Thesis for the degree
Doctor of Philosophy

Submitted to the Scientific Council of the
Weizmann Institute of Science
Rehovot, Israel

By
Noam Morali

Advisor:
Dr. Haim Beidenkopf

September 2019
Acknowledgements

Writing this dissertation concludes a very significant chapter in my personal life, spanning the five years I have spent in the WASP lab. Acknowledging the amazing people I got to spend time with, is also great opportunity for me to recap and truly appreciate this experience.

Haim, my Ph.D advisor, who offered me a place in his group as a M.Sc student and guided me onward to this stage. I quickly learned you are truly a brilliant physicist with a remarkable intuition in both research and technological development. I later further discovered your mesmerizing presentations and tantalizing wording. I had the fortunate access to these core skills, which any experimental physicist wishes to have. Thank you for entrusting me with the VT-STM project, and for advancing the vision in all our joint works. I am positive the accomplishments listed in this dissertation would not have been possible without you.

Nurit, a true asset of our the group, offering a knowledgeable and unique perspective at any scientific junction. Your passion and love to physics, together with your deep insights triggered me continuously in my research. I am also thankful for your guidance in our joint projects, reminding me to follow data derived conclusions, and separating wheat from chaff.

Rajib, my main work partner. You and I worked so long together, it is impossible for me to imagine any milestone along the way without you in the frame. The synergy between us was just perfect, which really helped push forward any project we took on together. I wish you all the best in your new postdoc.

Special thanks goes to my current and past lab mates. Jonathan, who welcomed me into the lab and has since became my mentor in physics and beyond. Abhay, who taught me how to do everything in work more effectively. Aviram, who is firstly my friend, but has later became a colleague whose physical and engineering intuition really benefited me along the way. Tom, whose diligence ignited me and the whole lab. Pranab, my second work partner, thanks for putting your time and dedication into the measurements, which no doubt allowed their successful outcome. Mathi, who had to endure loads of information, both in MBE and the VT-STM, but still remained eager to contribute and achieve more. Alon, allegedly a summer student, but bonded well with all, kudos for your hard work who greatly contributed the VT-STM. Ben, whose excitement of science is stimulating. Finally, to our new M.Sc students - Ambikesh and Yotam, thanks for striving to learn, I am positive that the lab is in good hands.

No lab is an island, and I am also thankful for the partners I acquired along the path. Tal, who taught me all I know about proper grounding and carpentry, and the other memebers of Dan Shahar ‘s lab - Adam, Idan and Franzi, who were there for
me at times when I needed a physics advice, an inch screw or a cup of coffee; I will surely miss our joint lunch breaks. Binghai and Ady who contributed their enlightening theoretical modelling to our measurements. Claudia for her help in conceiving the experiments, together with the support of Yan and Qiunan. To Eli and Karen, my Ph.D committee members. I am further thankful to Aviram, Nadav and Yuri from Eli’s lab; Asaf and Lior from Shahal’s lab; Yonatan, Amir, Anna and Ron from Moty’s lab; Gal and Ran from Ofer’s lab, and other students from our faculty - for all of their advice and willingness to help.

Much gratitude goes to the condensed matter administrative staff for much needed logistic support and beyond - Hava my lunch partner and friend; Merav who shares my love to jachnun and fun; Einav and her appreciation of inappropriate jokes; And Yuri whose diligence and knowledge solves any issue. I would also like to acknowledge supporting arms outside of department. The physics machine workshop and the CNC workshop, ran by Harel and Haim, who literally made the microscope a reality, and their wonderful professional staff - Alex, Shmuel, Evgeny and Ady. Solomon, Ronen, Asher and Dima from the Helium liquid center, for their friendly and competent services.

Finally I am most thankful to my dear life companion Noy, and our beloved daughter Dolev. Thanks for rejuvenating me with your love and support every morning and evening. Our family, which was formed during my Ph.D, is by far my greatest accomplishment.
## Contents

Acknowledgements iii  
Abstract ix  

1 Introduction 1  

2 Methods 5  
2.1 Scanning Tunneling Microscopy 5  
2.1.1 The Tunneling Current 5  
2.1.2 Modes of operation 7  
2.2 Quasi-particle interference (QPI) 10  
2.2.1 QPI from Point Defects 10  
2.2.2 QPI from Step Edges 12  
2.3 Sample preparation 13  
2.3.1 Calibrating Field of View 13  
2.3.2 Single crystal cleaving 14  

3 Non-centrosymmetric Weyl Semimetal 15  
3.1 Introduction 15  
3.2 Results 16  
3.2.1 Quasiparticle interference of trivial bands and Fermi arcs in TaAs 16  
3.2.2 Bulk-Boundary correspondence 17  
3.2.3 Structure of the surface wave function 18  
3.2.4 Topological Bloch wave function 20  
3.3 Discussion 20  

4 Magnetic Weyl Semimetal 23  
4.1 Introduction 23  
4.2 Results 24  
4.2.1 Diverse surface band structure 24  
4.2.2 Fermi arc connectivity and time reversal symmetry breaking 25  
4.2.3 Inter Brillouin zone Fermi arc connectivity 28  
4.2.4 Weyl cone dispersion on the S termination 30
4.3 Discussion ........................................................................ 31

5 Construction of VT-STM ...................................................... 33
  5.1 Introduction ..................................................................... 33
  5.2 Microscope Assembly .................................................... 34
    5.2.1 Electro-polishing .................................................... 34
    5.2.2 Gluing piezo stacks ............................................... 37
    5.2.3 Gluing sapphire prism ........................................... 39
    5.2.4 Electronic wiring .................................................... 41
  5.3 System Grand Assembly .................................................. 44
    5.3.1 Suspension springs ................................................ 44
    5.3.2 Radiation shields ................................................... 46
    5.3.3 Cryostat .............................................................. 47
    5.3.4 Scanning Electron Microscope ................................ 49

Bibliography ........................................................................... 53

A Supplementary information TaAs ........................................ 59
  A.1 Sample synthesis ......................................................... 59
  A.2 Extended q-space map .................................................. 59
  A.3 $dI/dV$ maps: Raw data and symmetrization ................... 59
  A.4 Fermi arc scattering signature ........................................ 60
  A.5 Agreement between vacancy- and step edge–induced QPI .... 60
  A.6 Fermi arc dispersion ..................................................... 64
  A.7 Correlation between scatterer-free $dI/dV$ modulations and replications of QPI patterns ......................................................... 64
  A.8 Correspondence between QPI patterns and Bloch wave function .... 65
  A.9 Band structure calculations ........................................... 68
  A.10 Extracting the intensity of QPI features .......................... 68
  A.11 Splitting the line-cut $dI/dV$ into submaps ....................... 69

