates maximum positive photovoltage (red diamond Figure 4.4 b), while antialigned local charge current generates negative photovoltage (blue triangle Figure 4.4 b). Conversely, V_{ph} becomes zero when the local photocurrent is perpendicular to the photocurrent streamlines, consistent with Eq. 1. To further illustrate this, we numerically calculate $\vec{S}(\vec{r})$ for the device and perform a Gaussian convolution to mimic the finite beam spot size. Figure 4.5 c and d show the resulting field in the same manner as the experimental data. The experimental data exhibit remarkable quantitative agreement to the photocurrent streamlines expected from the Shockley–Ramo theorem (7). Importantly, when voltage reading leads are reconfigured (e.g., by switching voltage readout to other contact pads in our device), the experimentally obtained photocurrent streamlines

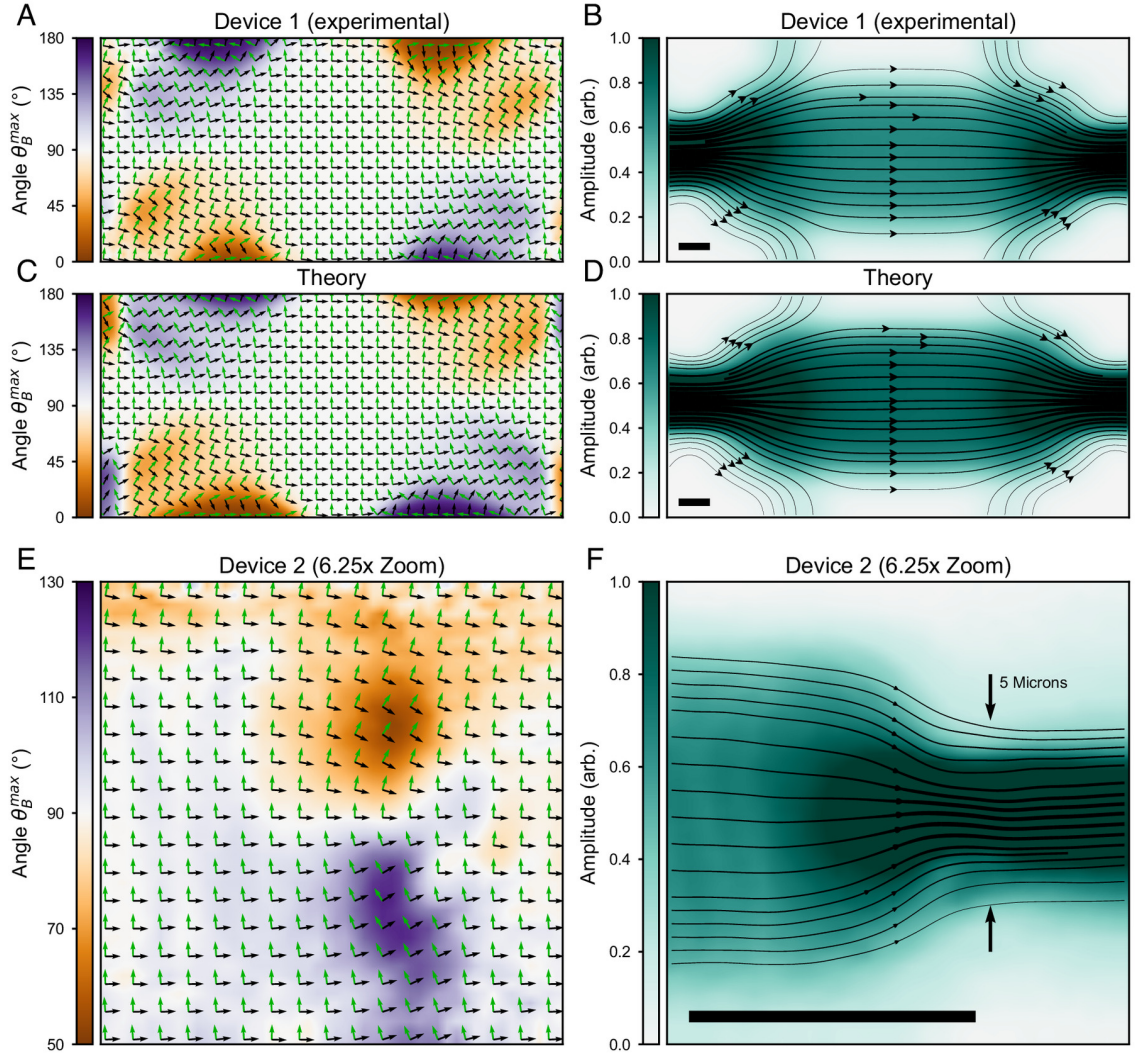


Figure 4.5: Imaging Photocurrent Streamlines Through a Micromagnetic Heterostructure

similarly contort to favor strong photocurrent streamlines that directly connect the reconfigured voltage readout pads in accordance with that expected from Shockley-Ramo theory. Such examples vividly demonstrate the ability of our technique to sense different readout configurations as well as the concomitant direction of the preferred current flow.

