ELECTRONIC TRANSPORT AND STRUCTURE OF A SURFACE QUANTUM WELL

BY

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DISSENTATION

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Abstract

Lattice-mismatched heteroepitaxy is present in the growth of any thin-film heterostructure, and has inevitable consequences on the nature of film growth. A mismatch in the lattice parameter between two materials leads to strain within both films, which can lead to dislocations and deformations in the lattices. The formation of dislocations is a complex phenomenon with both thermodynamic and kinetic influences. In this work, a structural study of antimonide buffer layers on GaAs substrates was performed. Using *in situ* diffraction and *ex situ* microscopy, we examine the lattice transition between GaAs substrates, and the highly mismatched material GaSb. When GaSb grows on GaAs, it follows a complex Stranski-Krastanov 2D to 3D transition. Using our real-time diffraction data, we determine that the change in lattice constant and the transition to 3D growth occur simultaneously.

Our work on the growth of antimonides is motivated by the desire to grow high quality InAs films. InAs is closely lattice-matched to the antimonide compounds studied, making them candidates for the composition of buffer layers on which to grow InAs. The superconducting proximity effect in a highly spin-orbit coupled material such as InAs is key to the realization of Majorana zero modes and future advances in topological computing. The surface accumulation layer that InAs forms when in contact with a superconductor makes it ideal for proximity effect-based devices, as the carriers are physically connected to the superconductor.

We grew and measured InAs surface quantum wells grown on GaSb buffers, characterizing their electronic properties and preparing measurements of the proximity effect in the wells. While electrons in the InAs surface quantum wells are scattered more than those buried InAs
quantum wells, an electron mean free path approaching 100 nm was obtained. Fabrication methods are being developed for the creation of Josephson junction arrays on our InAs films, with which the proximity effect can be studied.
In memory of Dr. Lynda Bradley-Vacco.
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# Table of Contents

Chapter 1  Motivation ........................................... 1
  1.1 Lattice-mismatched epitaxy ................................. 1
  1.2 InAs surface quantum well transport ..................... 3
    1.2.1 Proximity effect .................................. 4
    1.2.2 Josephson junction arrays ......................... 5
  1.3 Thesis outline ........................................... 6

Chapter 2  Background .......................................... 8
  2.1 Dislocations in GaSb on GaAs ............................ 8
    2.1.1 Stranski-Krastanov growth ........................ 8
    2.1.2 Interfacial misfit dislocations .................... 12
  2.2 Transport in InAs surface quantum wells ................ 13
    2.2.1 InAs surface accumulation layer .................. 13
    2.2.2 Proximity effect .................................. 14
    2.2.3 BKT transition in Josephson junction arrays ...... 15
  2.3 Majorana fermions ........................................ 16
    2.3.1 Majorana, Dirac, and Kitaev ....................... 16
    2.3.2 Emergent Majorana states ......................... 20
    2.3.3 Proposed Majorana heterostructures ............... 21
    2.3.4 Experimental background: Majorana nanowires .... 24

Chapter 3  Experimental Methods ............................... 28
  3.1 Molecular beam epitaxy .................................. 28
    3.1.1 System B .......................................... 28
    3.1.2 Ultra-high vacuum ................................ 31
    3.1.3 Growth of GaAs .................................... 33
    3.1.4 Generalizing GaAs techniques to antimonide growth . 37
  3.2 Film analysis techniques ................................. 39
    3.2.1 RHEED ............................................ 39
    3.2.2 Transport measurements ............................ 43
    3.2.3 AFM ............................................... 44
    3.2.4 TEM ............................................... 45
Chapter 1
Motivation

1.1 Lattice-mismatched epitaxy

In general, perfectly lattice-matched epitaxial growth would be the ideal for any study of thin films, as when growth occurs on mismatched substrates, strain can cause the crystal to dislocate in unpredictable ways. This has consequences for the resulting material quality, for example by adversely affecting the material’s mobility when dislocations act as scattering sites. The ideal substrate would not only exactly match the lattice of the target material in size and crystal structure, but would be highly insulating, so that transport measurements would only probe the properties of the film itself.

However, many technologically significant material systems have no such perfect substrate available. For this reason, lattice-mismatched heteroepitaxy is an essential technique in thin film experiments. Prominent examples include the growth of Ge on Si or InP on GaAs, the formation of quantum dots in a number of materials, and many varieties of superlattices and other heterostructures[1]. The entire 6.1 Å family, consisting of the three nearly lattice-matched III-V semiconductors InAs, GaSb, and AlSb, are a set of materials with a variety of band structures and energy gaps. This makes them useful in a wide array of heterostructure applications, ranging from field effect transistors to broken-gap quantum wells and type 2 superlattices[2]. However, there is no insulating substrate lattice matched to these materials, and their electrical and quantum behavior is adversely affected by a conducting shunt. Therefore, it is preferable to use an insulating substrate and attempt to optimize the growth dynamics despite the lattice mismatch.
The search for Majorana modes and other topological physics necessitates growth of high quality InAs- and InSb-based layers, motivating our study. Both materials have a large Rashba spin-orbit coupling: $\alpha \approx 50 \text{ meV} \cdot \text{Å}$ in InAs and $\alpha \approx 200 \text{ meV} \cdot \text{Å}$ in InSb[3][4]. This serves to separate spin bands widely enough to eliminate spin as a degree of freedom in certain regimes, essential to realizing Majorana modes.

For this work, we have chosen to work with InAs, as it is closely lattice matched with the antimonide materials GaSb and AlSb, which would make ideal substrates for its growth by MBE. Unfortunately, both GaSb and AlSb are unsatisfactory as substrate materials for Majorana applications. GaSb, while commercially available in wafer form, is highly conductive even when unintentionally doped. The substrate effectively shunts transport measurements made on the eventual InAs film and makes it difficult to extract the properties of the film alone. AlSb is much more insulating, but is highly reactive due to its aluminum content and thus unavailable as a substrate. Instead, the antimonides serve as a buffer layer material that is used to change the lattice constant when growing on highly mismatched GaAs substrates. In our crystallographic study, we explore the transition during growth from GaAs to GaSb as a way of providing a suitable platform for InAs epitaxy while preserving the ability to isolate the transport properties of the InAs layer.

The lattice parameter mismatch of nearly 8% between GaSb and GaAs is high enough to cause severe material consequences due to the presence of strain and the resulting dislocations that eventually form. In agreement with previous literature, we find that crystal dislocations in GaSb form in a regular array of 90 degree misfits, on top of which the growth is fully relaxed of strain and relatively free of defects. The growth follows a complex Stranski-Krastanov island growth sequence. Probing the formation of the misfit dislocations is the center of our materials study. We report on growth techniques for antimonides, including buffer structures, interface control, superlattices, and the use of a cracked antimony source. Using RHEED, AFM, and TEM/STEM, we study the structure of GaSb misfit dislocations and chronologically probe the growth process in order to describe in time-resolved detail how
the crystal relaxation occurs. We present the first real-time data showing that the formation of Stranski-Krastanov islands and the lattice relaxation occur simultaneously. AFM and TEM data show that the 2D surface underlying the islands is complex and not described by a simple wetting layer model.

1.2 InAs surface quantum well transport

The growth of the very highest quality 2D quantum wells requires careful layer structure engineering in order to protect the well from sources of scattering, which reduce carrier mobility, and unintentional dopants. For instance, modulation doping, which separates donor atoms from the quantum wells themselves, is a crucial technique employed in growing the highest mobility AlGaAs-GaAs wells[5]. InAs based wells are usually constructed with strong confining layers on either side of the InAs, often AlSb, which protect the carriers in the well[6].

However, buried InAs wells are less optimal for applications involving the superconducting proximity effect. Recently, experiments have been conducted demonstrating the ability to induce superconductivity into InAs/GaSb wells as part of the study of the quantum spin Hall effect[7]. The success of such experiments depends on precise growth architecture in order for the superconductivity to reach the well, requiring pristine, highly transparent interfaces between all the materials involved. Our experiment, on the other hand, probes the transport of a quantum well confined to the surface layer of an InAs film, where superconductivity is induced by an overlaying Nb film. Such a geometry results in strong superconducting order in the InAs due to its physical proximity to the superconducting layer and the very transparent contact between the two films.
1.2.1 Proximity effect

The study of superconductivity has continued with unabated pace since its discovery in 1911 by Kammerlingh Onnes. It is one of the most salient of macroscopic quantum phenomena to be experimentally accessed. The introduction of the phenomenological Ginzburg-Landau theory, along with the microscopic Bardeen-Cooper-Schrieffer (BCS) theory, elucidated the phenomenon of the condensed ground state and Cooper pairing, as well as the fundamental nature of superconducting materials. High temperature and other exotic forms of superconductivity have proven a fertile ground for the study of highly interacting systems. This thesis is motivated by the possibility of inducing superconductivity into the surface quantum well of an InAs layer, which is expected to result in topological superconductivity and provide an avenue for probing Majorana fermions. The surface quantum well introduces experimental advantages regarding the proximity effect, but also causes difficulties regarding transport properties.

Many theoretical and experimental studies have been performed to study the proximity effect, where the pair correlations involved in superconductivity bleed into adjacent, normally non-superconducting, materials. The proximity effect is mediated by Andreev reflection, the process by which electrons can scatter between the normal and superconducting materials, conserving energy and momentum during the exchange and extending pair correlations beyond the normal interface. The process is highly dependent on the transparency of the contact, a materials problem studied previously by this group in the work by Vissers[8],[9]. In his experiment he studied the contact between InAs and Nb, a system which forms no Schottky barrier between the materials. By depositing the junction entirely in vacuum, the experiment achieved a transparency of $|T|^2 = 0.7$, near the theoretical maximum. The resulting method for growing highly transparent in situ InAs-Nb junctions enabled an experiment where superconductivity was proximity-induced into the edge channel of a quantum Hall state in a 2-dimensional semiconductor quantum well[10].
The proximity effect is essential in the currently flourishing field of topological quantum computation. In condensed matter, the excitations termed Majorana fermions are actually non-Abelian anyons which have unusual exchange statistics. By performing exchange operations, known as braiding, it is possible to store and manipulate quantum information that is encoded in these states. A nonlocal implementation of non-Abelian states would be immune to local sources of decoherence, making them appealing for fault-tolerant quantum information storage and calculations. The topological superconductivity in which these states manifest relies on the proximity effect for its realization.

As the field of topological quantum computation is built on very exotic physics that is difficult or impossible to find in a fundamental excitation, theoretical and experimental work has focused on the construction of complex heterostructures that manifest the unusual band structures. Specifically, inducing superconductivity into strongly spin-orbit coupled materials can allow for a topologically nontrivial regime wherein states are protected from certain kinds of noise. Understanding the nature of the proximity effect is essential to making such heterostructures work.

1.2.2 Josephson junction arrays

Josephson junction arrays are an important device configuration in the study of the superconducting proximity effect. A single Josephson junction consists of two pieces of superconductor separated by a weak link, a region that may be affected by the proximity effect. The weak link may be composed of a normal metal, semiconductor, topological insulator, or even a constriction in the superconducting material. A Josephson array is made up of many superconducting islands arranged in a grid on top of a non-superconducting, or normal, material, which provides many weak links for the array of junctions. If the resistance of the array is measured as a function of temperature, the curve shows transitions that correspond to various regimes of superconductivity in the sample. First there is evidence of individual islands becoming superconducting, followed by the beginnings of the proximity effect in
the semiconductor regions between them. Finally, the entire array becomes phase coherent, a phenomenon called the phase stiffening transition, which is similar to the Berezinskii-Kosterlitz-Thouless (BKT) transition seen in 2D superconducting films. The presence of superconducting vortices is the reason a finite resistance manifests in the superconductor above the transition. The study of vortex behavior connects this proximity study back to the Majorana fermion search. The details of these transitions provide insight into how the proximity effect functions inside the chosen normal material. In our lab, island arrays on bismuth selenide were studied by Mulcahy[11], who observed fully coherent transport in an array of niobium below the BKT transition, and fabrication processes are being developed for creating island devices on InAs as well.

In addition to quantifying the proximity effect, Josephson junction arrays should themselves be an avenue for the observation of Majorana physics. Kitaev noted that the presence of Majorana states should alter the periodicity in the Shapiro steps observed from the AC Josephson effect for a single junction. It will be fruitful to measure the current-voltage characteristics, current-phase relationship, and magnetoresistance of the devices carefully for deviations from the behavior of typical SNS junctions.