B Supplementary information $Co_3Sn_2S_2$ .......................... 71
  B.1 Single crystal growth of $Co_3Sn_2S_2$ .............................. 71
  B.2 ab-initio calculations ................................................... 71
  B.3 Identification of Co,S,Sn terminations .............................. 72
  B.4 Raw Fourier transformed $dI/dV$ data measured on Sn, Co and S terminated surface ......................................................... 73
  B.5 Spin and orbital character from ab initio calculation ............ 75
  B.6 The electronic structure of the Sn terminated surface ........ 76
B.7 The electronic structure of the Co terminated surface .......................... 78
B.8 The electronic structure of the S terminated surface .......................... 81
Abstract

Faculty of Physics
Department of Condensed Matter Physics

Doctor of Philosophy

Scanning Tunneling Spectroscopy Investigation of Topological Weyl Semimetals

by Noam Morali

The dispersion of the topological surface bands in Weyl semimetals is correlated with the topological bulk bands dispersion. The bulk boundary correspondence in these is manifested by a unique open contour surface state known as Fermi arc, connecting two surface projected Weyl nodes of opposite chirality. Bulk Weyl nodes are formed under broken inversion or time reversal symmetry.

In my thesis report I describe our contribution to this novel electronic phase, on two different materials, the non-centrosymmetric TaAs Weyl semimetal, and the magnetic Co₃Sn₂S₂ Weyl semimetal. Our spectroscopic STM experiments were among the first to demonstrate the contour and dispersion of the Fermi arc bands and their relation to the bulk Weyl cone. Moreover we found unique surface characteristics of the Fermi arc bands as opposed to trivial surface states. We obtained these results using an analysis technique, based on the role of the Bloch wave function in shaping quantum electronic interference patterns. We were further the first to spectroscopically verify a time reversal broken Weyl semimetal phase. Our results show how the distinct surface potentials imposed by different terminations of the crystal modify the Fermi arc contour and Weyl node connectivity. These joint studies revealed profound experimental insights by which we were able to resolve the topologically protected electronic properties of a Weyl semimetal and its unprotected ones.

In parallel to my research, I also constructed a home built variable temperature scanning tunneling microscope with an integrated scanning electron microscope system. The system granted the lab access to previously uncharted territories, in the realm of microscopic samples, such as WP₂ and SnTe, as well as silicon based devices for the study of InAs nanowires.
To Noy and Dolev
Chapter 1

Introduction

The field of topology was first introduced to condensed matter physics at the 1970s. Kosterlitz and Thouless theorized (Kosterlitz and Thouless, 1972; Kosterlitz and Thouless, 1973) the role of topological defects in phase transition, solving a long debate about phase transitions in two-dimensional (2D) systems. A decade later, Thouless (Niu, Thouless, and Wu, 1985) has turned to topological arguments in explaining the quantum Hall effect (QHE). QHE (Klitzing, Dorda, and Pepper, 1980) is a quantum-mechanical version of the Hall effect. In the quantum case, the conductor is confined to 2D and quantized values of the Hall conductance can be measured. This quantization does not depend on the type of substrate, and remains robust in the presence of defects or impurities.

Topology can be used to characterize phases of electronic matter as shown by Haldane on his 1D spin chains model (Haldane, 1983), and QHE in 2D lattices in the absence of a macroscopic magnetic field (Haldane, 1988). More recently, this framework gave rise to the field of topological insulators (Kane and Mele, 2005; Bernevig, Hughes, and Zhang, 2006) — phases of matter which exhibit conductance only on their surfaces, which are protected by time reversal symmetry. For their contribution to this field, Thouless, Kosterlitz and Haldane were awarded the Nobel prize of physics in 2016.

Note that in all of the above topological systems, strict constraints are in place. The QHE requires high magnetic fields to induce band gap, confinement to 2D, and low temperatures. With the incorporation of spin-orbit coupling, the 2D topological insulator phase no longer requires the usage of magnetic field. However, it maintains the dimensional confinement as well as the requirement of a bulk gap. For three-dimensional (3D) topological insulators, we further remove the former constraint, and solely demand a bulk gap, though of topological nature. In 2015 (Lv et al., 2015b; Xu et al., 2015b; Yang et al., 2015) the last constraint was relaxed with the discovery of topological semimetals - a topological phase of matter with no bulk gap.

In Dirac semimetals the conduction and valence bands touch at accidental discrete critical points in the Brillouin Zone (BZ) (Herring, 1937), and disperse linearly in all three directions around them (Wan et al., 2011). The low energy effective Hamiltonian
Chapter 1. Introduction

takes the massless Dirac or Weyl form, $H(k) = v_{ij}k_i\sigma_j$, where $\vec{\sigma} = \{\sigma_x, \sigma_y, \sigma_z\}$ are the Pauli matrices and $\det[v_{ij}] \neq 0$. Such a Hamiltonian is especially robust against perturbations because $H(k)$ contains all three Pauli matrices, thus any perturbation will simply shift the location of the Weyl point in energy or momentum but cannot remove it.

This protection can be expressed in topological terms by the Chern number of the valence band on a sphere surrounding one such point, which can be shown (Young et al., 2012) to be $\text{sign}(\det[v_{ij}]) = \pm 1$. Assuming a Weyl point at a specific $k$ point in the BZ, time reversal (TR) symmetry implies another Weyl point will exist at $-k$ with equal Chern number. However, since the overall Chern number of the Fermi surface is zero, two additional points are required with opposite Chern number, at $k'$ and $-k'$. Inversion (I) symmetry suggests the points at $k$ and $-k$ have an opposite Chern number. Ergo, for a material with both TR and I symmetry we infer $k = k'$. In this case, the band structure consists of doubly degenerate valence and conduction bands, and the two Weyl points overlap to give a Dirac point (Fig. 1.1A). In general, two Weyl points with opposite Chern numbers annihilate, which is why an additional crystal symmetry is needed to support this electronic phase. Indeed this was reinforced experimentally in $\text{Cd}_3\text{As}_2$ (Wang et al., 2013) where the $C_4$ rotation symmetry protected the doubly degenerate Dirac cone from gapping. Alternatively, by breaking either TR or I symmetry a Dirac point can separate to two Weyl points (Burkov and Balents, 2011; Burkov, Hook, and Balents, 2011).