These photocurrent streamlines are compressed within the Pt film as the boundaries become more restrictive. As shown in Figure 4.5 b, the photocurrent streamlines are

highly sensitive to the device geometry: Photocurrent streamlines are uniform and dilute far from the boundaries yet converge and stream into a high-density bundle near narrow constrictions. To further illustrate this effect, we repeated the same measurements on a significantly smaller device using a nearly diffraction-limited beam spot to allow higher resolution measurements. With a device less than one-sixth the original size, Figure 4.5 f shows the same pattern of current compression near the contact as in Figure 4.5 e. For a finite spot-size laser illumination, the density of photocurrent streamlines corresponds directly to the intensity of the photovoltage response. As shown in Figure 4.5 b and f, V_{ph} is strongly enhanced in regions of high photocurrent streamline density (e.g., along the narrow horizontal contacts) and suppressed in regions of low density.

In addition to the dramatic current compression examined in Figure 4.5, photocurrent streamlines—and the global photoresponse—can be manipulated far away from device boundaries in much the same way as fluid streamlines are guided to flow around an airplane wing. To test this, we repeated our experiment on specially fabricated devices with cross-sectional wing-shaped cut-outs, or electrofoils. Apart from an unpatterned blank device that is used as a control (Figure 4.6 a, Left), the devices were created by removing regions of the Pt film in the shape of aerodynamic Clark Y airfoils (27) (Figure 4.6); the electrofoils exhibit a convex upper profile and flat lower surface (gray outlines in Figure 4.6 a), and each is fabricated with an increasing angle of attack. Similar to the Hall bar device, we measure V_{ph} vs. θ_B and fit the resulting sinusoidal characteristics to obtain $V_m(x, y)$ and $\theta_B^{max}(x, y)$ (Figure 4.6 a and b). From these maps, we then produce images of the photocurrent

streamline flow through the electrofoil devices (Figure 4.6 c) using the protocol described above.

Figure 4.6 d compares a detailed image of the photocurrent streamline flowing over the top surface of the 35° electrofoil to the theoretical expectations (Figure 4.6 e). At such a high angle of attack, photocurrent streamlines curve sharply to traverse the electrofoil (black lines Figure 4.6 d), creating regions of high- and low-density current flow. The photovoltage amplitude V_m in Figure 4.6 d reaches a peak value above the leading (Left) edge of the electrofoil. By introducing the asymmetric cambered boundary, photocurrent streamlines are forced to curve more sharply around the leading edge of the electrofoil compared to the trailing edge. In this way, we gradually manipulate the photocurrent streamline density—and thus photovoltage intensity—along the length of the electrofoil surface.

4.5 Summary and outlook

Imaging the photocurrent streamline field $\vec{S}(\vec{r})$ gives a unique fingerprint for each device, clearly identifying the lines along which photocurrent naturally flows. We emphasize that these photocurrent streamline patterns—while device specific—are agnostic to the mechanism through which spatially local photocurrents are generated. The experimental data of Figure 4.6 d directly visualize these natural photocurrent streamlines inside the electrofoil devices. Even as $\vec{S}(\vec{r})$, strictly speaking, only visualizes the directions along which spatially local photocurrent produces maximal photovoltage at remote contacts (an optoelectronic device property), it is nevertheless interesting to consider how it relates to