1.3 Thesis outline

The topics discussed in this thesis are eventually motivated by two of the core building blocks of the solid-state implementation of Majorana states. First, a strongly spin-orbit coupled material is required; we choose to study the compound III-V semiconductor InAs. The semiconductor must be of high quality and free of defects which would hinder the quantum behavior of its transport properties. Here we study the the crystal structure of the antimonide buffer layers on which MBE InAs is grown, exploring the nature of dislocation formation in GaSb. Next, the proximity effect must be able to induce a robust superconductive order into the InAs. InAs is also a material of interest for this purpose due to its band
structure. An InAs layer in contact with a metal tends to form a surface accumulation layer of electrons near the interface, where it is easily influenced by superconducting ordering. We characterize these surface wells, with the eventual goal of probing the Berezinskii-Kosterlitz-Thouless transition, a macroscopic quantum phase transition with topological properties that depends strongly on the proximity effect. Our study addresses these topics in order to pave the way for future Majorana devices based on thin films.
Chapter 2

Background

This chapter details the background behind the main thrusts of this thesis: dislocation formation in GaSb grown on lattice-mismatched GaAs substrates, transport in InAs surface quantum wells and the vortex dynamics underlying the superconductor-insulator transition in Josephson junction arrays, and Majorana fermions as manifested in topological superconductors. This material informs the design of our experiment by influencing the choice of materials for growth, as well as device architecture for transport measurements.

2.1 Dislocations in GaSb on GaAs

2.1.1 Stranski-Krastanov growth

The transition from GaSb to GaAs has long been thought to follow the Stranski-Krastanov growth mode, where after a certain thickness is reached, the growth transitions from a stable 2-dimensional pseudomorphic wetting layer to 3D island growth. The Matthews-Blakeslee critical thickness model describes how thick the strained 2D film can grow before the growth transitions to 3D[12]. Following the work of Frank and van der Merwe[13], they calculated the energy balance between crystal strain and misfit dislocation formation, finding that forming dislocations can be energetically favorable over strain alone when the film is sufficiently thick. Simulations of Stranski-Krastanov growth suggest that the 2D to 3D transition is an energetic one, where the surface energy (especially the contribution due to entropy) keeps the growth of the wetting layer stable until the critical thickness is
reached. Then, the strain energy in the volume of the crystal becomes enough to break the surface of the wetting layer[1]. In real materials, the concept of a critical thickness is often an oversimplified description, but it provides a convenient starting point for modeling more complex growth behavior.

Experiments have shown evidence agreeing that the early GaSb growth does form into island shapes with low coverage of the substrate surface, but which widen and eventually coalesce into a 2-dimensional film with continued growth[14][15][16]. It is suggested that the very first layers of GaSb likely form a wetting layer strained to the GaAs surface, after which the crystal relaxes under the strain of the lattice mismatch[17]. In addition to being important crystallographic features, the Stranski-Krastanov 3D islands are interesting in themselves as spontaneously formed quantum dots.

The lattice transition between GaAs and GaSb is interesting due to its practical usefulness for buffer layers, but also has some unusual features worth studying. In 2006 Huang et al. showed that it is possible to grow a relaxed bulk GaSb layer on GaAs after only 3 ML of growth, in spite of the 7.78% lattice mismatch, and that the strain is relieved by a highly periodic array of misfit dislocations. This result narrows down the critical time that needs to be studied to the first few ML of growth, but no further. Highly time-resolved information about those first 3 ML, however, is missing from the literature. Furthermore, under the right growth conditions, the misfits are 90 degree dislocations that do not propagate along the growth direction into the film, which means that the subsequent GaSb growth is relaxed and free of the threading dislocations that usually plague mismatched epitaxy[16]. It is this unusually quick relaxation that makes antimonides so appealing as preparatory buffer layers for InAs. High resolution cross sectional TEM shows the presence of an extra monolayer of (110) GaAs every 14 ML, which correspond to 13 ML of GaSb. A 90 degree misfit is seen every 56 Å at the interface.

A number of MBE studies of the GaSb/GaAs system have been performed over the last two decades. The existing body of literature spans many different purposes and describes
film growths performed at a wide range of growth parameters. The main techniques used to characterize the incipient growth of GaSb include RHEED, AFM, TEM/STEM, and XRD. While many experiments describe the same qualitative growth chronology, they tend to differ on certain small but crucial details. For example, the experiments took place at a wide range of substrate temperatures, a parameter that can have observable consequences for the growth[16]. Interfacial shutter sequences, shown to greatly impact sample mobility, also vary from experiment to experiment or are not described at all. Because essential features of the transition from GaAs to GaSb happen very quickly (within the first few monolayers of GaSb growth), even detailed descriptions of the growth timeline tend to have a relatively course time resolution. There have not been studies with high enough time resolution to track the growth mode together with the lattice parameter. The occurrence of a regular misfit dislocation lattice is universally seen, but how this is obtained remains unanswered. Our study is an attempt for a finer-grained chronological study, building on the literature discussed next.

In the 1996 paper from Thibato[18], an attempt is made at a time-resolved description of GaSb growth. Four distinct phases to the growth, performed at 490 C, are described. They claim the first two ML are 2-dimensional, taking the form of “platelet-like” features around 10 nm in diameter. Between these platelets are gaps 1 ML in depth. The second ML then grows atop these platelets, leaving the gaps. The third ML is when the quantum dots are formed with an area density of around 100 per square micron, which are coherently strained to the platelets and free of dislocations according to TEM. STM revealed that once the dots formed, the 2D “wetting layer” remained beneath them, meaning the dots were not formed by a crumpling of the 2D layers lying below them. Above 3 ML, the film was relaxed and contained dislocations as shown by TEM. The dots appeared to merge as they grew thicker, but growth remained 3 dimensional until around 100 nm of material had been grown, when the growth became flat again, nucleating in a spiral fashion around threading dislocations. At 5 ML, the threading dislocation density was 50 per square micron, which was reduced to
In papers from 1996 and 1997, the same group studied the self assembly of GaSb quantum dots and the “wetting layers” that precede their formation[15][17]. Their STM measurements showed that GaSb at first formed platelet-like 2D island structures on a highly anisotropic ribbon-like wetting layer, each ribbon separated by around 50 Å. They observed the onset of 3D island formation around a nominal GaSb thickness of 2.5 ML, but noted that this thickness could vary depending on growth conditions. The RHEED signature of 3D growth in the form of transition spots had first appeared around 1 ML, coexisting at first with the streaky pattern denoting the smooth GaAs substrate. By 3.5 ML, the transmission spots had strengthened and moved closer to each other, indicating the increased bond length. However, TEM measurements showed the 3D dots to still be coherently strained at this thickness. This means that crystal dislocations had not yet formed and performed their function of relaxing the lattice strain, in contrast to a large number of subsequent results.

The important feature that distinguishes the above growths from ours is their use of the migration-enhanced epitaxy technique (MEE), as opposed to the more typically employed codeposition method. MEE involves shutter sequencing to rapidly alternate between short depositions of Ga and Sb, and shutting off the flux of each when the other material is being deposited. Thermodynamically, this is a highly non-equilibrium situation, where the kinetic conditions are constantly changing, and the surface keeps switching between group III and group V stabilized conditions. In contrast, codeposition involves minimal kinetic control besides the beginning of growth. In our experiment, the underlying physical consequences of growing GaSb starting on an Sb-stabilized surface are able to be studied as a natural evolution under thermodynamic conditions. Our result is also much higher in time resolution, showing that the 2D to 3D transition happens after less than a monolayer of growth.

Bennett et al. claimed a Stranski-Krastanov critical thickness of 2.5 ML, which falls in the middle of a wide range of experimentally determined and theoretically predicted figures[17]. Müller-Kirsch, in a study of films grown by MOCVD, report a critical thick-
ness of around 4 ML. In their theory paper, Eisele and Dähne reported experimental and theoretical determinations of critical thickness to be between 0.4 and 4 ML, also observing that further study is necessary to forestall the “major hassle” that would result from an “uncontrollably varying” critical thickness[19]. Their calculation involved finding the strain fields for infinite 2D films, and gave a result of 1.2 ML for the critical thickness, provided the terminating As on the final GaAs surface has been replaced by Sb atoms in some way. By a static calculation that only involves the relevant forces, their method attempted to bypass thermodynamic considerations and dependence of the critical thickness on growth parameters. This leaves unresolved the important question of whether the lattice transition is dominated by thermodynamic or kinetic effects, which would greatly affect the result[19].

2.1.2 Interfacial misfit dislocations

The features of the misfit dislocations at the interface of GaSb and GaAs appear consistent when microscopically probed in many different experiments. Cross-sectional TEM and STEM are the preferred methods for visualizing the dislocations. As discussed in the 2006 paper by Huang[16], misfits appear in a regular array, with a periodicity equal to 56 Å in two dimensions. The spacing originates from the size of the mismatch between GaSb and GaAs - 14 lattice sites in GaAs correspond to 13 in GaSb, and one dislocation forms to accommodate one extra GaAs lattice site. Burger’s vector analysis of atomic-resolution STEM reveal that these dislocations are 90 degree misfits, meaning the dislocation does not continue to propagate upward in the growth direction as the crystal forms. Moiré fringes seen in plan-view TEM show that the dislocation lattice is 2-dimensional and extremely regular[20]. Huang[16] also shows that the formation of 90 degree misfits is growth temperature dependent. When growth at 560 C, an atypically high growth temperature for GaSb, the 90 degree misfits are replaced with 60 degree misfits, which do lead to dislocation threading. When the growth temperature is 520 C, the misfits are all of the 90 degree type, and the density of threading dislocations is minimized. Any remaining threading dislocations are thought to form due to
island coalescence, should the misfit lattices of different islands form misaligned relative to one another.

2.2 Transport in InAs surface quantum wells

2.2.1 InAs surface accumulation layer

Exposed surfaces of InAs display the unusual property of a charge accumulation layer, which has consequences for any interface made with another material[21]. This is due to the pinning of the surface Fermi level above the minimum of the conduction band, which leads to band bending and the formation of a potential well. The area density of accumulated electrons is around $10^{12}$ per square centimeter, and has been shown to be robust against a variety of surface treatments, bulk doping levels, and growth parameters[22].

However, complex structures based on InAs tend not to make use of its surface properties. The highest mobility InAs quantum wells are made by burying InAs between thick layers of AlSb, whose wide gap provide excellent confinement for the charges in the well. While these wells are of pristine quality, burying them deep in the layer structure makes them more difficult to interact with, such as in proximity effect experiments, where physical distance
from the superconductor has the effect of weakening the pairing strength. Choosing to study the surface wells in InAs is likely to trade some of the carrier mobility for a stronger proximity effect, but is still a worthwhile avenue for investigation.

2.2.2 Proximity effect

When a superconductor is placed in contact with a normal material, its characteristic pairing interaction can diffuse the non-superconducting region. By the process of Andreev reflection, Cooper pairs are able to scatter into the normal material at the interface, creating quasi-particles that can diffuse a finite distance before the pairing is broken. The length scale on which this happens is called the normal metal “coherence length” \( \xi_N = \sqrt{\hbar D/\pi k_B T} \), where \( D = v_F l/3 \) is the diffusion constant of the metal[24]. The Fermi velocity is denoted by \( v_F \) and the mean free path by \( l \). This figure sets the length scale of devices that may be made using proximity-induced superconductivity, such as Josephson junctions. The presence of superconductive correlations in the metal causes it to exhibit some of the characteristics of superconductivity, like dissipationless current flow.

A normal material can be used as a weak link between two superconducting contacts to form a Josephson junction. If the separation between the contacts is not much larger than \( \xi_N \), the phases of the two contacts will be correlated and the Josephson relations for the supercurrent and voltage-phase relationship will manifest.

\[
I_s = I_c \sin \Delta \phi \quad \quad \quad \quad \quad V = \frac{\hbar}{2e} \frac{d\phi}{dt} \tag{2.1}
\]

\( \Delta \phi \) is the difference in phase between the two contacts, and can be used to find the Josephson coupling energy as a function of phase:

\[
E_c = -\frac{I_c \Phi_0}{2\pi} \cos \Delta \phi \quad \tag{2.2}
\]
$\Phi_0$ is the magnetic flux quantum $\hbar c/2e$[25].

### 2.2.3 BKT transition in Josephson junction arrays

A system in which both the proximity effect and topological effects play a substantial role is that of the Josephson junction array. This geometry consists of a large but finite array of superconducting islands spaced a regular distance apart on top of a thin film layer of normal material. In the infinite limit of the array size, this system maps onto the thin superconducting film model studied by Berezinskii, Kosterlitz, and Thouless, which exhibits finite resistance at finite current even in the fully superconducting state. This is due to vortex dynamics that emerge at finite temperatures. For a finite Josephson array, the vortex behavior combines with the proximity effect to produce observable transitions in the resistivity of the array as a function of temperature.