The manifestation of the bulk boundary correspondence in the topological Weyl phase can be understood by examining the Berry flux lines around the two Weyl nodes, governed by the fact one node acts as a source and the other as a drain (Fig. 1.1B). This in turn leads to a non-zero Chern number in the region between the Weyl nodes. Therefore, one may think of a 2D plane cut through this intermediate region as a quantum Hall system. The accumulated chiral edge states of that quantum Hall like slices generate the 2D surface states known as Fermi arcs. These states, as their name implies, possess a unique property of having an open contour shape - emanating from one surface projected Weyl node, and terminating in another. It is of relevance to this work to note the topological properties of the band structure determine one Fermi arc will connect two opposite charge Weyl nodes. However, in materials hosting more than one pair of Weyl nodes, the topological properties do not entail the pairing scheme between the nodes.

The transport properties of Weyl semimetals vary from other topological electronic phases. In topological insulators the topological surface states exhibit weak antilocalization due to the helical spin structure and the lack of backscattering (Roushan et al., 2009). In stark contrast, the dc conductivity of Weyl semimetals is mostly governed by
the bulk states. Interestingly, the electronic crystal system of Weyl semimetals demonstrate a mathematical similarity to Fermionic systems of lattice gauge theories, thus exhibiting an effect analogous to the mechanism of the Adler-Bell-Jackiw axial anomaly (Adler, 1969; Bell and Jackiw, 1969), also known as the chiral anomaly.

For lattice systems, this anomaly manifests itself as the inter-valley charge pumping of electrons between the two cones of a Weyl semimetal (Nielsen and Ninomiya, 1983). One can ascribe the chiral anomaly to the chiral zeroth Landau levels which possess opposite velocity signs for Weyl cones of opposite chirality (Fig. 1.1C). Noting that the degeneracy of a Landau level is proportional to magnetic field, one infers that the application of a parallel electric field will give rise to a negative magnetoresistance, essentially favoring one chiral mode over another. This was indeed experimentally observed (Huang et al., 2015b; Zhang et al., 2016; Liu et al., 2018), and stand in contrast to conventional metals and semiconductors in which the magnetoresistance is usually weak, positive and isotropic.

In my Ph.D research I used scanning tunneling microscopy (STM) to investigate the electronic properties of two single crystal Weyl semimetals candidates. In parallel I also designed and constructed a variable-temperature STM (VT-STM). The structure of the thesis is as follows:

Chapter 2 describes the basics physics governing STM, focusing on the different modes of operation, as exemplified on a simple Copper metal surface.

In chapter 3 I present our study (Batabyal et al., 2016) of the non-centrosymmetric Weyl semimetal carried out on a $\text{TaAs}$ crystal. This study was among the first STM studies done on a Weyl semimetal and has indeed contributed to the validation of the field. Chapter 4 reports our most recent study (Morali et al., 2019) of the magnetic Weyl semimetal $\text{Co}_3\text{Sn}_2\text{S}_2$, whose novelty stems from showing the first experimental spectroscopic evidence of a time-reversal breaking Weyl semimetal. Together chapters 3-4 embody a unique and timely experimental overview of the key topological aspects of Weyl semimetals.

Finally in chapter 5 I describe in detail the main features of the VT-STM, with corresponding recent results. This chapter also includes a useful technical road map for future STM construction projects.
Chapter 1. Introduction

**Figure 1.1:** Bulk and surface states of a Weyl Semimetal. (A) Dirac to Weyl: Top left - One dirac cone in a material with TR, I and additonal crystal symmetry. Top right - without an additional crystal symmetry, weyl cones annihilate and gap out. Bottom left - Breaking I symmetry, we are left with a minimum of two Weyl pairs. Bottom right - Breaking TR symmetry, we are left with a minimum of one Weyl pair. (B) Fermi arc in a Weyl semimetal connecting the surface projected locations of two opposite charged Weyl nodes. (C) The zeroth Landau levels exhibit chirality with opposite sign velocity. With the application of electric field one can create an imbalance between the charges, effect known as the Chiral anomaly. (adapted from (Zhang et al., 2016))
Chapter 2

Methods

2.1 Scanning Tunneling Microscopy

STM is a tool that is used for imaging and spectroscopy of surfaces at the atomic scale. It was invented in 1982 by Binnig and Rohrer (Binning et al., 1982), and its enormous impact was recognized soon after, awarding its inventors the 1986 Nobel prize. It is paramount for many fields of research, including material and surface science, molecular physics, condensed matter physics and more.

2.1.1 The Tunneling Current

The STM is based on a tunnel junction (Chen, 2007) formed by an atomically sharp conducting tip positioned a very short distance from the surface of the studied sample (Fig. 2.1A). According to the quantum theory of tunneling, application of voltage bias $V$ across the tunnel junction will result in a tunneling current. The tunneling probability amplitude $M$ can be calculated using a model for a metal-vacuum-metal tunneling junction. This model can be solved with the Wentzel–Kramers–Brillouin (WKB) approximation for a barrier of width $s$ and height $\phi$, determined by the values of the work functions of the tip and the sample. Typically $\phi$ is couple of electronvolts and $eV \ll \phi$, so the vacuum barrier can be considered as a square barrier. This scenario has been carefully considered by Bardeen, who argued that under reasonable assumptions on the exponentially small overlap between the wavefunctions (Fig 2.1B), $M$ is virtually independent of energy provided by the voltage bias (Bardeen, 1961).

$$|M|^2 = \exp \left( -2 \frac{\sqrt{2m_e\phi}}{\hbar} s \right) = \exp (-2\kappa s) \quad (2.1)$$

Typically, the exponential factor can be estimated as $\kappa \sim 0.5 \text{ Å}^{-1}$, and the resulting amplitude drops by an order of magnitude for every angstrom. This exponential decay with the tip-sample separation is the key feature that allows the realization of atomic-scale topography. The tunneling current is calculated by using Fermi’s golden rule,
Figure 2.1: (A) Schematic diagram of a tunnel barrier with two metallic electrodes. The electrodes are separated by thin vacuum barrier and biased by voltage $V$. The tunneling current is facilitated by the exponential tails of the wavefunctions. Inset: The STM realization of a tunneling barrier (B) Energy diagram of the respective DOS of the tip (assumed to be flat) and the sample. Only electrons in the range $[E_F, E_F \pm eV]$ contribute to the tunneling current (up to thermal broadening).