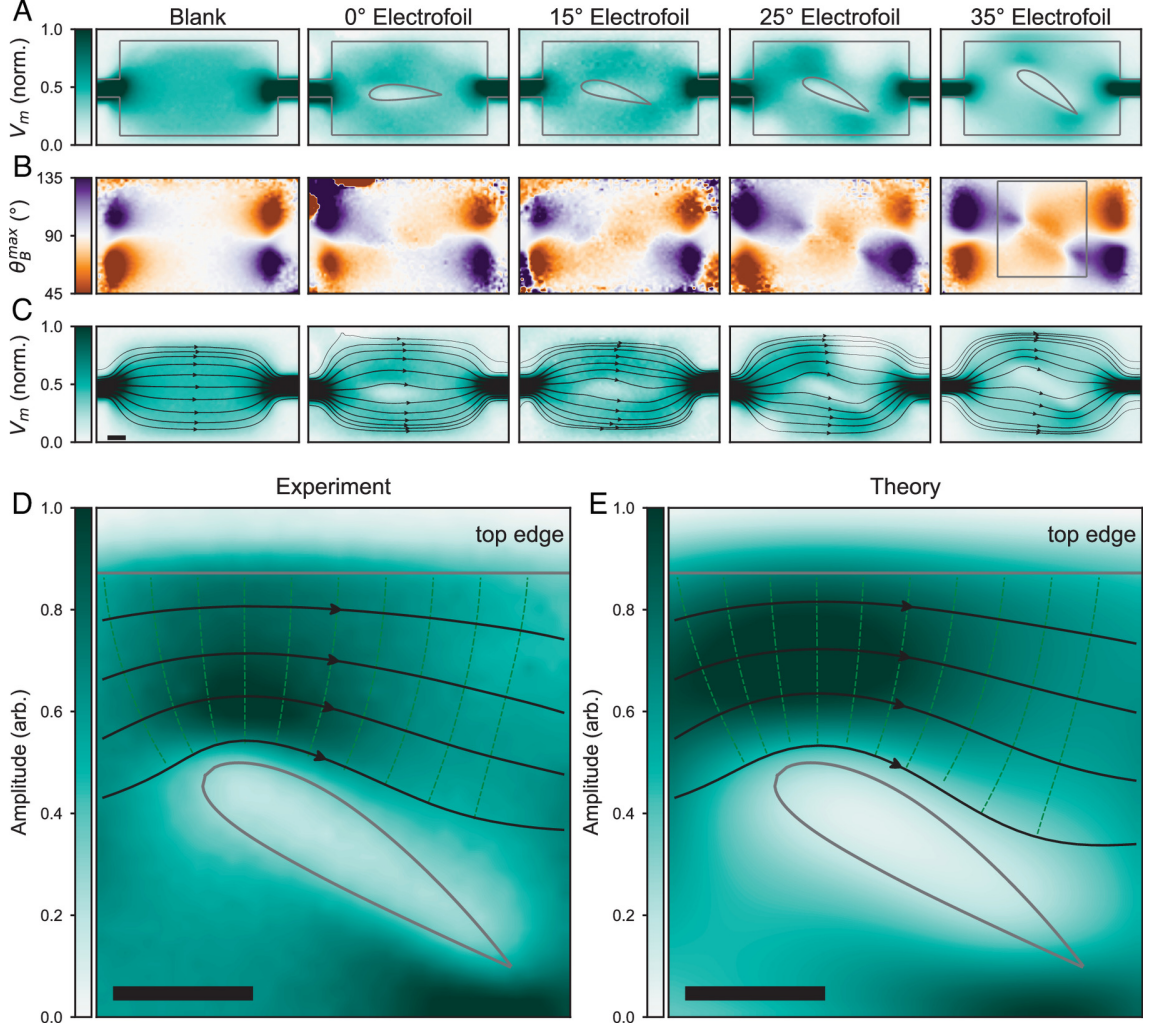


Figure 4.6: Contortion, Compression, and Expansion of Photocurrent Streamlines in a Micro-magnetic Wind Tunnel

other device characteristics. For instance, in the absence of an in-plane Hall effect, $\vec{S}(\vec{r})$ can mimic the electric field spatially distribution of the device when it is operated purely electrically [51] (i.e., when only electric current is flowed from one contact to another without photoexcitation). Since $\vec{S}(\vec{r})$ can be contorted and compressed (as shown in Figure 4.5 and 4.6) close to sharp corners and near constricted geometries, we anticipate that the electric field distributions (when operated electrically) close to these regions are similarly morphed.

This is especially important since the spatial distribution of electric fields, especially in nonuniform systems, can dictate the presence of nonuniform joule heating that affect device performance, e.g., for spintronics in metal/magnetic material systems [52, 53]. With imaging resolution spanning over an order of magnitude in spatial scale, we have shown that photocurrent streamline microscopy is a robust experimental tool for obtaining detailed visualization of photocurrent in quantum materials. The key requirements for this tool are a spatially tight laser illumination as well as directional control of local photocurrent (in our case, through B-field manipulation); when combined, this enables detailed visualization of global photocurrent streamline fields. While we have used a photoinduced Nernst effect in a micromagnetic device to enable directional control of the local photocurrent, other means of directional control of local photocurrent can be employed as well. For example, the bulk photovoltaic effect [54, 55] features local laser-induced photocurrent that has a direction that is sensitive to the polarization and helicity of light [56]. Combined with a tight laser spot size, we anticipate that such light polarization control of the local photocurrent direction can also allow the ability to trace out the photocurrent streamlines in a wide variety of material systems that support the bulk photovoltaic effect [55]; the key material requirement is the breaking of bulk inversion symmetry. Another exciting future prospect is the ability to exploit the sensitivity of $\vec{S}(\vec{r})$ to the details of the conductivity tensor [51]. For instance, irradiation incident on a sharp tip can produce near-field and highly localized photoexcitation [57] with extremely spatially tight local photocurrent generated; this may enable to probe nonlocal quantum geometric corrections to the conductivity [58, 59, 60] are difficult to access with conventional transport methods.[61]

Chapter 5

Layer photovoltaic effect in a gapless van der Waals semimetal heterojunction

5.1 Motivation

Photocurrent is a powerful probe of electrons, holes, and their interactions in van der Waals (vdW) solids. In conventional materials, photovoltaic effects occur in semiconductor systems with a band gap, while photothermoelectric effects are expected to dominate in gapless metals and semimetals. In 2D semiconductors, light generates voltage via the photovoltaic effect, giving direct experimental access to the valley degree of freedom [62, 63, 64, 65, 66], highly interacting electron-hole phases [67, 68, 69], and interlayer exciton transport and trapping [70, 71]. This contrasts with photocurrent generation in graphene