The Berezinskii-Kosterlitz-Thouless (BKT) model is an elegant explanation for the mystifying observation of finite resistances in certain fully superconductive systems. These include very narrow wires and thin films, in which the energy to create a vortex in the superconductor is very small. In the presence of thermal fluctuations when above a certain temperature, the free energy can be lowered by introducing a vortex, a normal region accompanied by an integer multiple of $2\pi$ winding in phase. The change in phase around the vortex must be a multiple of $2\pi$ so that the order parameter stays single-valued at each position. The net phase winding about the whole sample, or the vorticity, is a conserved topological invariant, protected by the superfluid angular momentum. When current is applied, vortices experience a Lorentz force and flow from one boundary of the sample to the other. At one edge, they disappear, while at the other, they form at an equal rate in order to preserve the vorticity. As the core of a vortex is made up of normal (unpaired, non-superconducting) electrons, the energy required to break pairs as vortices cross and leave the sample is lost. This is an avenue for dissipation otherwise not found in a superconductor, and a lossy current results in finite resistance, or finite voltage to accompany current flow.
The BKT transition occurs at a temperature at which vortices go from being free to move and flow, and thus dissipate energy, to being bound in vortex-antivortex pairs, on which the Lorentz forces cancel, preventing dissipation. This means there should be a transition from finite to zero resistance as the temperature drops below the BKT critical temperature. In a Josephson junction array, this corresponds to macroscopic phase stiffening, where the phase as a function of position across the sample is constant in time. Phase stiffening occurs when superconductivity, resulting from the proximity effect in the weak links connecting the islands, becomes strong enough to lock the phase between adjacent islands. Thus, observation of the BKT transition in the resistance of the island array is intimately connected to the nature of the proximity effect.

2.3 Majorana fermions

2.3.1 Majorana, Dirac, and Kitaev

Ettore Majorana first proposed his namesake fermion in 1937 in the context of particle physics. When the relativistic Dirac equation was first found, its negative energy eigenfunctions caused great consternation among physicists. Prior to the discovery of antimatter, there was no reason to believe the negative energy solutions to physically exist, but no mathematical reason to discard them. To reconcile his discomfort with the result, Majorana recast the Dirac equation so that it provided only real fermionic solutions. Eliminating negative-energy solutions also removed the distinction between particle and antiparticle. However, eventually further progress in high energy physics demonstrated the existence of antimatter, vindicating the Dirac equation’s original interpretation. No elementary particle was found to obey the physics laid out by Majorana, and his calculation remained a mathematical curiosity with no real-world application for many years[26].

In homage to Majorana, the condensed matter community uses the term “Majorana fermion” to describe emergent self-conjugate states, whose creation and annihilation opera-
tors are equal to one another. These are not elementary particles, but rather quasiparticles, or collective excitations of a complicated underlying system. Majorana quasiparticles have unique properties that the elementary Majorana fermion does not share, since the quasiparticles arise as zero-energy modes in certain systems with particle-hole symmetry. In fact, in condensed matter they are not fermionic at all, but obey non-Abelian exchange statistics, the foundation for fault-tolerant topological quantum computation schemes[27][28]. These systems are completely distinct from Majorana’s treatment of the Dirac equation, only sharing the property of self-conjugation. To avoid confusion, in this thesis terms like Majorana modes, excitations, or quasiparticles will be preferred over “Majorana fermions” when referring to condensed matter applications.

A number of systems have been suggested to host Majorana zero modes, among them fractional quantum Hall states and cold fermion systems[29]. Superconducting systems are prominent among those in which Majorana modes can arise, due to their inherent particle-hole symmetry near the Fermi surface. The excited states of a superconductor are given by the Bogoliubov operators, which consist of superpositions of electron and hole states. For the standard s-wave Bardeen-Cooper-Schrieffer (BCS) Hamiltonian, they take the following forms:

\[ b_0^* = u^* c_{\uparrow}^* - v^* c_{\downarrow} \]  
\[ b_1^* = u^* c_{\downarrow}^* + v^* c_{\uparrow} \]  

Here \( c_\sigma \) and \( c_\sigma^* \) annihilate and create an electron of spin \( \sigma \), respectively, and \( u \) and \( v \) are the probability amplitudes for a state being unoccupied and occupied, respectively[25]. Such states would be a natural place to search for excitations that might obey the Majorana condition, were it not for the spin dependence that prevents them from being self-conjugate. Therefore, initial theoretical efforts focused on spinless quasiparticles, which pair two particles of the same spin \( \sigma \) with equal weight. By inspection, such an quasiparticle will be equal
Figure 2.2: Schematic of the Kitaev chain. The upper row depicts the trivial configuration, where Majoranas belonging to the same site are paired. The lower row shows the nontrivial state, with pairing shifted over by one Majorana site from the trivial state, leaving unpaired Majoranas at either end. Reprinted by permission from Springer Customer Service Centre GmbH: Springer Nature Nanotechnology[28], © 2013.

to be its own antiparticle[30]. We label the operator $\gamma$ to indicate a Majorana state.

$$\gamma = uc_\sigma^* + u^*c_\sigma$$  \hspace{1cm} (2.5)

Alexei Kitaev’s seminal work provided the foundation for how Majorana states arising in superconductors can be used to implement fault-tolerant quantum information storage[31]. As detailed in his 2001 paper, his Majorana nanowire scheme provides protection against qubit dephasing, one of the prominent sources of error in these quantum systems.

Qualitatively, the Kitaev chain “toy model” consists of a row of superconducting (non-Majorana) fermionic electron sites, numbered by a position index $j$. The Hamiltonian for this system, in terms of Dirac fermion operators $c$ and $c^*$, includes the nearest-neighbor hopping amplitude $w$, the superconducting gap $\Delta$ and phase $\theta$, and the chemical potential $\mu$.

$$H = \sum_j \left[ -w(c_j^*c_{j+1} + c_{j+1}^*c_j) - \mu(c_j^*c_j - \frac{1}{2}) + \Delta|e^{\imath\theta}c_jc_{j+1} + |\Delta|e^{-\imath\theta}c_j^*c_{j+1} \right]$$  \hspace{1cm} (2.6)

Here spin is simply not included as a degree of freedom, the most straightforward way to create operators taking the form of equation 2.5.
Next, Kitaev defined Majorana operators as follows:

\[
\begin{align*}
\gamma_{2j-1} &= c_j + c_j^* \\
\gamma_{2j} &= \frac{c_j - c_j^*}{i} \\
&j = 1, \ldots, N
\end{align*}
\] (2.7)

Effectively, each fermionic site of the chain has been divided into two Majorana contributions. In most situations, this is a merely mathematical equivalence that has no effect on the physics of the system. Each Dirac fermion can be considered as the superposition of two Majoranas, but because the two are localized very closely to one another they cannot be individually probed. However, should a pair be somehow spatially separated, preventing their wavefunctions from overlapping, the fermionic state the Majorana pair encodes becomes delocalized. This has the effect of protecting it from local sources of decoherence that only affect one of the Majorana states[32][30]. If this immunity to decoherence can be employed in a qubit, a significant challenge of quantum computing will be mitigated.

The Hamiltonian is then written in terms of the Majorana operators:

\[
H = \frac{i}{2} \sum_j \left[ -\mu \gamma_{2j-1} \gamma_{2j} + (w + |\Delta|)\gamma_{2j} \gamma_{2j+1} + (-w + |\Delta|)\gamma_{2j-1} \gamma_{2j+2} \right]
\] (2.8)

The parameters \(\mu\), \(w\), and \(|\Delta|\) form a phase space in which to explore the behavior of the system. In the limit \(|\Delta| = w = 0\) and \(\mu < 0\), the remaining terms in the Hamiltonian pair Majoranas from the same Dirac site, forming a trivial ground state with no unpaired Majorana states on the ends. However, in the limit \(|\Delta| = w > 0\), \(\mu = 0\), the Hamiltonian pairs each Majorana with another Majorana lying on the adjacent Dirac site. Kitaev explained that these two situations, illustrated in figure 2.2, are the model’s two universality classes, and are two topologically distinct situations with the same bulk properties but distinct boundary characteristics. The dangling end Majorana fermions are the “unpaired” fermions that may be individually probed.

Kitaev’s also showed that a topological phase transition separates the two pairing states
in the wire. Since the bulk quasiparticle gap must close to undergo the transition, it offers the unpaired Majorana states a certain “topological protection,” and thereby the robustness that may allow for developments in quantum computing with MFs. Kitaev suggested a measurement setup of a Josephson junction in the form of a “quantum wire bridge,” an important construction to keep in mind[31].

Once Kitaev demonstrated mathematically that the decoherence protection is topological in nature, he then proposed to use unpaired Majorana modes as a fault-tolerant method for storing quantum information. As mitigating decoherence in qubits is the primary engineering challenge in quantum computing, using Majorana states is an attractive path toward increased coherence times in qubits, motivating our present study.

### 2.3.2 Emergent Majorana states

While Majorana fermions have not been found as fundamental particles in nature, exciting possibilities have emerged in condensed matter, where the interactions of different materials can be used to realize complex band structures that would not occur naturally in a known system. Specifically, the goal is to construct a superconducting system whose excitations can obey the Majorana criterion. Read and Green constructed an effective mean-field Hamiltonian having such excitations under certain conditions[33].

\[
H = \sum_{k} \left[ (\varepsilon_{k} - \mu) c_{k}^{\dagger} c_{k} + \frac{1}{2} \left( \Delta_{k}^{*} c_{-k}^{\dagger} c_{k} + \Delta_{k} c_{k}^{\dagger} c_{-k}^{\dagger} \right) \right] 
\]  

Here \( \varepsilon_{F} \) is the single-particle kinetic energy and \( \Delta_{k} \) is the superconducting gap function, and spin degrees of freedom are again ignored. Next, the gap function was written as

\[
\Delta_{k} \equiv \Delta_{0}(k_{x} - ik_{y})
\]  

20
where $\Delta_0$ is a constant. The Hamiltonian then describes a superconductor in 2 dimensions with only one spin degree of freedom, and exhibiting chiral $p$-wave pairing in the form of the gap function. The key result was that at vortices and sample edges, where the order parameter $\Delta_0$ vanishes, the lowest energy solution of the Bogoliubov-deGennes equation $H_{2D}^p \Psi(r) = E \Psi(r)$ is non-degenerate due to the absence of spin degrees of freedom, and has energy $E = 0$. The zero-energy condition results from particle-hole symmetry, which in terms of second quantized creation and annihilation operators appears as

$$c_{-E}^* = c_{-E}$$  \hspace{1cm} (2.11)

Therefore, the second quantized operator creating the Bogoliubov quasiparticles is Hermitian at $E = 0$, confirming the Majorana criterion, as we are left with $c^* = c$. Without spin as an additional degree of freedom, the creation and annihilation operators for zero-energy states at order parameter defects are identical and thus particle and antiparticle are no longer distinct\cite{29}\cite{33}. If this Hamiltonian could be realized in a physical system, this would provide an avenue for probing Majorana excitations.

### 2.3.3 Proposed Majorana heterostructures

Unfortunately for experimentalists hoping to observe actual physical signatures resulting from the presence of Majorana modes, the spinless $p$-wave superconductivity described by Read and Green, while theoretically straightforward, rarely occurs spontaneously, if at all. The most promising material, strontium ruthenate ($\text{Sr}_2\text{RuO}_4$), is still being studied for conclusive proof of the presence of $p$-wave superconductivity. Its exotic pairing has also proven fragile and easily ruined by the presence of disorder in the system\cite{34}. For this reason, theorists have turned their attention toward the creation of systems that can be used to simulate spinless $p$-wave superconductivity as the effective behavior of an underlying system built up out of more experimentally accessible components.
Fu and Kane first proposed a hybrid system coupling ordinary \( s \)-wave superconductivity by physical proximity to the surface states of a strong 3-dimensional topological insulator (TI)[35]. Working out the physics of the proximity-induced surface states, they obtained the a Hamiltonian that is formally equivalent to the Read and Green \( p \)-wave system, and hosts similar Majorana bound states at vortices and edges. This proposal proved a breakthrough in the condensed matter field, as unlike the exotic \( p \)-wave form of superconductivity, \( s \)-wave superconductors are ubiquitous and extensively used experimentally. Other \( s \)-wave based proposals were soon to follow, with many experimental efforts focused on 1-dimensional nanowire devices.

The semiconductor heterostructure approach underlying the Majorana nanowires discussed next, pioneered by Sau[3] and expanded by Lutchyn[36] and Oreg[37], uses proximity-induced \( s \)-wave superconductivity, like Fu and Kane’s TI-based model. In this setup, a semiconductor exhibiting strong spin-orbit coupling (such as InAs or InSb) is used so that the spin up and spin down bands become separated. With the addition of a magnetic field to cause Zeeman splitting, along with induced superconductivity, the system will enter into a state that is effectively a spinless \( p \)-wave superconductor. Lutchyn \textit{et al.} proposed the realization of the heterostructure system as a layered structure consisting of a magnetic insulator lying underneath a semiconductor, which provides the Zeeman field, while a layer of superconductor is deposited on top. Alternatively, the magnetic field can be applied externally, which has the advantage of being tunable but is not inherent to the heterostructure.

The semiconductor-based proposals provide us with a checklist of ingredients that must be present to realize a Majorana-hosting topological phase in a quantum wire. First, superconductivity must be present to enforce the requisite particle-hole symmetry. Two effects, spin-orbit coupling and the Zeeman effect, combine under certain conditions to produce the necessary spin-polarization. We will now examine the underlying theory in further detail.