Accounting for electron (holes) flow from the filled (empty) states of the sample to the empty (filled) states of the tip, by summing over $\rho_s(E)$ and $\rho_t(E)$ - their respective local density-of-states (LDOS), as depicted in Fig. 2.1B.

\[
I = -\frac{4\pi e}{h} \int_{-eV}^{\infty} |M|^2 \rho_s(E) \rho_t(E + eV) \left[ f(E) \left(1 - f(E + eV)\right) - (1 - f(E)) f(E + eV) \right] dE \quad (2.2)
\]

One can simplify this expression and write the tunneling current as:

\[
I = -\frac{4\pi e}{h} \rho_t(0) |M|^2 \int_0^{eV} \rho_s(E_F - eV + \epsilon) d\epsilon \quad (2.3)
\]

The physical assumptions that justify this simplified form are:

1. The metallic tip is characterized by a flat $\rho_t(E)$ around $E_F$ and can therefore be taken out of the integral.

2. The temperature is low enough so that the Fermi distribution function cuts-off sharply at $E_F$. In practice at a finite temperature $T$ this assumption can be relaxed and accounted for by thermal broadening of the LDOS by a factor of approximately $3.5K_B T$.

3. $|M|^2$ is independent of the energy difference between the tip and the sample.
2.1.2 Modes of operation

STM is a rather versatile tool, which can be used for many purposes. We will briefly describe three common measurement methods which have been used extensively in our study, and are illustrated in Fig. 2.2.

• **Topography** - in this mode, one fixes $V$ and raster the tip across the surface while employing a feedback loop that controls the $z$ piezo to keep a predetermined tunneling current $I_{\text{set}}$ fixed. The voltage applied to the piezo is recorded and translated to units of distance, and due to the exponential dependence of the tunneling current on $z$ it gives a very precise mapping of the surface topography. It should be noted that the actual distance from the surface is a bit arbitrary and depends on $V$ and $I_{\text{set}}$. In fact, even the notion of distance from the surface is a bit vague, because in STM we measure electrons, not atoms. For instance because of the dependence of $I$ in $\rho_s$, spatial variations in electron density might also register in the topography, but since this dependence is in the integrand of Eq. 2.3 and the $z$ dependence is exponential, it’s usually simple to distinguish between the two. Fig. 2.2B shows the topography of the surface of a copper crystal, where the atomic step edges and a screw dislocation can be seen.

• **Scanning tunneling spectroscopy (STS)** - The LDOS as a function of energy can be extracted from the tunneling current by linear expansion of the integrand in Eq. 2.3 and differentiation with respect to $V$ at fixed bias $V_0$:

$$\left. \frac{dI}{dV} \right|_{V_0} \propto \rho_s(E_F - eV_0)$$  \hspace{1cm} (2.4)

In practice, in order to reduce the noise we do not take a numerical derivative, but use a lock-in amplifier to modulate the bias by an AC signal $dV$ around the DC bias $V_0$ and measure the current modulation $dI$ at the same frequency. Importantly, the feedback loop is open at the time of the measurement so no contribution of the tip-sample separation is expected, unless there is a mechanical drift in their relative positions. During the experiment we scan a range of distinct DC voltages, where the energy resolution we aim for is determined by the specific features we want to probe. The result is resolving $\rho_s(E)|_{E=eV}$, and providing a significant input on the electronic structure. Figure 2.2C shows a cusp in the $dI/dV$ that comes from the onset of the copper surface states band on the featureless background of the 3D metal LDOS.

• **Spectroscopic maps** - Instead of taking a single $dI/dV$ curve, one can acquire multiple curves at different $(x,y)$ points, e.g. along some line-cut or on a two-dimensional grid which comprises a $dI/dV$ map or spectroscopic map for each energy - $\rho_s(E; x, y)$. A spectroscopic map can be viewed as stack of sliced 2D or
1D spatial maps, where each slice corresponds to a different energy. In Fig. 2.2D a spectroscopic linecut (1D map) close to an atomic step edge in copper shows the energy dependence of the interference patterns. The spatial structure of the oscillations due to scattering from impurities at a specific energy is presented in a 2D map in Fig. 2.2E. Fourier transforming the spectroscopic map, spatial modulations in the LDOS, also known as quasi-particle interference (QPI) are translated into momentum space, $\mathcal{F}T(\rho(E,x)) = \rho(E,q)$. These reveals information on the momentum transfer $q$ associated with the scattering processes of surface electrons as detailed below.
2.1. Scanning Tunneling Microscopy

Figure 2.3: (A) A low bias topographic image of a CO molecule on Cu(111) surface. Note that at low bias QPI can be seen also in topography. (B) Illustration of the 2D free-electron-like dispersion of copper surface electrons. (C) Fourier transform of the QPI at low bias - the bright ring corresponds to the Fermi surface. (D) Dispersing QPI pattern at a line cut across the CO molecule (along the blue dotted line in (A)) (E) Fourier transform of (D) shows the surface states dispersion $E(q)$. (F) The low bias topographic image of a crystallographic step edge on Cu(111) surface. (G) $dI/dV$ line cut at 200 meV away from the step edge (along dotted line) fitted with appropriate Bessel function. The measured decay is faster due to decoherence. (H) Dispersing QPI pattern at a line cut across step edge (along the dotted line in panel (D)). (I) Fourier transformation of (E) shows the surface states dispersion $E(q)$. Adapted from (Avraham et al., 2018)
2.2 Quasi-particle interference (QPI)

The method of QPI has been used to study a variety of electronic systems including metallic surface states (Crommie, Lutz, and Eigler, 1993; Bürgi et al., 1999), various correlated states as high temperature superconductors (Hoffman et al., 2002a; Vershinin et al., 2004), heavy fermion systems (Aynajian et al., 2012; Schmidt et al., 2010) and charge density waves (Arguello et al., 2015), graphene (Miller et al., 2009; Zhang et al., 2009), and topological electronic systems as topological insulators (Roushan et al., 2009; Alpichshev et al., 2010), Weyl semimetals (Inoue et al., 2016; Batabyal et al., 2016; Morali et al., 2019), and Majorana states (Nadj-Perge et al., 2014). We first demonstrate the application of the QPI method on the well-studied system of the 2D Shockley surface states bound on the (111) surface of Cu single crystals (Crommie, Lutz, and Eigler, 1993). The band structure of these states is known to be fairly free-electron like (Gartland and Slagsvold, 1975). It has a parabolic dispersion,

\[ E(k) = \frac{\hbar^2}{2m^*} k^2 \]

with an isotropic effective mass of \( m^* \sim 0.4m_e \). The bottom of the surface state band lies about 440 meV below \( E_F \). The extremely weak spin-orbit coupling in Cu leaves the surface band practically doubly degenerate (Tamai et al., 2013). We use this electronic system as a case study for demonstrating the principles of QPI method. The LDOS is given by

\[ \rho(E, r) = \sum_k |\psi_k(r)|^2 \delta(E - \epsilon_k) \]  

where \( |\psi_k(r)| \) is the electronic wave function and \( \epsilon_k \) is its energy determined by the dispersion. The typical LDOS \( \rho_0 \) found on Cu(111) surface far from any scatterer is shown in Fig. 2.2A. The broad peak-like feature in the LDOS that sharply onsets 440 meV below \( E_F \) (above the dotted line) is the surface state contribution to the surface projected bulk LDOS.