- a Dirac semimetal - where light energy heats electrons and holes, giving rise to a photo-thermoelectric response [72, 73, 74, 75, 76, 77, 78]. Here, we find a layer photovoltaic effect that emerges in a hybrid semimetal heterojunction composed of WTe₂ - a compensated electron-hole Weyl semimetal - and graphene. We investigate the interlayer photocurrent at the WTe₂-graphene interface as a function of applied gate voltage V_g and laser power. Comparing the interlayer photoresponse to the expected behavior of the photothermoelectric effect, we find that photocurrent does not arise from a thermoelectric origin, particularly when the system is tuned near charge neutrality. Detailed investigation of the interlayer photoconductance reveals a sharp enhancement near the WTe₂-graphene charge neutrality point. This enhancement exhibits a shift in V_g as it increases with laser power and occurs in precisely the regime in which electrons and holes form a strong layer polarization under photoexcitation. From the inferred band alignment, we attribute this striking photoconductance enhancement to the interlayer photovoltaic effect. Our observation enables a unique view of the intrinsic photoresponse in Weyl-Dirac semimetal heterojunctions and heralds the emergence of next-generation infrared sensor technologies based on hybrid semimetal vdW solids.

5.2 Introduction

In the field of 2D material optoelectronics, the investigation of pure semimetal heterojunctions composed of atomically thin materials presents a new frontier of research. These heterojunctions exhibit unique photocurrent characteristics, which have important implications for various photogeneration effects such as photo-thermoelectric and photo-

voltaic processes. Two-dimensional materials, including graphene and transition metal dichalcogenides, have garnered significant attention due to their exceptional optical, and electrical properties. [79, 80, 81] graphene has shown great promise for applications in photodetection due to its high charge carrier mobility and broadband absorbance in the near-infrared and visible spectral ranges. However, the semi metallic nature of graphene with zero bandgap caused limitations on its application in photodetection.[82, 83] On the other hand, WTe₂ exhibits a type-II Weyl semimetal state, characterized by its distinctive electronic band structure. The material's band structure features pairs of Weyl points boasts a distinctive feature in its band structure—the presence of charge-compensated points. These points, known as charge-compensated Weyl nodes, contribute to the material's exotic electronic properties.[84, 85, 86, 87] In terms of optical properties, WTe₂ demonstrates strong light-matter interactions, making it a promising candidate for optoelectronic applications.[88, 89] The material's electrical conductivity is noteworthy, with a high carrier mobility and a significant anisotropy in its electronic transport properties. The combination of its unique band structure and exceptional electrical and optical characteristics positions WTe₂ as a promising material for the development of novel electronic and photonic devices, potentially paving the way for advancements in next-generation technology.[90, 91, 92] When graphene is bonded to WTe₂ through weak Van der Waals interactions, the excess flow of non-equilibrium carriers can move more directionally, leading to enhanced photoresponsivity in the band.[93] However, the investigation of photocurrent characteristics in pure semimetal heterojunctions remains relatively unexplored. In this paper, we propose to investigate the photocurrent characteristics of a exotic heterojunction consisting of a Dirac

semimetal and a charge-compensated Weyl semimetal. Our study aims to understand the dynamics of photoexcited electrons in this heterojunction and elucidate the electrical and optical conductivities at the interface. To achieve this, we employ near-infrared scanning photocurrent microscopy, which provides us with high-resolution imaging of the photoexcited electron dynamics. Furthermore, we aim to clarify the heterojunction band alignment between WTe₂ and graphene, which have charge-compensated electron-hole bands and a Dirac point, respectively. We expect that our findings will contribute to potential applications in photodetection and energy harvesting.

5.3 Experimental methods

Once the device is prepared, it will be installed within a cryostat equipped with electrical probes that establish and stabilize electrical connections. These probes will facilitate the tuning of the backgate voltage and the measurement and tuning of the source/drain voltage. Subsequently, the cryostat lid will be securely closed, and the internal pressure will be reduced to approximately 10^{-7} torr, thereby stabilizing the device.

The experimental setup described in Chapter 3 (MPDPM) will be employed to focus a laser beam on the device region of interest or to scan across the entire device surface. The intensity of the laser beam can be modulated using a filter.

5.4 Results and discussion

As schematically shown in Figure 5.1 a the studied devices are made of layered material starting with backgate graphene, electrical insulator hexagonal Boron Nitride and

the main interface graphene and WTe₂ which are each electrically connected to a gold metal contact separately and a top hBN layer serves as a protective coating to enhance both the quality and lifespan of the device. Detailed information regarding the fabrication steps and methods employed for device construction can be found in the Method section. The measurement of resistance (dark current) in the device, reflecting the conduction between WTe₂ and graphene, exhibited the anticipated dependence on Source/Drain voltage (V_{sd}). Simultaneously, adjusting the gate voltage (V_g) provided the capability to fine-tune the chemical potential. The resistance curve, depicted in Figure 5.2 a, reveals a Dirac-shaped behavior at the backgate around zero. [94] This can be interpreted as the Dirac point in the graphene band structure or the comprehensive charge compensation between WTe₂ and graphene, as illustrated in Figure 5.1 c.