Solving the Schrödinger equation for one occupied subband of free electrons confined in 1D results in the familiar parabolic dispersion relation \( E_k = \hbar^2 k^2/2m \). The energy
spectrum is doubly degenerate due to the presence of spin up and spin down components. The introduction of Rashba spin-orbit coupling adds another term to the Hamiltonian, which takes the form $H_R = \alpha k_x \sigma_y$. The resulting dispersion relations are

$$E_k^\pm = \frac{\hbar^2}{2m} (k \pm k_{SO})^2 - \frac{\hbar^2 k_{SO}^2}{2m}$$

(2.12)

where $k_{SO} = m \alpha / \hbar^2$, and $E^+$ and $E^-$ are the energies for spin up and spin down electrons, respectively[38]. The parabolic form of the bands is preserved, but the Rashba coupling serves to move the parabolas down and to the left (for spin up) and right (for spin down), as seen in part (a) of figure 2.3.

![Figure 2.3: Band splitting due to Rashba and Zeeman effects. (a) shifted parabolic bands for spin up (left) and spin down (right) due to the Rashba effect in the absence of a magnetic field; (b) opening of a Zeeman gap at small field; (c) Zeeman gap at large field, with a wider effectively spinless regime; (d) same as (c) but with proximity-induced superconducting gap added. © IOP Publishing. Reproduced with permission. All rights reserved.[30].](image)

The addition of an external magnetic field perpendicular to the spin-orbit coupling introduces a Zeeman term into the Hamiltonian, $H_Z = g \mu_B B_x / 2$, where $\mu_B$ is the Bohr magneton and $g$ is characteristic of the material. This has the effect of hybridizing the spin bands around $k = 0$, lifting the degeneracy there by opening up a gap. If the chemical
potential happens to lie inside the Zeeman gap, the system has effectively only one spin band. This is shown in parts (b) and (c) of figure 2.3. As long as the chemical potential can be tuned (by gating or otherwise) into the Zeeman gap, the spinlessness requirement is met. As both of the suggested high spin-orbit semiconductors also have large $g$-factors, ($g \approx 15$ for InAs; $g \approx 50$ for InSb), the Zeeman gap can be made large without applying too high of a magnetic field.

Finally, superconductivity must be introduced into the system, opening up yet another gap in the system, seen in part (d) of figure 2.3. The proximity effect, which introduces Cooper pairing into a normally non-superconducting material, has been shown to exist in a number of target materials. Lutchyn et al. demonstrated that when combined with the effective spinlessness created above, the proximity induced superconductivity takes on the unusual chiral $p$-wave form [36]. The resulting single-particle Hamiltonian is isomorphic to the Kitaev chain and manifests Majorana modes at its endpoints.

The Majorana checklist of superconductivity, spin-orbit coupling, and Zeeman effect are what determine the material parameters in our experiment. We focus on the the surface quantum wells of the semiconductor InAs, when in proximity with Nb, a strongly spin-orbit coupled system that has been shown to form highly transparent interfaces. Pair correlations formed by the proximity effect in InAs have also been studied.

### 2.3.4 Experimental background: Majorana nanowires

The prominent experimental results on hybrid nanowire devices have employed epitaxially grown wires, which are grown vertically from seed crystals by various deposition methods. After growth they must be made to lie horizontally on a substrate and electrically contacted ex situ. The crystalline quality of grown nanowires is excellent, providing high electron mobilities and few defects, and can even be grown inside in situ shells of aluminum for highly transparent interfaces [39].

The 2012 experiment by the Kouwenhoven group at Delft [4] possibly demonstrated the
Figure 2.4: (a) Device schematic from the 2012 Kouwenhoven experiment. The InSb nanowire is in blue, the superconducting lead in yellow, the normal metal lead in silver, and gate electrodes are black lines. The magnetic field direction is indicated by red arrows. Reprinted by permission from Springer Customer Service Centre GmbH: Springer Nature Nanotechnology[28], © 2013. (b) Soft gap and zero bias conductance peak seen in differential conductance peaks at various magnetic fields, from Kouwenhoven experiment. From [4]. Reprinted with permission from AAAS.

The first signal of the presence of Majorana modes in a nanowire. The experiment consisted of InSb wires grown by MOCVD lying atop a series of narrow Au gates. Contact electrodes were defined \textit{ex situ}, so the wire had to be sputter cleaned in argon before deposition to improve the coupling to a superconductor, NbTiN, at one end of the wire and a normal metal, Au, at the other. Measurements of differential conductance, which is proportional to the density of states at a given bias voltage, were taken at various values of bias voltage and applied magnetic field. The $dI/dV$ traces showed signs of a “soft” proximity-induced superconducting gap, indicating that the proximity coupling could be improved. Their key finding was a robust zero-bias conductance peak (ZBCP) when the external magnetic field was within a range of 100-400 mT, which disappeared when the field was applied parallel to the spin-orbit field. Other groups have confirmed the ZBCP result using devices with a wide variety of parameters. Deng[40] used InSb wires similar to those from the Delft experiment, contacted by two superconducting Nb electrodes separated by 110 nm, while a global back gate was used to tune the Fermi level in the nanowire. In addition to the ZBCP, they observed a Josephson supercurrent and multiple Andreev reflection when the wire was
in the trivial state. The van Harlingen group[41] observed similar results in wires made of InAs. They also measured changes in the width of the ZBCP as system parameters were tuned, as well as peak splitting and re-forming, an observation consistent with hybridization of the two end Majoranas.

While consistent with theoretical expectations, a ZBCP result is not considered a “smoking gun” positively indicating the presence of Majorana bound states. Debate continues over whether signatures consistent with Majoranas can be explained away by more trivial factors, including the Kondo effect, Andreev bound states, and weak antilocalization[42][43]. A much more conclusive result was published in 2017 in a study on a very different system, the edge transport in a magnetic topological insulator film stacked beneath a layer of niobium[43]. This experiment measured half-integer conductance quantization, a sign that in the presence of a Majorana mode, current is not only carried by Cooper pairs of electrons.

New developments in processing techniques continue to emerge and improve the prospects for Majorana nanowire experiments. The “soft gap” seen by Kouwenhoven group, a consequence of the inhibited proximity effect due to ex situ superconductor deposition, was mitigated by in 2015 by Chang et al.[39]. This group developed a method to grow Al shells around InAs nanowires in situ, which improved the interface to the point where a full superconducting gap could form. Our approach, focused on 2D film growth, also follows this strategy of in situ superconductor deposition to maximize the strength of the proximity effect.

The device geometry in the nanowire approach is unfortunately limited by the random patterns into which the wires fall. Attempts to manipulate more than one nanowire simultaneously result in low device yield and thus difficulty in scaling up to larger multi-qubit systems. A method of fabricating quantum wire circuits out of thin films by means of lithographic techniques is an important step along the route to large-scale topological quantum computing. For this reason, thin film approaches to device production are indispensable. The fabrication processes required provide significant engineering challenges to overcome,
but if they can be solved the film approach will also be a viable avenue toward the creation of device networks involving many Majorana modes.
Chapter 3
Experimental Methods

3.1 Molecular beam epitaxy

Molecular beam epitaxy (hereafter MBE) is a crystal growth technique employed mainly for its precision. Slow growth rates and a clean ultra-high vacuum environment distinguish it from other growth methods, allowing the precise control of growth conditions required for atomically smooth layer-by-layer growth[44]. By matching exacting conditions on the molecular beam fluxes and substrate temperature, it is possible to reproducibly grow single crystal materials in a variety of architectures, with abrupt variations in composition and contamination free interfaces between materials[45]. MBE is used to produce high mobility 2-dimensional electron gases (2DEGs) in which the quantum Hall effect and conductance quantization can be measured[5]. Quantum dots, wells, and solid-state lasers are grown for optics applications[46].

The following discussion of the growth of III-V semiconductors centers around the most common among these compounds, GaAs. However, the techniques can be applied in a qualitatively similar way to the other semiconductors in the III-V family, with some adjustments to the relevant fluxes and temperatures.

3.1.1 System B

Our III-V MBE growth is conducted in a dedicated ultra-high vacuum chamber ("system B") equipped with source materials, vacuum pumps, and peripheral instrumentation which aids
Figure 3.1: Schematic of our MBE system. Essential components include: residual gas analyzer (RGA), ion gauge, load lock, RHEED system, evaporation sources, and various pumps.
in the growth process. Eight source ports accommodate evaporation cells (mostly Knudsen
cells), thermal evaporation sources loaded with elemental arsenic, gallium, aluminum, and
indium. All eight ports are covered by pneumatically controlled shutters, which can be
activated manually or in timed sequences by a LabVIEW program. A two-stage valved
antimony cracking source from SVTA is also present in one of the source ports. Cracking
sources are often preferred for elements, such as arsenic and antimony, that evaporate in
clusters of multiple atoms, and use a two-stage heater to break the molecules into smaller
and more reactive species. A variety of dopant sources can also be included depending
on the application, such as silicon, tellurium (in the form of GaTe), and manganese. For
III-V growth, the sources are placed in solid form into a crucible made of pyrolitic boron
nitride, a ceramic that does not outgas or decompose except at very high temperatures[47].
Once under vacuum, current is run through the cell’s heating filaments, which wrap around
the crucible and lead to evaporation of the material. The higher the cell temperature, the
greater the flux of atoms that leave the crucible. An ionization gauge placed in the path
of the molecular beams can be used to estimate the beam equivalent pressure in order to
calibrate the flux exiting a cell, although this measurement does not account for differences
in gauge sensitivity to different elements[46]. A substrate placed in the line of sight of the
crucible opening will be struck by most of the oncoming source atoms, as the vacuum in
the chamber leads to a mean free path on the order of kilometers. For the correct choice of
substrate material, temperature, and growth conditions, the necessary chemistry will occur
to form a single-crystal layer on the substrate.

Substrates are loaded into the vacuum system through a secondary chamber called the
load lock, a small volume with its own pumping system that can be isolated from the main
chamber with valves. The load lock can be brought up to atmosphere and pumped down
without affecting the vacuum in the main system. Some mechanism to transfer the substrate
holder from the load lock into the chamber is required, and the substrate manipulator inside
the chamber must be able to position the sample in the orientations required for transfer
and growth. In our system, substrates sit on an elevator inside the load lock, which is lowered into a transfer tube, also kept at ultra-high vacuum. Inside the tube is a system of manually operated forks and pulleys that can pick up the holder and move it to any of the four MBE chambers connected to this tube. From the tube, another set of forks transfers the substrate into the growth chamber itself, where it is attached to the manipulator. The manipulator provides complete 360 degree rotation of the substrate, and its angular position can be raised to place the substrate in the line of sight of the sources. After growth, the manipulator must then be lowered again to transfer the sample out of the chamber. The substrate heater filament, externally powered and controlled in a similar manner to the source cells, lies behind the sample puck, heating it radiatively. Substrate temperature can be monitored by means of a thermocouple in direct contact with the substrate, or with an infrared pyrometer through the chamber windows; in system B the latter method is used.

Reflection high energy electron diffraction, or RHEED, is the main in situ method for monitoring the crystalline quality of the terminating surface of a growing film. Within the RHEED gun, electrons are emitted off of a high-current filament, accelerated by 10 kV, and collimated such that the beam strikes the substrate at a glancing angle. Reflected and diffracted electrons then travel to a phosphor screen across the chamber from the electron source, where diffraction features can be observed and recorded by a camera. Because only the first few layers of the film are being probed, the features of the RHEED pattern are highly sensitive to the morphology of the growth surface, especially its relative flatness. Changes in the RHEED pattern that alert the growth controller to potential problems enable real-time corrections to suboptimal growth conditions. Recorded images of RHEED patterns allow further analysis of the state of the growth at a frame-by-frame resolution.

3.1.2 Ultra-high vacuum

Achieving ultra-high vacuum places certain restrictions on the construction of the system. The load lock enables loading and unloading of samples while protecting the cleanliness of
the growth chamber from exposure to air. All sealing surfaces are stainless steel knife edges pressed into a copper gasket, minimizing leaks to air.

Materials for the in-vacuum components are chosen with their outgassing characteristics in mind, as the wrong materials can both inhibit the vacuum and introduce contaminants into samples\cite{48}. Anything to be introduced into the chamber must undergo degreasing with acetone followed by isopropanol. Metal parts that must be heated to high temperatures, such as heater filaments and the substrate holder, are made of the refractory metal tantalum, which is especially suited to these applications due to its high melting point. Molybdenum can also be used, though not to such a high temperature as tantalum. Electrically insulating parts are, like the source crucibles, made of pyrolitic boron nitride.

The chamber is initially roughed out with a dry mechanical pump such as a diaphragm pump or roots blower, which evacuates the system to below 1 torr, low enough to start a turbomolecular pump which should then bring the chamber to around $1 \times 10^{-5}$ torr. At this point, the high vacuum pumps, a cryopump and an ion pump, can be activated. Leak checking is accomplished with the help of a residual gas analyzer (RGA), which identifies the species present by molecular weight, and a helium bottle to locate leaks which are then tightened.