2.2.1 QPI from Point Defects

We begin with examining the response of the surface electrons to a point-like scatterer located at \( r_o \) and adds a potential \( V(r) = V_0 \delta(r - r_0) \). A typical surface topography taken around such point-like scatterer on the Cu(111) surface is shown in Fig. 2.3A. The location of the scatterer appears in the topography as a local dip which is surrounded by a circular ripple-like pattern. These circular ripples are the interference pattern of the scattered electrons embedded in the LDOS. They testify on the existence of a point scatterer such as a vacancy or an adatom at the origin of the circular waves. The common adatom species on the surface of Cu are CO molecules. These form a

\(^1\)This section is adapted from (Avraham et al., 2018)
potential barrier for the electrons tunneling from tip. Consequently, although topographically the molecule extends out from the surface, in order to maintain constant tunneling current under constant bias, the tip moves closer to the surface. The CO molecule is accordingly registered as a topographic intrusion rather than an extrusion. In presence of disorder such as the CO molecule, other impurities or crystal defects, translational invariance is broken and elastic scattering processes mix equal-energy states of momenta \( k \) and \( k' \), corresponding to the incoming and outgoing momenta of the scattered electron. The standing wave resulting from the scattering process will be seen in the LDOS map as an interference pattern with a wavevector \( q = k - k' \), as illustrated in Fig. 2.3B. Fourier transform of the spectroscopic map will thus reveal the underlying scattering processes and the available phase-space volume for their occurrence. This in turn can be used to reconstruct the dispersion and the structure of the Fermi surface or any other constant energy surface. This is illustrated in Fig 2.3C that shows the Fourier transform of the low bias topography (Fig. 2.3A), therefore capturing the circular Fermi surface (corresponding the contour of constant energy at zero bias) of Cu(111) surface states.

By measuring a \( dI/dV \) line cut across the impurity as a function of bias voltage one can directly image the evolution of these standing waves as a function of energy. The energy dispersion of the standing waves obtained around the impurity is shown in Fig. 2.3D. It was measured along a line cut that crosses the impurity as marked by the blue line in Fig. 2.3A. Note that the wavelength of these standing waves increases upon decreasing the energy until it diverges at the bottom of the surface states band, approximately at -440 meV. This dispersing wavelength corresponds to the momentum transfer wave vector \( q = k - k' \), which in Cu(111) is dominated by the backscattering channel \( k = -k' \), implying \( q = 2k \). At the Fermi energy this momentum transfer equals \( q = 2k_F \). This can be obtained by solving explicitly the scattering problem with cylindrical symmetry imposed by the scatterer, which to lowest order yields:

\[
\rho(r, E) = \rho_0 \left( 1 + \frac{2}{\pi k_F r} \left( \cos^2 \left( k_F r - \frac{\pi}{4} + \eta_0 \right) - \cos^2 \left( k_F r - \frac{\pi}{4} \right) \right) \right) \quad (2.6)
\]

where \( \eta_0 \) is the phase shift affected by the detailed scattering potential (Crommie, Lutz, and Eigler, 1993). For a \( \delta \)-like potential with \( \eta_0 = \pi/2 \) the modulated LDOS reduces to:

\[
\rho(r, E) = \rho_0 \left( 1 - \frac{2 \sin (2k_F r)}{\pi k_F r} \right) \quad (2.7)
\]

The \( q = 2k_F \) oscillatory modulation becomes apparent, as does the radial attenuation in the amplitude of the oscillations. This suppression originates from the spreading out of the standing wave pattern over the circumference that grows as \( r \). The quantization
of the dispersing band seen in 2.3D results from the presence of an additional adjacent scatter (As can be seen for example in Fig 2.2E).

### 2.2.2 QPI from Step Edges

Crystallographic step edges, as the one shown by the topography in Fig. 2.3F presents another type of scattering source with different geometry than the one presented by a point impurity. The atomically ordered step edge yields a translational invariant scattering potential along the step edge, \( V(r) = V_0 \Theta(x) \), where \( x \) is the distance from the step edge. As a result the impinging electrons undergo a unidirectional scattering perpendicular to the step edge such that \( k_x = k'_x + q_x \) and \( k_y = k'_y \). In a material with isotropic dispersion like Cu, summing over these scattering processes yields a LDOS given by

\[
\rho(r, E) = \rho_0 \left( 1 - J_0(2k_F x) \right) \tag{2.8}
\]

where \( J_0 \) is the zero-order Bessel function which asymptotically goes as \( \cos(qx)/\sqrt{qx} \) (Crommie, Lutz, and Eigler, 1993). The resulting standing wave pattern along a line cut normal to the step edge (blue line in Fig. 2.3F) is shown in Fig. 2.3H. Its Fourier transform, presented in Fig. 2.3I, shows the energy dispersion \( E(q_x) \) which remarkably agrees with the \( E(q) \) obtained from scattering off a point impurity (Fig. 2.3E).

Finally, Fig. 2.3G presents a single energy cut of the standing wave pattern taken at 200 meV (dotted line). The expected oscillatory behavior with \( x^{-1/2} \) decay is fitted to it (solid line). It becomes evident that the decay of the oscillation amplitude is stronger than that predicted. The reason for that is decoherence of the scattered electrons away from the step edge. The injected hot electrons lose their phase coherence due to interaction with the environment which facilitates their relaxation. These can be either interactions with the cold electrons occupying the Fermi sea or with phonons. In any case it leads to an exponential suppression of the standing wave amplitude as a function of the distance from the step edge

\[
\rho(r, E) = \rho_0 \left( 1 - e^{-x/L_{\phi}} J_0(2k_F x) \right) \tag{2.9}
\]

over a length scale defined as the phase coherence length \( L_{\phi} \) (Bürgi et al., 1999).