Using a photocurrent microscopy system designed and constructed in-house, which is illustrated in detail in the experimental method section, we illuminate the device with NIR laser and systematically scan different locations, including the interface (2), graphene (1), and WTe₂ (3), as illustrated in Figure 5.1 a and d. Subsequently, we measure the photocurrent generated at each specified location, producing a photocurrent map shown in Figure 5.1 d. The regions where the device is positioned exhibit a high photoresponse, with negative observed in the WTe₂ region (3) and interface region (2), and positive in the graphene region (1). These photocurrent mapping results underscore the high photoresponse of the graphene/WTe₂ heterostructure compared to pure graphene/contact [93]. The enhanced photoresponse implies the joint contribution of both photovoltaic and photo thermoelectric effects to the generation of photocurrent in the heterostructure.

Fig. 1.

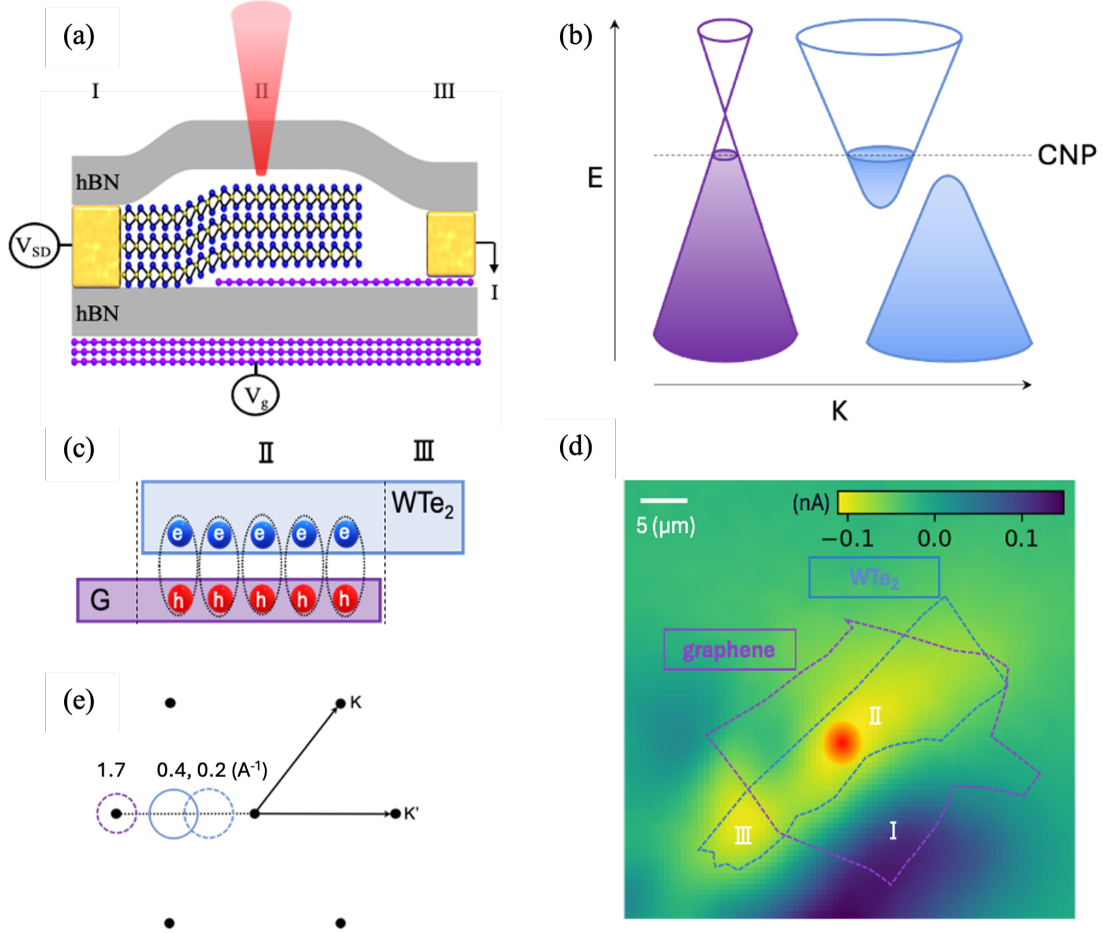


Figure 5.1: a. Device's Schematic, consist of backgate graphite, monolayer graphene and WTe₂. b, Schematic of proposed band alignment between graphene and WTe₂ in k space. c, Charge balance between graphene and WTe₂. d, Photocurrent map, showing three main areas. e, Graphene Dirac point (purple) and WTe₂ charge neutrality point.

Fig. 2.