It is necessary to undergo an outgassing and bake procedure before commencing growth. Outgassing involves heating each source cell above its typical growth temperature for a few hours to ensure that water, solvents, oxides, and other volatile contaminants are removed from the cell components and the source material itself. This is a process that must also be extended to the materials constructing the chamber, as anything that was exposed to room air will have collected an adsorbate layer of mostly water. Bakeout lasts for around 2 weeks, during which levels of the contaminants are monitored using the RGA. When the bake is complete, the partial pressure of each harmful contaminant is below the resolution limit of the RGA, around $10^{-11}$ torr.

A set of cooling shrouds built into the chamber walls can be filled with liquid nitrogen
(LN2) during growth to further reduce the residual gases, especially water and hydrocarbons, and thus the level of background contamination[45]. Finally, a titanium sublimator filament provides additional pumping by depositing a few monolayers of titanium on the chamber walls, increasing adsorption of gas molecules. The base pressure attained in system B with all available pumping systems active is below $10^{-10}$ torr.

### 3.1.3 Growth of GaAs

The following discussion describes the most commonly grown III-V semiconductor, GaAs, but the technique can be generalized to several of the compounds in that category.

Arsenic, when heated by a Knudsen cell in solid form at temperatures and pressures relevant to MBE, sublimates into $\text{As}_4$ molecules, while gallium evaporates from liquid as monomers with unity sticking coefficient[49]. The equation modeling the flux coming out of a Knudsen cell, in units of molecules per square centimeter per second, is

$$F = \frac{P a}{\pi L^2 \sqrt{2 \pi m k_B T}}$$  \hspace{1cm} (3.1)

$P$ is the equilibrium pressure in the cell at temperature $T$, while $a$ is the area of the cell opening, $L$ is the distance from the cell to the substrate, and $m$ is the mass of the evaporated molecules[47]. $P$ is the parameter we pay the most attention to, as it is determined by the vapor pressure of the material being evaporated. Vapor pressure is a strong function of temperature, meaning that incident flux is controlled by changing the cell temperature. A rough estimate of flux can be made using data like the RCA tables, which record vapor pressure as a function of temperature for many materials.

Historically, the GaAs crystal was thought to form by a second order reaction in which two arsenic dimers react with each gallium atom. The efficiency of this reaction is capped at 50%, while excess arsenic atoms will re-evaporate from the substrate above 300 C without being incorporated into the crystal. Subsequently, both As- and Ga-stable growth regimes
were shown to be possible, in which the sticking coefficients can vary from this model[50]. However, the As-terminated regime is chosen for several reasons. First, a shortage of As to react with runs a risk of Ga droplet formation instead of GaAs growth, which is an irreversible degradation in the crystal quality. Excess As also helps to combat the effects of As loss due to desorption by the substrate. Since a higher substrate temperature is preferred in any case to decrease impurities and results in smoother morphology, it is safest to provide an overpressure of As throughout the growth, so that lost As may be replaced. Layer by layer epitaxy tends to happen by a process of island nucleation. First, growth nucleates at a certain spot, and then as the lateral growth rate is much higher than the vertical rate, the islands enlarge and coalesce into a 2D film.

Before growth, a substrate must be selected and prepared with great care to ensure a high-quality and uncontaminated growth. Our substrates are commercially manufactured, epitaxy-ready 2-inch GaAs wafers. They are oriented with the growth surface in the (100) crystal direction and cleaved into four quarters. At minimum, substrates must be solvent cleaned to minimize introduction of contaminants. GaAs wafers may be etched in 10:1:1 H\textsubscript{2}SO\textsubscript{4}:H\textsubscript{2}O\textsubscript{2}:H\textsubscript{2}O for 30 seconds after sonication in trichloroethylene, acetone and isopropanol. GaSb substrates may be etched in HCl. These treatments serve to selectively flatten taller elevations in surface topography without also causing pitting. It is vital that etchant residue be thoroughly rinsed away with deionized water before the substrate is introduced to vacuum, again for reasons of cleanliness. The DI water rinse also assists in forming a protective passivation layer on the surface. Once loaded into the vacuum system, each substrate is heated to a temperature of about 200 C and outgassed overnight in the transfer chamber to eliminate adsorbed water and any volatile contaminants.

Once the substrate is outgassed, it can be loaded into the growth chamber. Liquid nitrogen is introduced into the cryoshrouds to cool the chamber, and the titanium sublimation pump is run for a single 2 minute cycle so that the system reaches its base pressure. Next, the source cells must be heated by ramping up the heater power over the course of about an
hour. Once they have achieved growth temperature, they are allowed to stabilize for another
hour.

![RHEED pattern before and after oxide blowoff.](image)

Figure 3.2: RHEED pattern before and after oxide blowoff. The difference in brightness is clearly detectable when monitoring intensity with a camera, and is used to calibrate the pyrometer for temperature measurement.

The final step before growth can begin is oxide desorption. When exposed to air, or when etched with an oxidizing solution containing H$_2$O$_2$, GaAs, GaSb, and most other III-V compounds form an amorphous oxide layer on the outer surface, which must be removed to expose the crystalline structure underneath. This is accomplished by heating the substrate. In the case of GaAs, the native oxide desorbs around 600 C. This can be seen by observing the RHEED pattern as the heater filament current is increased: at the time of desorption, diffuse reflections morph into the streaks expected for a 2D crystalline surface, indicating the substrate surface is ready for epitaxial growth[44], as seen in figure 3.2. Noting the temperature, as measured with a pyrometer, at which desorption occurs for each substrate allows temperature measurements to be calibrated from growth to growth. The high vapor pressure of arsenic means that above a temperature of around 300 C, a steady flux of arsenic must be supplied to replace that which evaporates out, otherwise gallium droplets will form, ruining the crystalline order[49]. For practical purposes, this means that the arsenic source remains on from the first stage of substrate heating before oxide blowoff, until the substrate heater is turned off after growth. A side effect of the continuous high partial pressures
used is the accumulation of arsenic on the pyrometry window glass, gradually increasing its opacity. This leads to a decrease in the readings given by the pyrometer over time. One can compensate by decreasing the pyrometer’s emissivity setting, nominally $\epsilon = 0.68$ for GaAs, to match the occurrence of oxide blowoff to the same temperature each time.

With oxide blowoff complete, growth can now begin. Since the arsenic overpressure is already present from the oxide blowoff step, opening the gallium shutter will start growth. Under typical growth conditions, the group III elements have a unity sticking coefficient on the substrate, while excess group V molecules will evaporate away. Therefore, the growth rate of stoichiometric GaAs is entirely determined by the gallium flux, so long as sufficient arsenic is provided[44]. The sticking coefficient of arsenic is more complex, determined by whether the evaporated species consist of dimers or tetramers. Evaporated arsenic monomers are not relevant because at the relevant temperatures they tend to reform into dimers[49]. In the absence of an arsenic cracker cell, as is the case in system B, the predominant arsenic species is As$_4$. However, optical evidence suggests that using an As cracker to break the tetramers into dimers can lead to higher quality growth.

GaAs grows epitaxially under a fairly wide range of substrate temperatures and source fluxes. A higher substrate temperature is correlated with decreased impurity levels and smoother growth morphology, up to a limit of around 640$^\circ$C where gallium atoms cease to stick to the surface as readily[44]. However, a high growth temperature also necessitates increases in the arsenic flux in order to maintain the arsenic-stable growth regime, so a balance must be struck. Our GaAs is grown at temperature of 570-590$^\circ$C, with a low carrier concentration of $3.5 \times 10^{15}$ per cubic centimeter and a mobility of $2.29 \times 10^4$ cm$^2$/V·s at 77 K, indicating good system cleanliness.

Techniques can be employed to improve the growth quality even more. Regular interruptions in growth allow extra migration time for atoms that have just reached the surface, which can lead to fewer defects overall but also increases the chance of incorporating contaminants. Strategic insertion of superlattice layers, commonly AlGaAs alternated with GaAs,
are used to trap and bury contaminants that stick to the aluminum content of the alloy. We insert an AlGaAs-GaAs superlattice before the growth of thick GaAs layers in our films.

The GaAs films grown in system B are high-quality, smooth, semi-insulating, and high mobility samples, which have been used in a variety of device applications. The work of Vissers[8][9] and Law[10] focused on studies of the proximity effect, utilizing the quality of the in situ interfaces with niobium. Vissers quantified the high transparency of the superconductor-semiconductor junction using a 3-terminal device, and found that the transparency matches the highest predicted value of 0.7. Law studied the proximity effect induced into the 1-dimensional transport channels of the quantum Hall effect in an AlGaAs-GaAs 2-dimensional quantum well. Her study showed evidence of supercurrent and Andreev reflection, indicating that superconductivity was indeed occurring in the semiconductor channel.

3.1.4 Generalizing GaAs techniques to antimonide growth

Growth of binary antimonide compounds bears qualitative similarities to the well-studied growth of GaAs and other arsenides. In both cases the preferred growth regime is group V-stabilized, occurring under a continuous overpressure of in this case Sb. Both As and Sb evaporate as tetramer molecules, meaning that growth occurs by a higher order process at the substrate surface in order to result in stoichiometric growth. Growth rate is controlled entirely by adjusting the flux of the group III element. The use of RHEED for rate determination and real-time growth monitoring is similar, though with some important differences to be discussed. A significant element of the work done for the growth portion of this thesis was to establish optimal procedures for using a new Sb cracker, purchased from SVT Associates and installed in 2014.

On the other hand, Sb has some bulk properties which As does not share, resulting in adjustments to the procedures used to handle it. Sb atoms have a tendency to cluster together and form aggregates, which has both chamber maintenance consequences and effects on resulting growths[51]. From a practical point of view, the use of Sb in a deposition
chamber means that care must be taken not to allow Sb aggregation to clog openings or short electrical connections. This results in reduced time between vents and decreases machine throughput. It also has the effect of requiring higher Sb fluxes than would actually be needed for optimal growth, to make up for Sb atoms lost on the way to the substrate. Figure 3.3 shows the practical consequences of a few months of such high Sb flux growth.

![Figure 3.3](image.png)

Figure 3.3: Several centimeters of Sb accumulated onto a vacuum part in system B, the thickness of which is indicated by the red arrows. While using a Knudsen cell, a large amount of Sb gathered over a few months on a piece of shielding near the opening of the cell. None of this Sb ever reached the substrate.

The alternative to a regular Knudsen cell is the use of a cracking source for Sb, which helps the growth of antimonides in several ways. A cracker consists of two main zones. The bulk zone works like a Knudsen cell, containing a crucible filled with bulk material and heated to an appropriate temperature for evaporation or sublimation. Before the flux from the bulk zone reaches the substrate, it passes through the second zone, or cracking zone. Heated to a much higher temperature than the bulk zone using an independent filament, the cracking zone provides enough thermal energy to crack Sb tetramers into dimers, and dimers into monomers. At a cracking zone temperature of 900 C, as used for the growths in this experiment, the evaporated species are 90% monomeric Sb with the rest being Sb$_2$;
practically no Sb tetramers are present in the molecular beam[52]. The advantages of dimer and monomer Sb is that the film-forming reactions at the substrate surface are more efficient, unlike the tetramer reaction, which wastes an Sb atom for each one incorporated into the film. Already this reduces the amount of flux needed for an antimonide growth. In addition, the opening of our Sb cracker is much narrower than the opening of our Knudsen cells. Thus, fewer Sb atoms are intercepted on the way to the substrate, reducing the need for regular clean-up and overall using less Sb. Finally, there are also benefits to the properties of antimonide films themselves from using cracked Sb. Use of monomer Sb has been shown to improve the properties of optical devices[51], and reduce film defects[52]. Care must be taken to prevent clogging of the different zones of the cracker, which is accomplished by the presence of extra heater filaments where the path of the flux is constricted.

3.2 Film analysis techniques

3.2.1 RHEED

Reflection high energy electron diffraction, known as RHEED, is the primary in situ technique for monitoring growth in real time, as it is performed within the chamber itself and modulates depending on the growth parameters. RHEED signatures, read by an experienced grower, give vital information about how the crystal is forming as it happens, both allowing for real-time corrections and later analysis.

The RHEED system consists of a tungsten filament through which a high current is run while under vacuum. The electrons that boil off the filament are then accelerated by a 10 kV potential through a collimating region and a focusing lens. Parallel plates at the tip of the RHEED gun allow for controlled deflection of the electron beam when varying voltages are applied. The electrons are aimed so that they strike the surface of the substrate at a glancing angle, around 1 or 2 degrees. The electrons diffract off the first few layers of a crystalline substrate and then strike a phosphor screen, allowing for visual evaluation of the diffraction
pattern. A CCD camera is used to record the RHEED images into a computer for brightness analysis in real time.

RHEED patterns from a nearly atomically flat MBE growth show patterns made up of spots, which denote where the Ewald sphere intersects the allowed reflections in $k$-space, illustrated in figure 3.4. For a 3D crystal, the permitted reflections would be dots in $k$-space, but in 2D they are elongated into rods. The sharper the RHEED spots, the smoother the surface of the film. However, a complex crystal like GaAs does not manifest a perfectly smooth surface, but complex, anisotropic reconstructions. For GaAs and the other III-V semiconductors, the best RHEED patterns look like sharp and narrow elongated streaks. Depending on the orientation of the substrate, different periodicity in the streak patterns can be seen, as shown in figure 3.5.