The above examples demonstrate that measuring the interference pattern electrons embedded in the LDOS as they scatter off various crystallographic irregularities allows one to study the electronic dispersion, scattering processes, and the phase decoherence of the scattered electrons.
2.3 Sample preparation

2.3.1 Calibrating Field of View

Single crystals of TaAs and Co$_3$Sn$_2$S$_2$ were grown by our collaborators at the group of Claudia Felser. These samples vary in size, but are typically about 1 mm$^2$, therefore pose a challenge to approach with a STM tip. Due to the limited scan range, a slight misplacement of the crystal on top of the sample holder stage may result in a failed approach, as illustrated in Fig. 2.4A. In order to tackle this issue, we had to properly calibrate the maximum STM scan range with our sample holders. To that end, we have mounted a large Au sample (25 mm$^2$) and poked holes using the STM tip in a predefined pattern (Fig. 2.4B). The sample was then imaged using an optical microscope (Fig. 2.4C-D), which easily resolved the pattern formed. The calibrated scan window allows for a precise positioning of samples, which accounts for the inherit XY offset.
2.3.2 Single crystal cleaving

Before introducing the sample to the STM, a freshly cleaved surface needs to be exposed *in-situ*. Accordingly, we have glued a 10 mm long aluminum post, of 3 mm diameter using silver epoxy (Epo-Tek) on top of the crystal surface (Fig. 2.5A). The sample assembly was then transferred to a cold stage, and was cooled to 80K (Fig. 2.5B). Subsequently, using a push-pull manipulator a fast blow is applied to the post transferring a torque to the crystal. Finally, observing (Fig. 2.5C) both the post and the sample allows us to determine whether the cleave is successful. In such event, the sample is quickly transferred to the STM.
Chapter 3

Non-centrosymmetric Weyl Semimetal

3.1 Introduction

Topological states of matter harbor strikingly unique boundary states, such as the chiral edges of the quantum Hall effect (Wen, 1990), the surface states of topological insulators (Hasan and Kane, 2010; Qi and Zhang, 2011), and the Majorana end modes of topological superconductors. The properties of these surface states, such as gapless surface spectrum, relativistic dynamics, and evasion of localization by disorder, are determined by the topological nature of the bulk and are protected by the energy gap in the bulk’s spectrum. These states cannot be realized as stand-alone systems without the coupling to the topological bulk. Surprisingly, these states exist even on two-dimensional surfaces of three-dimensional Weyl semimetals (Wan et al., 2011; Xu et al., 2011; Burkov and Balents, 2011; Weng et al., 2015; Huang et al., 2015a; Sun, Wu, and Yan, 2015), despite the absence of a bulk energy gap (Murakami, 2007). The defining characteristic of these states is the Fermi arcs, which may not be realized on stand-alone two-dimensional systems. Whereas in two-dimensional systems, lines of constant energy must form closed contours in momentum space, Fermi arcs are open contours that emanate and end in states associated with bulk Dirac cones whose nodes are termed Weyl nodes. Here, we use scanning tunneling microscopy (STM) and spectroscopy to study surface states in the Weyl semimetal tantalum arsenide (TaAs). Bulk and surface band structures of TaAs have been modeled (Weng et al., 2015; Huang et al., 2015a; Sun, Wu, and Yan, 2015) and mapped by photoemission spectroscopy (Xu et al., 2015a; Lv et al., 2015b; Yang et al., 2015), and its unique electrodynamics (Vazifeh and Franz, 2013; Son and Spivak, 2013; Potter, Kimchi, and Vishwanath, 2014; Baum et al., 2015; Baireuther et al., 2016) have been probed in magnetotransport (Zhang et al., 2016; Huang et al., 2015b). Previous STM studies of this material have identified scattering processes among the non topological surface bands (Zheng et al., 2016) and between these surface bands and a Fermi arc band, where absence of other scattering processes involving Fermi arc states was attributed to their connectivity with topological bulk bands (Inoue et al., 2016). Here, we report an exhaustive visualization of
the scattering processes available among the different surface bands, including intra-
arc scatterings. Processes involving Fermi arc states are identified by their distinctive
attributes and real-space structure. This provides a comprehensive characterization
of the unusual properties of the Fermi arc states and their unique correspondence to
topological bulk bands.

High-quality single crystals of TaAs (see appendix A.1) were cold-cleaved at 80K
under ultrahigh vacuum, exposing a fresh (001) surface that was measured at 4.2K.
QPI patterns that elastically scattered electrons embed in the local density of states
were measured in differential conductance ($dI/dV$) mappings. We reveal four distinct
attributes of the Fermi arcs by measuring different aspects of their scattering processes:
(i) their relatively isotropic QPI profile, revealed by scattering off atomic vacancies (section 3.2.1) (Chang et al., 2016; Belopolski et al., 2016; Kourtis et al., 2016; Mitchell and Fritz, 2016); (ii) the linear energy dispersion and its relation with bulk Weyl nodes, by
scattering off a crystallographic step edge (section 3.2.2); (iii) their localization on the
Ta layer, by tracing the spatial origin of their QPI patterns with subatomic resolution
(section 3.2.2); and (iv) the weak coupling to the surface atomic structure, as opposed
to the strongly coupled trivial states, deduced from the manifestation of their wave
function structure in the QPI pattern (section 3.2.4).

3.2 Results

3.2.1 Quasiparticle interference of trivial bands and Fermi arcs in TaAs

The As terminated surface topography shown in Fig. 3.1A hosts naturally occurring
vacancies which serve as excellent scattering centers for electrons. Indeed a differential
conductance ($dI/dV$) map taken at fixed energy (Fig. 3.1B) shows clear quasiparticle
interference (QPI) patterns around such vacancies. Fourier decomposition of the QPI
pattern reveals several scattering processes. We recognize three prominent QPI pat-
tterns: ellipse shaped patterns around $G_0$ and $G_{\pm Y}$, half of bowtie-shaped patterns at
$G_{\pm X}$, and portions of rounded square-shaped patterns on the four corners of the central
zone (dashed lines). These are indeed in good agreement with spin selective probabil-
ity (SSP) calculation, based on the DFT calculated Fermi surface shown in Fig. 3.1D,E
respectively. They originate from dangling bonds and hence are topologically trivial
(Zheng et al., 2016). Careful observation in the region near $G_0$ reveals two leaf-like
features that extrude beyond the ellipse which we identify to result from the Fermi arc
(Q1 in Fig. 3.1F).
3.2. Results

Figure 3.1: QPI study of As terminated surface. (A) (001) topography of TaAs, inset showing atomic resolution. (B) dI/dV map taken at Fermi energy shows clear elongated QPI patterns around As vacancies. (C) Fourier transform of B. (D) SSP calculations of the first BZ in q space. (E) DFT calculation of the Fermi surface, arrows represent the scattering processes we observe. (F) Blown up comparison of the QPI signal around G₀ (left), and an SSP calculation involving the ellipse and arc, shows evidence of scattering process involving the Fermi arc. (G) Measured dI/dV spectrum over a broad energy range (solid) shows nice agreement with DFT calculation of As termination (dashed).