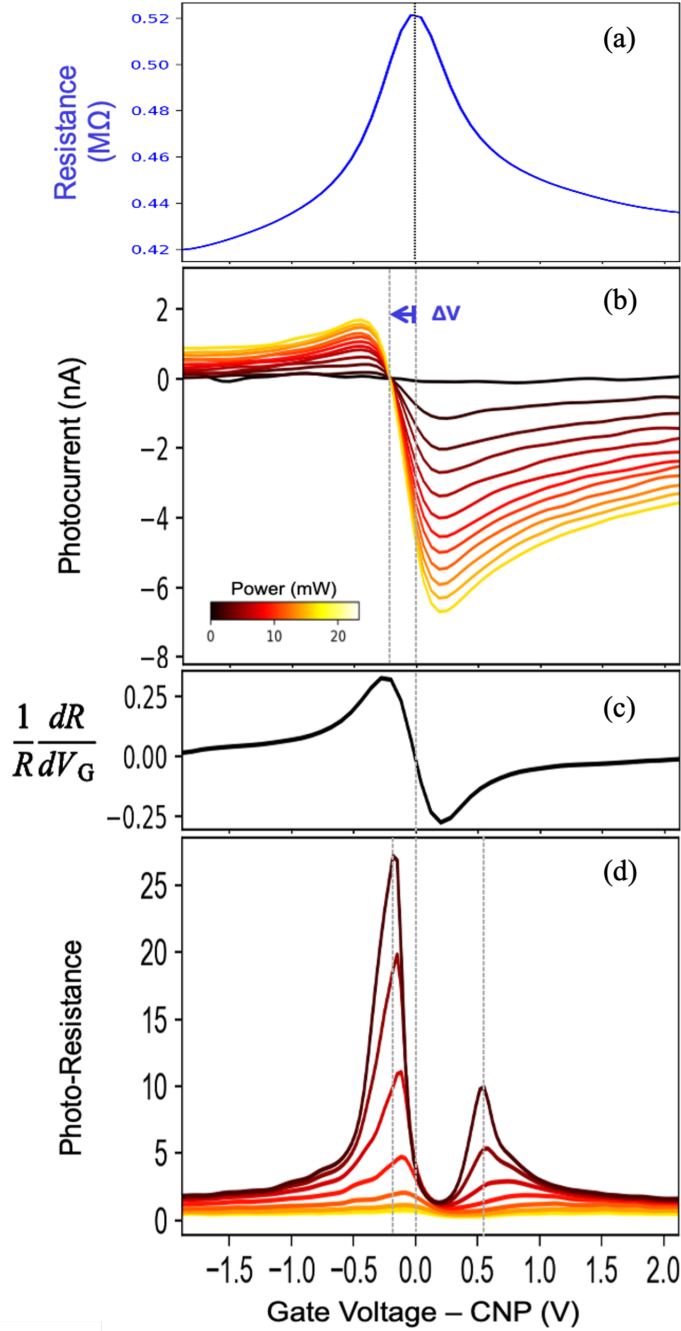


Figure 5.2: a, Dark resistance curve vs backgate voltage - CNP voltage. b, Photocurrent at the interface (region 2) as a function of backgate voltage - CNP voltage, different colors represent different incident laser power. c, Calculated seebeck coefficient based on resistance. d, Photo resistance as a function of backgate voltage - CNP voltage

For visualization purposes we shifted the data to the right so the peak location in resistance locates at zero and we named it Gatevoltage - $V_{\text{Charge Neutrality Point (CNP)}}$ which now peak is at zero. In Figure 5.2 b, we presented the photocurrent generated at the interface location plotted against the backgate - V_{CNP} , varying the laser power from 0 to 17.83 mW. While the Z shape photocurrent bears some resemblance to the Seebeck S shape, notable differences exist. This indicates that the photo-thermoelectric factor is a significant contributor to photocurrent generation at the interface, but it is not the sole factor. The photocurrent not only exhibits asymmetry with distinctly different peak and valley but also displays a non-monotonic pattern towards the two extremes of the backgate- V_{CNP} axis. Interestingly, the charge neutrality point (CNP) in photocurrent which is the crossing point, where there is no change in current at different laser powers, does not align with the resistance peak, and there is a shift to the left that we called it ΔV , this cause negative current at backgate- V_{CNP} equal to zero that we expected zero current.

The variation in laser power illustrates how the increased excitation of electrons and holes contributes to the generation of photocurrent (PC) also it creates perpendicular temperature gradient within the system. In this interface, the distinction between higher and lower power levels in PC generation is more pronounced at chemical potentials above the charge neutrality point (CNP) compared to chemical potentials below CNP, as depicted in Figure 2b.

According to the theory, the photocurrent's behavior is directly linked to the Seebeck coefficient as per Eq 2, assuming that the thermoelectric effect is the sole contributing factor to the generation of photocurrent within the system.

To delve deeper into the contribution of the photo thermoelectric effect in producing photocurrent at the interface, we computed the Seebeck coefficient using Eq 1 derived from the dark resistance depicted in figure 5.2 c. The Seebeck coefficient exhibits a symmetric S shape pattern with a peak and a valley, demonstrating comparable peak locations and heights. Additionally, a monotonic behavior is observed on the opposite side of the backgate - V_{CNP} equal to zero along the x-axis, indicating a consistent effect arising from both electrons and holes.