The RHEED reconstructions of GaAs and GaSb surfaces under normal growth conditions have been extensively catalogued in earlier work. In the As-stabilized regime, GaAs and InAs show a (2x4) reconstruction. This means that when the RHEED beam is aimed along the (110) direction, secondary streaks show half the periodicity of the primary streaks, while in the (-110) direction the secondary streaks have a quarter of the primary periodicity. The reason behind this is the structure of the dangling bonds on the As-terminated surface. If transitioned into a Ga- or In-terminated regime, for instance by reducing the As overpressure or increasing the substrate temperature, the reconstruction will switch to (4x2), where the period is halved in the (-110) direction and quartered along (110)\cite{46}. Observing the surface reconstruction transition is key to finding the appropriate growth parameters. A similar RHEED transition occurs for both GaSb and AlSb between group-III and group-V terminated regimes. However, in these cases the reconstruction along (110) is first order, having only primary streaks, and that along (-110) has a periodicity of one third. As in the case of GaAs, the reconstruction change is an important indicator in choosing the right growth parameters.

The moments where RHEED analysis is most essential are during drastic changes in
Figure 3.4: Ewald sphere construction for diffraction from a 2D film; side view and top-down view.
Figure 3.5: Half-order (left) and quarter-order (right) RHEED patterns for smooth GaAs.

the growth process. This includes the very beginning of the growth, and when material composition is changed. RHEED also gives a clear signature during the last step of substrate preparation, when the amorphous native oxide on a GaAs substrate must be desorbed. The rapid appearance of a very bright, streaky pattern is used to calibrate the infrared pyrometer used to measure the substrate temperature. Deviation from ideal growth parameters tends to show up first as transmission spots in RHEED. This means that the film is growing in faceted, 3 dimensional islands through which the electrons are diffracting, a growth mode we actively endeavored to observe in GaSb. Meanwhile, rings indicate polycrystallinity, while blurred and diffuse reflections indicate the development of an amorphous surface.

Figure 3.6: RHEED oscillations captured during the growth of GaAs.
Another important function of RHEED is in the determination of growth rate by use of RHEED oscillations. In a layer by layer growth mode, the surface of the film can be conceptualized as alternating between a rough appearance and a smooth one. That is, the film is rough when an incomplete monolayer is present, and becomes smooth when the monolayer completes, then becoming rough again as the next monolayer begins to form. The specular reflection of the RHEED beam is dimmer when reflecting off a rough surface due to scattering. By monitoring the light intensity of the specular spot appearing on the phosphor screen over time, very clear periodic oscillations can be seen, and since each one oscillation period corresponds to a single atomic layer, the growth rate can be extracted from the time graph. Pictured in figure 3.6 are a set of typical RHEED oscillations for a GaAs growth. Growth rates for all III-V semiconductors grown for this thesis were between 0.5 and 2 Å per second as determined by RHEED.

3.2.2 Transport measurements

Measurement of electrical resistivity is essential to characterizing a newly grown film. Using either a square van der Pauw sample or a Hall bar device geometry, it is possible to extract the carrier density and mobility of a sample, as well as study the dependence of resistivity on temperature. The traditional 4-terminal van der Pauw method for determining sheet resistivity was used for the square samples, for which

$$R_s = \frac{\pi R}{\ln 2}$$  \hspace{1cm} (3.2)

The Hall bars have an aspect ratio of 5 squares, so the sheet resistance is found by running current from end to end and taking measurements across the longitudinal voltage taps, then dividing the resulting resistance by 5. The Hall effect is measured in both types of samples by measuring voltage perpendicular to the direction of the current flow.

Van der Pauw samples were created by cleaving samples into an approximately square
shape with a diamond scribe. The four corners of the square were contacted with melted indium using a soldering iron, and contact fingers were pressed onto the indium and connected to a voltmeter and current source. Hall bar samples, on the other hand, were fabricated using UV photolithography, then etched down to the substrate in non-device areas. The contact pads made by the Hall mask were contacted by wire bonding with indium wire, and then connected to the voltmeter and current source. A LabVIEW program was used to record the raw voltage data. Measurements for this thesis were performed in a Quantum Design PPMS system, which reaches 1.8 K, or 300 mK with the low-temperature attachment.

3.2.3 AFM

Atomic force microscopy, or AFM, is a surface scanning probe technique that provides high resolution topographical data for thin films. From the nature of the surface, it is possible to extract information about the underlying layers as well, especially when paired with a cross-sectional technique like TEM.

AFM is performed by scanning a very fine tip across the sample surface. The tip is attached to a cantilever whose deflections are precisely monitored with laser interferometry. In tapping mode, which is how the AFM scans in this thesis were performed, the tip does not actually touch the sample, but is brought just close enough to experience van der Waals forces from the surface atoms. This is enough to extrapolate a topographical heat map of one of our semiconductor films, and is able to resolve single-monolayer steps in film height as well as complex terracing behavior. For our films, AFM resolves the locations of spiral defects at the center of pyramidal structures, from which growth spread outwards in an island-nucleation 2D growth mode.
3.2.4 TEM

Transmission electron microscopy (TEM) and related techniques high resolution-TEM (HRTEM) and scanning transmission electron microscopy (STEM) were performed by members of Professor Jian-Min Zuo’s group. These are cross-sectional techniques that allow us to view grown films from a point of view not achievable by looking only at the surface. Samples prepared for TEM were sectioned and thinned to 20-30 nm by focused ion beam. When a sample is sufficiently thin, electrons can transmit through the section and be detected on the other side. Variations in the diffraction and scattering that happens within the sample supply the contrast that appears in the final image. TEM is an imaging technique, in which a highly uniform electron wave diffracts through the section and the diffraction pattern is imaged. A Fourier transform performed on the diffraction pattern returns an image of the actual sample. STEM, on the other hand, is a scanning technique, where the TEM electron wave is focused down to a very small spot. The spot is then scanned across the sample. When columns of atoms within the crystal deflect the beam, the beam is detected and the location of the atoms precisely determined. The technique can distinguish elements of different masses, as well as be used for spectroscopic measurements. With these techniques, it is possible to distinguish layers of materials and analyze crystal dislocations from the high-resolution images obtained.
Chapter 4

Lattice Transition: A Crystallographic Study

In this experiment, we study the transition from GaAs to GaSb in real time. By closely examining the time evolution of RHEED during the transition, and aided by AFM and TEM images of films interrupted at various points in the transition process, we are able to reach conclusions about how exactly the lattice transition occurs. AFM taken after 1.4 and 14 ML of GaSb shows tall islands with low surface coverage, which enlarge and merge with neighboring islands as growth continues. STEM shows that the regular misfit dislocation array many experiments have observed in this system is already in place after 1.4 ML of growth. The moment at which 3D transmission spots appear corresponds to the same time at which the RHEED spacing begins to change. Therefore, we are able to determine that the formation of strain-relaxing dislocations occurs simultaneously with the appearance of a complex Stranski-Krastanov 3D island growth mode, which begins after only 0.4 ML of GaSb has been deposited onto the GaAs substrate. Rather than occurring independently or one effect leading into the other, the two phenomena are coincident. We also observe signs of the presence of strain in the underlying GaAs substrate once Sb atoms are introduced, leading to buckling or waving in the substrate surface. This is connected to the elastic Asaro-Tiller-Grinfeld (ATG) instability.
### Table 4.1: InAs and antimonide data from [2]

<table>
<thead>
<tr>
<th></th>
<th>GaAs</th>
<th>GaSb</th>
<th>AlSb</th>
<th>InAs</th>
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<td>Lattice constant (Å)</td>
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<td>6.10</td>
<td>6.14</td>
<td>6.06</td>
</tr>
<tr>
<td>Band gap (eV)</td>
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<td>0.78</td>
<td>1.35</td>
<td>0.36</td>
</tr>
<tr>
<td>% Mismatch relative to InAs</td>
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<td>0.7%</td>
<td>1.3%</td>
<td></td>
</tr>
<tr>
<td>% Mismatch relative to GaAs</td>
<td>8.0%</td>
<td>8.7%</td>
<td>7.3%</td>
<td></td>
</tr>
</tbody>
</table>

4.1 Growth of antimonides on GaAs

4.1.1 Substrate considerations

An ideal choice for nearly lattice-matched epitaxy of InAs would be AlSb. It is the most insulating of the 6.1 Å family ($\rho = 10^6$ Ω cm), and its lattice constant of 6.14 Å is a 1.3% mismatch from the 6.06 Å lattice constant of InAs. However, due to its aluminum content the compound is too highly reactive in air to be stable enough for substrate preparation[53]. For this reason GaSb (lattice parameter 6.10 Å) must be considered over AlSb[2]. Unfortunately, the technology of GaSb epiwafer production has not caught up to that of the much more frequently used GaAs. Commercially available wafers, while not intentionally doped, are $p$-type conductors with $n_h$ around $10^{17}$ per cubic centimeter. Such high substrate conductivity causes it to resistively shunt the devices grown and fabricated on top of it.

The absence of a sufficiently insulating lattice-matched substrate leaves open a significant question of what substrate should be chosen for a given application. Substrates that are often picked include InP and GaAs. However, InAs grown on InP tends to have a large number of threading dislocations[54]. Much III-V MBE growth is done on GaAs (with a lattice constant of 5.653 Å), as due to its ubiquity in semiconductor technology the material is widely available and commercially produced at a very high quality. These substrates are electrically insulating, largely free of defects, and smooth with the surface consisting of wide monolayer-thick terraces. For the growth of GaSb on GaAs to work, a detailed understanding of how the crystal transitions between lattice parameters is required, and is the focus of our crystallographic study.
To answer the question of how the dislocations form, we turn to both in situ and ex situ analysis of the film during and after growth. First of all, reflection high energy electron diffraction, or RHEED, is a method of real-time monitoring ubiquitous in MBE chambers, and provides a highly sensitive probe that measures the lattice spacing and characterizes the surface. RHEED can differentiate between 2D and 3D growth from the appearance of streaky or spotty patterns, respectively. 2D and 3D growth modes can also be shown to coexist by RHEED. As the diffraction pattern is a projection of \( k \)-space for a crystal, the distance between features is inversely proportional to the lattice spacing. Our results, detailed further on in this chapter, examine the qualitative RHEED features simultaneously with analysis of the spacing as a function of time during the first moments of lattice mismatched growth.

Atomic force microscopy (AFM) is an ex situ method for studying the topographical features of the film surface. AFM is a very high resolution mechanical scanning probe technique, in which a sharp tip is scanned across the sample repeatedly. Deflections in the tip position, monitored by optical and piezoelectric methods, are used to calculate the surface topography. AFM measurements performed for this thesis were able to resolve monolayer-thickness step variations, as well as the spiral dislocations in lattice-mismatched films, which correspond to threading dislocations underneath the surface.

Finally, our study is complemented by a third technique, transmission electron microscopy (TEM), and its more advanced sibling technique, scanning transmission electron microscopy (STEM). Both of these techniques use the diffraction of electrons through a very thin cross-sectional sample. TEM scans can show the propagation of threading dislocations, and properly interpreted can inform of their structure. High resolution TEM and STEM can even resolve the individual atoms making up a crystal, providing essential details in the study of film formation.
4.1.2 Growth Process

Our samples were grown on (100) GaAs substrates purchased from AXT. Each growth took place on a quarter of a 2 inch wafer. First, the substrate was cleaned by sonicating in solvents, loaded into the UHV transfer tube, and outgassed overnight. The native oxide was removed right before by heating the substrate to around 600 °C under an As flux. Oxide blowoff was monitored by infrared pyrometry and RHEED, which shows a clear qualitative change in brightness when oxide blowoff happens. The RHEED change was also used to calibrate the pyrometer from growth to growth.

After the oxide blowoff, the substrate temperature was adjusted to 580 °C. Next, around 200 Å of GaAs was grown, during which we observed the RHEED streaks brightening and becoming more established, indicating the rough surface post-oxide blowoff was becoming smooth. At this point, we grew 15 repetitions of AlGaAs-GaAs superlattices (Al percentage between 20 and 30 percent), which served to smooth and clean the surface further. The RHEED would become considerably brighter and streakier during the superlattice growth, indicating further improvement in smoothness. Specular spot oscillations provided a precise measurement of growth rates and confirmation of the amount of Al in the alloy. Following the superlattices, a layer of GaAs 2000 Å thick was grown at a rate of 1.8 Å per second to provide the best possible surface for the antimonide layers coming next, determined by the RHEED improvement leveling out around this point.