3.2.2 Bulk-Boundary correspondence

We further measured the dispersion of the Fermi-arcs by investigating the interference patterns from a crystallographic step edge that is oriented 49° with respect to the crystal axis (Fig. 3.2A). This experimental setup allows us to probe QPI patterns along the G₀ – GM direction. Measuring a dI/dV line cut normal to the step indeed shows clear dispersing modulations (Fig. 3.2C), which reveals several bands upon applying Fourier transform (Fig. 3.2D). Once again we turn to the SSP calculation and by comparing find the dispersing bands of the ellipse and square like features (blue and green arrows respectively in Fig. 3.2E). We further identify an additional band (Q2 marked in red arrow), which reflects scattering between two neighboring Fermi arcs. Its dispersion indeed agrees with the shrinkage of the Fermi arcs upon increasing of the energy, which corresponds to an average velocity of ~ 10^5 m/s per arc. The measured momentum transfer at the W₂ energy (E = 2meV) is 5.4 ± 0.1nm⁻¹ which equals to the inter-Weyl node separation. This quantitatively demonstrates the correspondence between the surface Fermi arc and the bulk Weyl node location in momentum space.

We examined the spatial distribution of the topological and trivial bands with respect to the topmost As layer. We decomposed the linecut to reflect signal arising
Figure 3.2: Fermi arcs on Ta and trivial bands on As. (A) Step edge topography oriented 49° with respect to the crystal axis. (B) The $dl/dV$ line cut probes scattering events approximately along the $G_0 - G_M$ line. (C) $dl/dV$ line cut as taken along the dashed line in a. Inset shows the topography as recorded while taking the measurement. (D) Fourier transform of C. (E) SSP energy dispersion along the dashed line in B. (F,G) As and Ta sites contribution from the line cut shown in C. (H,I) Fourier transform of F and G respectively. Inset shows the spatial distribution of the ellipse (H) and Fermi arc (I) wavefunction. (J) SSP of Fermi arcs only (left) and their extracted dispersive envelope (right).

from As sites (Fig. 3.2F) and Ta sites (Fig. 3.2G). Amazingly, the Fourier transform of these two sub-maps gives further insight to the scattering processes. The As sub-map Fourier transform (Fig. 3.2H) reproduces the previously seen ellipse dispersion. This is consistent with our understanding of the dangling bond origin of these states. In contrast, on the Ta layer (Fig. 3.2I) we observe two opposite V-shaped curves (Q3 red arrows) that agree with the SSP of intra-Fermi arc processes (Fig. 3.2J). Fascinatingly, the upper dispersing branch extrapolates to the energy of the $W_2$ Weyl node, again demonstrating the bulk-surface correspondence. Note the topological bulk Weyl cones are derived from the Ta atomic states (Inoue et al., 2016). Our measurements uniquely show that the Fermi-arc connecting these cones share this property.

3.2.3 Structure of the surface wave function

We now show that the Fermi arc bands differ from non topological bands, also in their structure, parallel to the surface within the unit cell. Figure 3.3 shows QPI maps at
three different energies (right panels), alongside $dI/dV$ maps in a vacancy-free region (left panels). At -300 meV (below Fermi energy), the vacancy-free $dI/dV$ map and the corresponding QPI map are approximately symmetric to 90° rotations, and the QPI patterns are concentrated around $G_0$. In contrast, at 85 meV, 130 meV, and the Fermi energy (Fig. 3.1B, inset), the vacancy-free $dI/dV$ shows a clear chain structure that changes its crystallographic orientation with energy. The QPI features at corresponding energies are strongly replicated along that modulation direction. Modulation in a vacancy-free region ought to be attributed to the structure of the wave function.

**Figure 3.3:** Correlation between modulations and replications set by the electronic wave function. (A to C) Left panels: $dI/dV$ in a vacancy-free region at three representative energies shows strong modulation, whose strength and orientation change with energy. The bars represent the Bragg peak intensities, $|A_{G_x,G_y}|^2$, along the two crystallographic directions. Insets: DFT calculation of the local charge density that captures a similar modulation. Right panels: Fourier analysis of extended $dI/dV$ maps in the presence of vacancies at the corresponding energies. The intensity and anisotropy of the replications of QPI features are correlated with the modulation detected in the vacancy-free region.
3.2.4 Topological Bloch wave function

The surface electronic wave function contains a background of density of states (DOS) modulations, commensurate with the lattice structure (inset Fig. 3.2C), including in vacancy free regions (inset Fig. 3.1B). This empirical fact suggests these modulations owe their existence to the structure of the wave function rather than the details of the scatterer. Motivated by these findings we turn to explore the Bloch wavefunctions of the trivial and topological states.

The Bloch theorem constrains a state with a crystal momentum \( k \) to be a superposition of momenta \( k + G \), where \( G \) is the reciprocal lattice vector,

\[
\Psi_k(r) = \sum_G C_G^k \exp(i(k + G) \cdot r).
\]

Consequently, the local DOS in a vacancy-free region becomes

\[
\sum_g A_g \exp(ig \cdot r),
\]

where

\[
A_g = \sum_{G,k} C_G^{*k} C_G^k \delta(E - E_k)
\]

is the amplitude of the Bragg peak that corresponds to \( g = G - G' \), \( r \) is the position, \( E \) is the energy, and \( E_k \) is the energy of the state with momentum \( k \). Namely, given a coefficient distribution with more than one dominant coefficient and thus a non zero \( g \) we expect to get modulation in the DOS along the \( g \) direction. With the introduction of a scattering potential \( V_q \) an electron may scatter between \( \Psi_k \) and \( \Psi_{k'} \) provided the momentum transfer satisfies \( q_g = k - k' + g \) with probability proportional to

\[
\sum_{G'} V_{q_g} C_G^{*k} C_{G'}^{k'}
\]

(Briner et al., 1998).