To understand the dominant mechanism behind this photogeneration, we plotted the photoresistance, which is essentially the slope of the photocurrent as a function of source voltage, against the backgate voltage (see Figure 5.2d). The figure reveals two peaks in the photoresistance, neither of which aligns with the dark current resistance shown in Figure 5.2a. The first peak occurs in the negative backgate region, near $-V_{\text{CNP}}$, and aligns with the photocurrent cross-section in Figure 5.2b, representing the overall charge neutrality point. The second peak appears in the positive region. Referring to the proposed band alignment in Figure 5.1b, the best explanation for this behavior is that the second peak results from the Dirac point of graphene. This indicates that the WTe_2 charge carriers, which are all n-type in this case, are responsible for photogeneration, causing the system to act like a perfect dipole and leading to the observed peak in photoresistance. The first peak corresponds to the WTe_2 charge neutrality point, where p-type carriers in graphene form a dipole, resulting in a decrease in photocurrent. Figure 5.3.

An interesting region exists between these two peaks, where the overall charge neutrality point occurs. Although this system consists entirely of semimetals, the charge

balance between the two materials behaves like a pn junction in semiconductors, suggesting enhanced photogeneration in this area. To further investigate, we plotted the photoconductance as a function of backgate voltage, which clearly shows a peak in this region, supporting our hypothesis. Figure 5.3 c.

In the final measurement, we increased the laser power and observed this peak. Contrary to our expectations, the peak shifted to the right as the power increased. This shift is due to the different relaxation dynamics in graphene versus WTe₂ or the heterostructure. In graphene, photo-excited carriers only need to lose energy to return to their initial band, whereas in WTe₂ or the heterostructure, carriers must lose both energy and momentum. This difference in relaxation time, with graphene being faster than WTe₂ and WTe₂ faster than the heterostructure, leads to an excess of charge in WTe₂ as laser intensity increases. As a result, the peak shifts to a higher backgate voltage in order to return to the PN junction state. Figure 5.3 b and d.

Fig. 3.

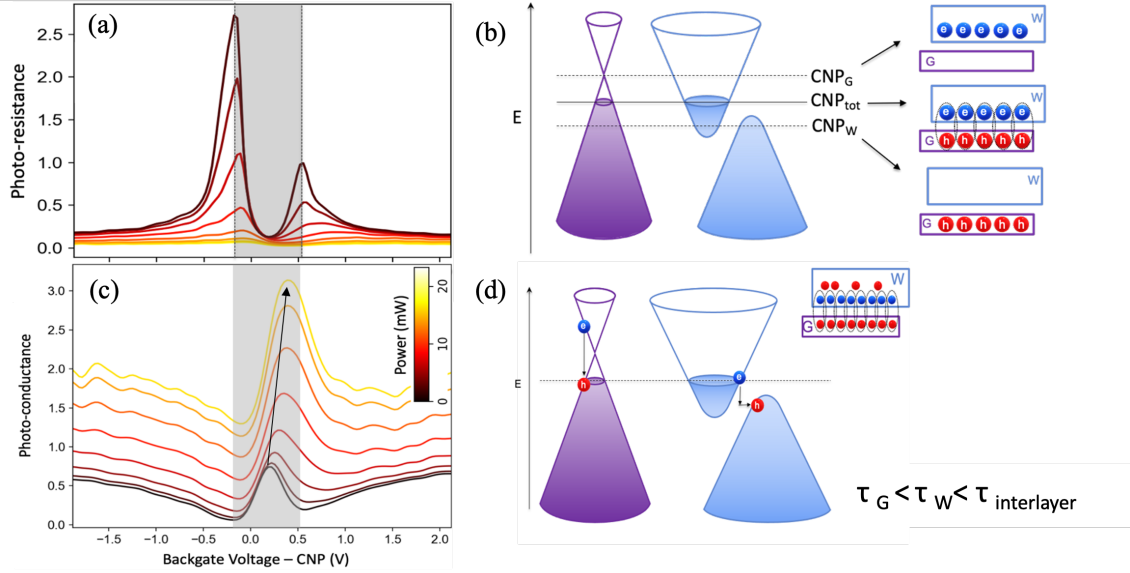


Figure 5.3: a, photoresistance as a function of backgate- V_{CNP} presenting two peaks and an interesting area in between them which aligns well with c, The peak of photocurrent as a function of backgate- V_{CNP} b, schematic of charge carriers (p type and n type) densities in different materials' band alignment at different chemical potentials d, comparing relaxation time of photoexcited carriers in different materials

To explore deeper, we studied the relationship between photocurrent and laser power at different chemical potentials, as shown in the Figure 5.4. The investigation reveals that the power-law behavior is sublinear/ nonlinear which is another proof to the conclusion that photo-thermoelectric power is not the only carrier generator at this interface.