As demonstrated by Huang[16] and Jallipalli[55], the interface conditions between the GaAs and the GaSb to follow are very important parameters that determine how the GaSb relaxes. Our growth experience concurs with theirs in that the GaAs-GaSb transition bond should involve a terminating layer of Ga atoms bonding to Sb atoms, as under less optimal conditions the RHEED quality suffers. Since the GaAs is grown in the As-stabilized regime, which terminates with As atoms, the surface As must be replaced with Sb. Once the substrate temperature has been lowered from the GaAs growth temperature of 580 °C to
around 520 (a number that is not trivial to determine due to GaAs being transparent in the infrared at this temperature), the background As pressure is stopped for 30 seconds. During this time, the RHEED dims significantly. The details of the RHEED progression with time are examined later in this chapter. Finally, once excess As has evaporated, the Sb shutter is opened for 30 seconds, so that the Sb atoms bond to the surface Ga atoms. At this point, antimonide codeposition can start.

### 4.2 Antimonide nucleation

We have a choice of nucleating the growth with either GaSb or AlSb. Of the two, AlSb has a slightly higher cohesive energy[56], but whether this would overall benefit the growth is not clear. A stronger crystal might resist dislocations better, but a weaker crystal might relieve strain more efficiently. The values for the elastic moduli of the two compounds are very similar, with different calculations placing one or the other slightly ahead[57].

Kroemer[2] notes that while GaSb nucleation leads to very rough growth initially, it does not persist as more GaSb is grown. On the other hand, while AlSb nucleation begins with less roughness than GaAs, the growth does not smooth out with increased thickness. Nguyen[6] studied thick layers, 1 micron or greater, of AlSb and GaSb for the purpose of growing InAs, and found that thick GaSb alone was preferred for obtaining high mobility InAs. In contrast, Zhou[58] and Wang[59], found that insertion of a very thin AlSb nucleation layer could improve the smoothness and some optical properties of the film. In our experiment, we evaluate these claims and attempt to provide more detail to characterize the varying growth methods.

We compare three compositions for the initial growth. Each sample is topped with a 2000 Å thick layer of GaSb. Sample 0600 has no buffer separating the GaSb from the substrate; sample 0603 contains 10 ML of AlSb as the nucleation layer prior to the thicker GaSb growth. Finally, 0604 begins with AlSb, but before the GaSb starts, a 10 repeat
(a) 0600 - no AlSb. Spiral dislocation density $2.6 \times 10^8$ per square centimeter; RMS roughness 911 picometers.

(c) 0603 - 10 ML of AlSb. Spiral dislocation density $3.3 \times 10^8$ per square centimeter; RMS roughness 723 picometers.

(e) 0604 - 10 ML of AlSb and a 10-repeat superlattice. Spiral dislocation density $5.0 \times 10^8$ per square centimeter; RMS roughness 612 picometers.

Figure 4.1: AFM showing monolayer-height terraces for samples 0600, 0603, and 0604, showing atomic terraces and spiral dislocations. A speck of debris may be seen on the surface of 0600 and 0603. Spiral dislocations were counted in a 5 µm by 5 µm area.
short-period superlattice layer is inserted, for a total superlattice thickness of 250 Å. Under AFM, all three samples show pyramidal terraced structures with monolayer step heights. Pyramids tend to center around one spiral dislocation, which appear to act as a nucleation site from which growth spreads out. Counting the spiral dislocations in a 5 square micron area shows that sample 0600, with no inserted AlSb layer, has the lowest dislocation density, while 0604, with AlSb and a superlattice, has the most, with the density of 0603 falling closer to 0604. The dislocation densities are \(2.6 \text{ per } \mu\text{m}^2 (2.6 \times 10^8 \text{ per cm}^2)\) for 0600; \(3.3 \text{ per } \mu\text{m}^2 (3.3 \times 10^8 \text{ per cm}^2)\) for 0603; and \(5.0 \text{ per } \mu\text{m}^2 (5.0 \times 10^8 \text{ per cm}^2)\) for 0604. However, the film is rougher without the insertion of AlSb. Over a 25 micron scan, 0600 has an RMS roughness of 911 picometers. 0603 has a roughness of 723 pm, while 0604 has a roughness of 612 pm. Our finding agrees with [59] here in that the AlSb reduces the height of the pyramidal mound shapes that form.

Cross-sectional TEM was also performed on the three films. Threading dislocations are visible in all the images, some of which follow a straightforward trajectory to the surface, while others have more complex and winding paths. Examining the number of dislocations in each sample, we see that 0604 has the most while 0600 has the least, matching what was seen with AFM. In some of the TEM, the interfacial misfit array is also visible as regularly spaced spots. This is the same phenomenon observed in previous studies[16].

From this portion of the experiment, we learn that the choice of antimonide compound for the nucleation is a trade-off between dislocation density and surface roughness. A future experiment might be performed using superlattices or thick buffers to further reduce threading dislocations. Our study of antimonides on GaAs focuses next on the initial nucleation GaSb, as we have chosen to prioritize reducing the number of dislocations.
Figure 4.2: Cross-sectional TEM showing threading dislocations in 0600, 0603, 0604.
Figure 4.3: TEM and STEM of sample 0579, a film nucleated with a thin layer of AlSb, showing both coarse and atomic resolution images of the regular misfit array. Analysis of the images gives an array spacing of 53.7 Å, which is consistent with previous work.
Figure 4.4: Evolution of GaAs RHEED into GaSb RHEED.
4.3 RHEED analysis of initial GaSb growth

We examined the initiation of the growth of GaSb on GaAs by recording the RHEED pattern with a CCD camera linked to a computer over the course of the growth of sample 0600. Frame captures of this video quantitatively reveal the salient structural transitions linked to each step preceding and following the GaSb nucleation. The full GaSb growth took just over 30 minutes from beginning to end. The video recording of the lattice transition was recorded at 7 frames per second, result in 962 frames of video, or 137 seconds. The timeline of the shuttering sequence is shown in figure 4.5a. Our new experimental finding is that as the larger lattice spacing GaSb is grown on the smaller lattice spacing GaAs, the lattice strain relaxes at precisely the same time as the growth converts from a 2D strained layer to 3D islands via a Stranski-Krastanov transition. We determine this by examining the diffraction patterns obtained by RHEED during the first moments of the growth of the GaSb film in detail as a function of time, noting both the shape of the patterns and the spacing of the features in $k$-space.

The data set begins with the final RHEED pattern at the end of the base GaAs growth. It is bright and streaky, and the growth generated RHEED oscillations after brief growth interruptions. Next, the substrate temperature was lowered from 580 C to around 520 C. The RHEED keeps its streaky quality, but dims substantially in brightness (figure 4.5b). Once the RHEED has stopped dimming, the As shutter is closed for 30 seconds (figure 4.5c). At this temperature, the RHEED does not switch reconstructions from (2x4) As-stabilized to (4x2) Ga-stabilized as would be expected at a higher temperature, but it regains some brightness.

After 30 seconds without As, the Sb shutter was opened for 30 seconds, in order to allow the replacement of desorbed excess surface As with Sb atoms. This converts the surface from As- to Sb-stabilized, and marks the first drastic change in the RHEED pattern. The RHEED switches to a (1x3) Sb stabilized reconstruction, seen in figure 4.5d, although at this point
the one-third order streaks are not yet visible. The spacing between streaks, corresponding inversely to the lattice spacing, has not yet changed at all, implying that the surface atoms have the same lattice spacing as the bulk atoms. Once 30 seconds of Sb soaking have elapsed, the Ga shutter is now opened, which starts the growth of GaSb. Within the first two seconds, transmission spots emerge along with and then eclipse the streaks (figure 4.5e). For thicker films, the initial GaSb is grown for 30 seconds and then interrupted to allow for annealing time. RHEED oscillation data determined a 1.2 Å per second growth rate for GaSb for a Ga temperature of 950 C. Within a minute of beginning GaSb growth, the transmission spots have begun to elongate and return to a streak shape, and by 5 minutes the spots have disappeared altogether.

Having recorded RHEED images throughout the entirety of the transition process, we analyzed them using image processing software to bring out the important features. The first of these is the streak spacing, which corresponds inversely to the lattice spacing of the crystal. Our analysis consisted of taking an intensity section over a wide line both perpendicular and parallel to the streaks, once for each frame of the RHEED video. Plotting the line scans for the first few seconds of GaSb on the same graph, we can pinpoint the exact frame at which the streak spacing begins to change in both the planar and $z$-directions to be frame 745, just after the first second of growth. The other characteristic we look for is the moment a spot pattern begins to emerge from the streaks, most easily spotted in the parallel section (figure 4.7. This indicates the beginning of the transition from 2D to 3D growth, or the transition from the Stranski-Krastanov wetting layer to 3D islands. Again, we pinpoint the correct frame by examining the spacing between features in $k$-space. Both events occur in frame 745 of the video, or 1.4 seconds into the growth of GaSb, which corresponds to a nominal GaSb thickness of 0.4 ML. The relaxation of the GaSb crystal and the formation of the islands occur simultaneously, which we have determined to a precision not previously observed. Before this occurs, RHEED indicates that the surface is a strained sub-monolayer of GaSb. Afterwards, the strain is relaxed and the atoms have migrated to form islands
Figure 4.5: Timeline of the GaSb nucleation process.

(a) Timeline

(b) 1. As-stabilized quarter-order streaks at 520 C.

(c) 2. Quarter-order streaks brightened by turning off As flux.

(d) 3. Switch from (2x4) As-stabilized to (1x3) Sb-stabilized after turning on Sb.

(e) 4. 3D transmission spots after 6 seconds of GaSb deposition.
Figure 4.6: Frame-by-frame intensity analysis of the RHEED pattern during the first 4 seconds of film 0600, made by taking a line scan parallel to the shadow edge. Each frame corresponds to one seventh of a second. Vertical lines denote the streak location prior to the change in material composition and make the change in streak spacing obvious, while the lower red line represents the position of the peaks of the quarter-order GaAs reconstruction for comparison. The first movement in streak position occurs three frames after 1 second of growth.
Figure 4.7: Frame-by-frame intensity analysis of the RHEED pattern during the first 43 seconds of film 0600, made by taking a line scan perpendicular to the shadow edge. The vertical lines denote the initial positions of 3D transmission spots when they first emerge. The first appearance of a 3D transmission spot occurs three frames after 1 second of growth.
on the surface through which 3D diffraction occurs, leading to the spots observed in the diffraction pattern.

4.4 Interrupted growths: STEM analysis

The group of films consisting of 0600, 0603, and 0604 were all topped with a layer of GaSb 2000 Å in thickness, and the information extracted from them informs us about the nature of the growth well after the initial transition has taken place, and the GaSb film has had plenty of time to relax and become smoother. In particular, the TEM images and the atomic resolution STEM images show that the lattice relaxes by the formation of a regular array of misfit dislocations running in the (110) directions, located right at the interface between the GaAs substrate and the overlaying GaSb film. The misfit array has also been observed earlier by other investigators[16][18]. The RHEED analysis in the previous section indicates that the lattice relaxation occurs at the same instant that the growth mode converts from 2D layer-by-layer growth to 3D Stranski-Krastanov growth. We would like to observe this in real space as well, by examining films interrupted just after the transition occurs. Here we relied on collaboration with the group of Professor Jian-Min Zuo and Dr. Xun Zhan, a post-doctoral researcher in his group. Our objective was to use TEM and STEM to examine similar films that have been interrupted at crucial moments closer to the initial nucleation, and gain more information about the early growth.

A pair of GaSb growths following the architecture of 0600 were grown, each one having been interrupted at a different moment during the formation of the GaSb layer. Growth of sample 0615 was stopped by closing the Ga shutter, after only 4 seconds of GaSb deposition. At a growth rate of 1 Å per second, the resulting film was nominally 4 Å thick, which corresponds to 1.3 ML (3.05 Å) in thickness. 0616 was a nominal 40 Å thick before its growth was interrupted. As expected from the background literature, however, we do not observe flat films under AFM for either of these growths. Rather, 0615 is covered in many
Figure 4.8: AFM topography maps and cross sections of islands at 1.3 and 13 ML of nominal GaSb deposition. Note the very different vertical scales for 0615 and 0616.
Figure 4.9: 0615 - 1.3 ML of GaSb. Misfit dislocations denoted by red arrows.
Figure 4.10: 0616 - 13 ML of GaSb. Misfit dislocations denoted by red arrows.
small dots which are considerably taller than the nominal thickness of the film, around 3 nm in height, with a width of around 20 nm. The height of the dot features make up for the greatly reduced film coverage. 0616, at nominally 10 times thicker, still shows the island-style morphology, but the islands have proliferated further, and features have widened to around 50 nm. The island height is around 8 nm at this point. Inspection of the AFM scans allows us to determine the island density: 237 per square micron for 0615, and 327 for 0616.

High resolution STEM images show the nature of the misfit dislocations at the highest level of detail, as individual atoms in the crystal can be resolved. In figures 4.9 and 4.10, it is clear that the islands already contain a dislocation array. We can also see to the left side of the 0615 island the bonds that are still stretched right after the second dislocation forms, which would eventually be brought to an equilibrium position if growth continued. The presence of misfit dislocations at 1.3 ML places an upper limit on our determination of the critical thickness for GaSb. It also agrees with the finding from the time-resolved diffraction analysis presented in the previous section.

Further examination of the AFM images of sample 0615, with 1.3 ML of GaSb deposition, reveals another interesting feature. In figure 4.11, it can be seen that the mostly flat surface beneath the islands is in fact not completely flat, but takes on a wavy morphology. Examination of the STEM images discussed above as well as a particularly revealing HRTEM image (figure 4.12) show the complex shape of the dots. The flatter island region below the visible misfit dislocation is attributed to surface GaAs buckling due to the influence of Sb and GaSb present on the surface and changing the energy landscape. Above the flat island is a rounder island composed of GaSb. The wavy morphology can be attributed to the ATG instability on the surface of the film.
Figure 4.11: 0615 - 1.3 ML of GaSb. Beneath the dots, a buckled or wavy surface is seen
Figure 4.12: 0615 - 1.3 ML of GaSb. Cross-sectional HRTEM image revealing the dual island shape.
Chapter 5

Transport in an InAs surface well

The purpose of the next portion of this work was to induce superconductivity into a strongly spin-orbit coupled semiconductor quantum well by means of the proximity effect. The majority of other quantum well studies for 2D physics use two-sided semiconductor heterostructures, so that the carriers in the well are located far from the surface, making the scattering time long enough to provide high mobility. The protection a large offset from the surface provides can give mobilities in excess of $10^6$ cm$^2$/V·s$^6$. However, the distance from the surface reduces the probability of Andreev reflection between the electrons in the quantum well and the overlaying superconducting material, which weakens the proximity effect. In this thesis, we explore the opposite limit, where the quantum well is located directly on top of the insulating semiconductor heterostructure. With in situ deposition of a film of superconducting niobium, this provides an essentially perfect coupling between the two layers of quantum well and superconductor. Vissers et al.\[9\], found that when deposited in this way, the contact to niobium can be at the Landauer limit, with transmission coefficient per channel equal to 0.7. In this work, we explore transport and scattering in very thin InAs surface quantum wells. We find that the scattering significantly limits the transport, but the transport is metallic and exhibits Shubnikov de Haas oscillations, and provides a low temperature mobility of 5369 cm$^2$/V·s.

The accumulation of carriers at the surface of a film was first studied for the purpose of creating electro-optics devices, such as light emitters\[60\]. Surface quantum wells have been observed in several thin film systems, including GaN and InP. That the surface accumulation of electrons in InAs could be employed in proximity effect experiments was noted back in
1987 by Kawakami[61], who found that interfacial properties were key to the induction of superconductivity into the quantum well. This experiment used very thick InAs and probed its bulk properties. In contrast, in our study we work towards characterization of very thin InAs surface wells and their use in island array devices, which are used to probe for interesting vortex behavior.

Having extensively studied the structural properties of the antimonide base layers, we now turn to the properties of InAs surface quantum wells. First, it was necessary to work out the optimal growth conditions for InAs. Transport measurements in order to characterize the mobility and carrier density of InAs films were performed, and a process for creating island array samples is currently in development.

5.1 Growth of InAs transport samples

The previous chapter detailed the process of growing antimonide buffers on GaAs for the end purpose of creating the best possible lattice matched surface on which to eventually grow InAs. For our InAs devices, we eventually elected to use a buffer layer containing only 2000 Å GaSb, with no Al content. While the buffer structures that contained AlSb resulted in smoother surfaces, the total spiral dislocation density was higher in the resulting films. Thus, in order to minimize threading dislocations we determined that GaSb alone would be the preferable buffer material.

Sample 0568, on which transport measurements were performed, was nucleated with AlSb and a short period superlattice, similarly to sample 0604 of the nucleation study. Its buffer layer was 2000 Å of AlSb, which was not our eventual optimal structure yet still yielded the transport characteristics of a high quality crystal. The InAs layer is 156 Å thick and contains no more than 2 quantum transport channels.

The growth of our proximity effect sample, 0618, began identically to that of sample 0600, one of the samples for our buffer layer AFM and TEM study. Growth began after
Figure 5.1: Schematics of transport samples. 0568 was measured in Hall bar form. 0618 is intended for island array measurements.

The oxide blowoff of the substrate at 600 °C with about 200 Å of GaAs grown at 580 °C. This was followed by 15 repeats of AlGaAs-GaAs superlattices, and then 2000 Å of GaAs. The substrate temperature was then lowered and the growth transitioned to GaSb, using the same shutter sequence detailed in the previous chapter to begin growth on an Sb-rich surface. After 2000 Å of GaSb was grown, the transition to InAs began. The InAs in this sample was 70 Å thick, calculated to have only one quantum channel. 200 Å of Nb was deposited \textit{in situ} after growth by electron beam evaporation.

For the best growth of InAs on GaSb, it is essential that the transitional layer be “InSb-like” rather than the alternative, “GaAs-like.” That is, the terminating element of the GaSb ought to be Sb, which must bond to the first In layer of the InAs. Extensive studies on the interface between GaSb and InAs were made in several papers by Haugan \textit{et al.}[62][63][64], for the purpose of optimizing InAs/GaSb superlattices for infrared detection. However, crystalline defects in optical applications are just as deleterious as they are in transport experiments, so we were able to apply their interface methods to improve the quality of our InAs.

One factor found to be important in the Haugan \textit{et al.} studies was the background As pressure, as they found that excess As that became incorporated into the GaSb would lead to defects and decrease the quality of the film[64]. For this reason, their growths were
performed at a low flux of As. For our growths, we endeavored to grow at an As to In ratio just above the point at which the surface reconstruction switches from As to In-terminated, staying in the flux ratio regime that is known to grow stoichiometric InAs. One consequence of straying too low in group V flux is the emergence of oval defects caused by nucleation of group III islands. As discussed below, this was observed in sample 0568, which showed very good transport properties but also manifested oval defects on the surface. It appears there is a delicate balance between these two concerns when growing the best InAs films, and the optimal parameter window is very small. Furthermore, in surface quantum wells the films are so thin that the transport properties programmed in by the crystal structure at the GaSb-InAs interface are likely to cause scattering to a degree similar to what the free surface causes. Therefore, transport in a surface well is likely to be strongly affected by scattering, and the question of whether metallic transport is crucial to understand.

More important for our purposes is the shuttering sequencing to ensure InSb-like bonds. Our method, again based on those used by Haugan et al., is to first ensure the terminating surface of the GaSb is composed of Sb. This is easily done in the Sb-rich growth regime in which our materials are grown. After the last of the GaSb is grown, it is allowed to sit and anneal under the Sb flux to fill any vacancies. However, if the Sb shutter is now closed and InAs deposition begun at this point, there is the possibility that Sb that desorbs will be replaced by As, resulting in GaAs-like bonds. To prevent this, the interface is grown by migration-enhanced epitaxy (MEE). The Sb shutter is closed at the same time as the In shutter is opened. After the In has been opened for the time it takes to deposit one ML of In. Next, the As is opened to begin InAs codeposition, and the interfacial bonds are ensured to be InSb-like. In our experience, the efficacy of the MEE approach can be seen in the evolution of the RHEED over the course of the InAs growth. With the single ML of In at the interface included, the RHEED noticeably improves over the course of the growth compared to growths without it.
5.2 Transport characterization

5.2.1 Device fabrication

The first step of the lithography process is to define the shape of the Hall bar device and contact pads. First, AZ5214E photoresist is spun onto the sample at 4000 RPM, then baked for 2 minutes at 110 C. The pattern is then exposed for 20 seconds through a soda-lime mask using a Suss MJB3 mask aligner, then developed for 40 seconds in AZ917 MIF developer. Next, the top layer of Nb is etched away from the non-device area by reactive ion etching using SF$_6$ (35W, 200 mT for 25 seconds). The now exposed semiconductor layers are removed down to the substrate by ion milling (75 mA, 300 V for 6 minutes), with the etch depth confirmed using a Dektak profilometer. The resist is then removed by sonicating in acetone. Control samples underwent a second UV exposure and RIE etch in order to remove all Nb from the Hall bar, so that Hall transport measurements could be made.

![Schematic of Hall bar design](image)

Figure 5.2: Schematic of Hall bar design, with current leads on the left and right sides, and 4 voltage leads. The entire device measures 3.2 by 3.2 mm. The bar itself, measured from the voltage tap locations, is 500 by 100 $\mu$m. For island array devices, a grid of single pixel lines 1 $\mu$m apart is drawn over the bar.

5.2.2 Hall effect measurements

Hall bar devices were measured in a Quantum Design Dynacool PPMS system with a base temperature of 1.8 K, having been bonded with indium wire to the contact pads of the
instrument’s sample holder. The current contacts are wired to a Keithley model 6220 DC current source, such that current flows along the length of the bar between the two current contacts. At the same time, two Keithley 2182A nanovoltmeters are connected to the voltage taps, one to measure the longitudinal voltage parallel to the direction of current flow ($V_L$), and one to measure the transverse voltage in the perpendicular direction ($V_T$). One full round of data points consists of four measurements: first, current is run in one direction and both longitudinal and transverse voltage readings are taken. Then the current direction is reversed, and a second pair of voltages are read. All four voltages, the value of the current, and the temperature and magnetic field values are recorded into one line of the data file. The process loops indefinitely, while the temperature and field can be changed at various speeds to adjust the data resolution of a sweep.

Hall measurements of sample 0568 revealed a 2D carrier density of $1.2 \times 10^{12}$ electrons per square centimeter, and a mobility of 5369 cm$^2$/V s. From this data it is possible to extract the mean free path, which comes out to be 96.9 nm. This number is what sets the length scale for the later island array devices. Measurements of the longitudinal resistance also displayed Shubnikov-de Haas oscillations, confirming the mobility data.

AFM taken of 0568 shows a high density of oval defects. These defects are thought to be caused by incorporation of excess Ga during growth[65], and are very common in growth of arsenides. From the cross sectional TEM, we can see that the oval defects lead to inhomogeneities in film thickness. Transport measurements taken of this sample are actually probing the smooth, homogeneous areas under and between defects. It has yet to be explained why InAs samples like this one with seemingly subpar morphology had the highest mobilities among our films, while films with much smoother surfaces had mobilities lower than 0568 by an order of magnitude.
Figure 5.3: Shubnikov-deHaas oscillations observed at 1.8 K in sample 0568. The number of oscillations is related to the mobility of the sample.

Figure 5.4: AFM and TEM of 0568. AFM shows a preponderance of oval defects, caused by excess As incorporation. The TEM image shows a cross section of one of these defects, revealing that the threading dislocations in the film are not correlated to these morphological defects.
5.3 Future steps

The next stage in our study of the proximity effect will be to fabricate island array devices. The experiment performed by Mulcahy[11] with Nb islands on films of Bi$_2$Se$_3$, a robust topological insulator, demonstrated how the resistivity of such a device reveals information about the proximity effect. As the array cools, its resistivity shows several transitions in the onset of superconductivity. In a measurement of resistance versus temperature, a drop will be seen around the critical temperature of the Nb. However, because the islands are not connected, the resistance will not drop to zero like we would expect for a continuous superconductor. Another drop will occur at a lower temperature, corresponding to the normal areas directly beneath the islands beginning to show proximity-induced pairing. Finally, at the phase stiffening transition, the normal regions between each island will become superconducting as well, leading to a uniform phase across the entire array. The phase stiffening transition is related to the Berezinskii-Kosterlitz-Thouless (BKT) transition, and is strongly dependent on the strength of the proximity effect as well as the vortex dynamics within superconducting regions. We would like to observe evidence of the phase stiffening transition in the surface quantum well of InAs, as superconducting vortices in this material have been predicted to manifest Majorana fermions.

The process of creating Nb islands on top of our InAs Hall bars essentially consists of etching many narrow trenches into the Nb slab. Our method being developed involves writing a grid of single pixel lines, spaced one micron apart, into a layer of polymethyl methacrylate (PMMA) using e-beam lithography. The lines are developed out in a solution of methylisobutylketone (MIBK) and isopropanol in a 1:3 ratio. After the lines are written into the PMMA, the sample is etched in SF$_6$ RIE. The Nb in the trenches is etched away, leaving a large grid of square islands spaced roughly 100 nm apart. A longitudinal resistance versus temperature measurement would reveal the proximity-related transitions and elucidate the nature of the superconductivity induced into the surface layer. Fabrication experiments for
these island devices are ongoing.

Figure 5.5: AFM images of Nb islands formed on sample 0618 using e-beam lithography.
Chapter 6

Conclusions

Our experiment examining the structural nature of antimonides grown on lattice-mismatched GaAs reveals essential information about the formation of misfits at the interface. We observe for the first time how the lattice relaxation of GaSb occurs simultaneously with the Stranski-Krastanov transition from 2D to 3D island growth. By understanding the transition between GaAs and GaSb, we are better able to engineer lattice matched growths of InAs material in order to study the properties of surface quantum wells. Finally, we are able to take the next steps into the study of the proximity effect in InAs and contributing to the search for Majorana physics in thin films.
References