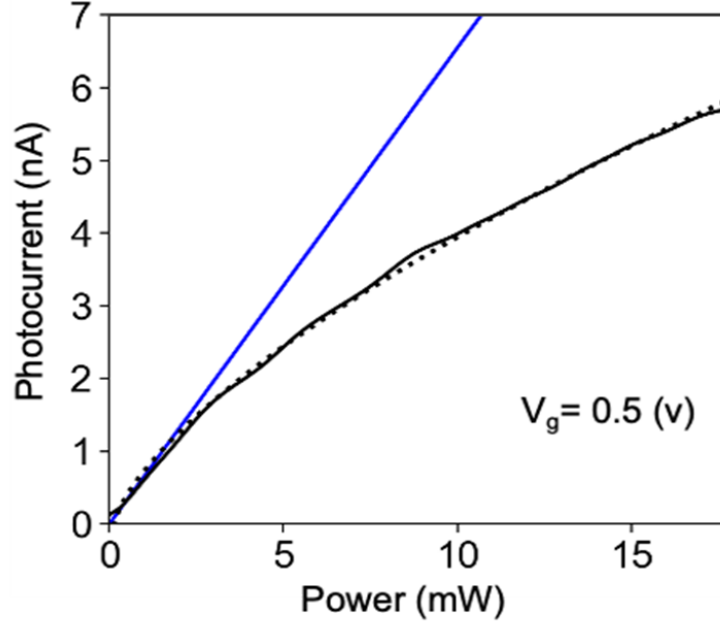


Figure 5.4: Photocurrent Power Dependence

5.5 Summary and outlook

In this chapter, we have explored the nature of photocurrent generation at the interface of van der Waals (vdW) semimetal systems, specifically focusing on the WTe₂-graphene heterojunction. Our findings reveal that traditional photothermoelectric effects, which are well-documented in gapless semimetals like graphene, are not the sole mechanism responsible for photocurrent in these systems. Instead, we have identified a layer photovoltaic effect that emerges uniquely in the WTe₂-graphene interface. This discovery indicates that the phenomena associated with PN junctions, previously observed exclusively in semiconductor systems, can also occur in semimetal interfaces, provided that the band alignment is suitably tuned.

The investigation demonstrates that the photocurrent response at the WTe₂-graphene interface does not originate from thermoelectric effects, especially when the system is near charge neutrality. Instead, a significant enhancement in photoconductance is observed near this neutrality point, suggesting a robust interlayer photovoltaic effect. This effect is attributed to the strong layer polarization of electrons and holes induced by photoexcitation, which dramatically influences the photocurrent behavior. The dependence of this enhancement on applied gate voltage and laser power further supports the notion that the observed photocurrent is due to photovoltaic rather than thermoelectric mechanisms.[95]

Our results open new avenues for research into the electronic and optoelectronic properties of semimetal heterojunctions. Future studies will aim to extend these findings to other semimetal interfaces to validate the generality of the observed phenomena. Additionally, we plan to explore the behavior of these systems at lower temperatures to uncover potential new mechanisms that might emerge under such conditions. By doing so, we hope to deepen our understanding of photocurrent generation in vdW semimetal systems and pave the way for advancements in infrared sensor technologies and other applications based on hybrid semimetal vdW solids.

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Chapter 6

Conclusion

This dissertation has explored two distinct but complementary aspects of photocurrent behavior in advanced materials: We initially introduced a mapping technique to locally image electron flow at the interface of a metal and a and then we studied the generation of this photocurrent when we lock the direction of its flow at the device design level. the detailed imaging of photocurrent flow and the characterization of photocurrent mechanisms in van der Waals (vdW) semimetal systems.

In the first chapter, we developed and utilized a novel photocurrent streamline imaging technique to visualize the spatial distribution of photocurrent within devices. This approach provided a unique fingerprint for each device, revealing the natural paths along which photocurrent flows. Our method, capable of high-resolution spatial imaging, demonstrated that photocurrent streamline patterns can mimic the electric field distribution in the absence of an in-plane Hall effect. This is particularly useful for understanding the effects of device geometry on photocurrent behavior. The insights gained from these patterns

are critical for optimizing device performance and for studying nonuniformities that could impact applications such as spintronics and quantum materials. Additionally, the ability to control local photocurrent direction through techniques like photoinduced Nernst effects or light polarization opens exciting avenues for future research and practical applications.

In the second chapter, we investigated the nature of photocurrent generation in the WTe_2 -graphene heterojunction, focusing on the emergence of a layer photovoltaic effect. This study revealed that, contrary to traditional expectations, the photocurrent in this system is not solely driven by thermoelectric effects but is significantly influenced by a robust interlayer photovoltaic effect. This finding suggests that similar phenomena to semiconductor PN junctions can occur in semimetal interfaces when the band alignment is suitably tuned. The observed photocurrent enhancement near charge neutrality, driven by strong layer polarization under photoexcitation, challenges previous understandings of photocurrent mechanisms in semimetals and opens new research directions.

By integrating these findings, this dissertation contributes to a deeper understanding of photocurrent mechanisms and their implications for device design and functionality. The ability to visualize photocurrent flow and identify underlying mechanisms in semimetal systems provides valuable insights that can drive the development of advanced optoelectronic devices. Future research will build on these insights to explore other semimetal interfaces and investigate behaviors at lower temperatures, aiming to advance technologies such as infrared sensors and hybrid vdW materials.

In summary, this work highlights the importance of both innovative imaging techniques and the exploration of new photocurrent mechanisms, providing a comprehensive

foundation for future research and technological advancements in the field of quantum materials and optoelectronics.