Fig. 3.4A,B lists the Bloch coefficients distribution of the ellipse and Fermi arc band as calculated deterministically by DFT. Amazingly, the coefficients distribution of the ellipse band agrees well with our experimental results (Fig. 3.1C), namely predicts the highest ellipse signal around the \( G_\pm Y \) bragg peak which diminishes beyond the bragg peak, a high signal around \( G_0 \) and a low signal around \( G_\pm X \). In contrast, the Fermi arc band contains one dominant coefficient, its topological wave function is rather uniform within the unit cell and resembles a plane wave. Ergo, the topologically derived Fermi arc states are fairly oblivious to the surface potential. We may introduce this physical understanding to the following analysis method. Fig. 3.4C describes the result of subtracting the QPI signal around \( G_0 \) from that of \( G_\pm Y \). Comparing to the SSP (Fig. 3.4D) we see that indeed this method succeeds in removing the replicated trivial ellipse band, while leaving the topological Fermi arcs unhindered. Moreover, it further reveals the previously unseen scattering processes (Q4) involving the Fermi arcs along the \( \Gamma - X \) direction.

3.3 Discussion

The topological nature of Weyl semimetals is manifested by the bulk Weyl nodes, their Berry flux, and the essential surface Fermi arcs that accompany them. The correspondence between the bulk and surface states gives rise to various physical phenomena
3.3. Discussion

**Figure 3.4:** Bloch-based QPI arithmetics. (A,B) Coefficients distribution of the ellipse (A) and Fermi arc (B) band. Arrows thickness represent the probability of each scattering process. (C) Subtraction of QPI signal around $G_0$ from that of $G_{\pm Y}$. $\alpha = 1.14$ is the ratio between extracted intensities of ellipse. (D) SSP of Fermi arcs alone. Inset showing the contributing scattering processes.

that characterize the topological semimetals and their unique electrodynamics (Vazifeh and Franz, 2013; Son and Spivak, 2013; Potter, Kimchi, and Vishwanath, 2014; Baum et al., 2015; Baireuther et al., 2016). However, in real systems, there are also non topological surface states that overlap in space and energy with the topological Fermi arcs. These states, which may originate, for instance, from dangling bonds, are ubiquitous. Their effects on phenomena that involve the Fermi arcs, such as the cyclotron frequency of cyclotron orbits that connect opposite surfaces, are not determined by topological considerations alone; rather, it is affected by the combined energy-momentum dispersion of both types of states, by the wave functions of both types of states, and by impurity induced scattering between the two types of states that we visualize.

The measurements we report here provide information on the interplay between the Fermi arc states and the non topological ones as well as on their correspondence with the bulk Weyl nodes. We visualized scattering processes among the Fermi arc surface bands, processes that scatter Fermi arc states to trivial states, and processes that scatter between trivial states. The two processes that involve only topological states were found to be correlated with the energy-momentum location of the bulk Weyl nodes. The intra-arc scattering channel (Fig. 3.2, I and J) extrapolates to the momentum separation of a Weyl pair, whereas the momentum transfer of the inter-arc scattering channel (Fig. 3.2, D and E) entails the momentum separation between Weyl...
nodes of adjacent pairs. We stress that this quantitative correspondence between the topologically classified bulk dispersion and the momentum extent of the Fermi arcs is unique to semimetallic topology classes. All previously studied topological electronic phases have a gapped bulk spectrum, which is thus spectrally featureless. Bulk-surface correspondence is also evident by the structure of the Fermi arc wave function that resides predominantly on the subjacent Ta sites, from which the bulk Weyl cones are also derived (Inoue et al., 2016).

We further showed that the lateral spatial structure of the Fermi arc wave functions within the unit cell is rather uniform and resembles a plane wave. It stands in stark contrast to the intricate structure of the non topological surface bands, as captured by their strongly replicated QPI patterns. This observation demonstrates that the topologically derived Fermi arc states are fairly oblivious to the surface potential, which is a property that is not shared by the non topological ones. The method of analysis that we developed and implemented, in which the replicated structure of QPI patterns is used to separate overlapping features in the pattern, will have further applicability in future studies of Fermi arcs in Weyl semimetals and in other electronic systems. Many topological surface states in different materials did not exhibit any clear replications in their QPI signatures (Roushan et al., 2009; Beidenkopf et al., 2011; Soumyanarayanan et al., 2013), possibly signifying their surface resilience. A counter example that calls for a closer examination is that of topological crystalline insulators whose Dirac surface states’ QPI signatures were found to be replicated (Zeljkovic et al., 2014). Strongly correlated electronic systems may also be probed in a similar fashion. For instance, QPI patterns in high-temperature superconductors (Hoffman et al., 2002b; Dalla Torre, He, and Demler, 2016), in which charge order has been recently reported, also exhibit replications. It would be enticing to apply our method of analysis to characterize the structure of the Bloch wave functions in such systems and to possibly unveil hidden spectroscopic features. On a yet broader scope, our resolution of the detailed structure of the Bloch wave function in local density of states and QPI measurements suggests that it will further affect other physical processes that involve quantum electronic interference. Among these are Friedel oscillations and their signature in transport, and surface state–mediated RKKY interactions. The role of the structure of the Bloch wave function in these processes calls for further theoretical elucidation alongside experimental verification.
Chapter 4

Magnetic Weyl Semimetal

4.1 Introduction

In topological semimetals, the dispersion of the topological surface bands is correlated with the topological bulk bands dispersion (Wan et al., 2011; Burkov and Balents, 2011; Burkov, Hook, and Balents, 2011; Young et al., 2012; Wang et al., 2012; Weng et al., 2015; Batabyal et al., 2016; Inoue et al., 2016). Examples include Weyl and Dirac semimetals whose nontrivial topological properties arise from the existence of band-touching points, termed Weyl or Dirac nodes, in the electronic bulk band structure. As presented in chapter 1, bulk Weyl nodes are formed under broken inversion or time reversal symmetry. They exhibit definite chirality and are associated with open-contour “Fermi-arc” surface bands that emanate from one Weyl node and terminate at another with opposite chirality within the surface 2D momentum space. Owing to this surface-bulk correspondence, the dispersion of the Fermi-arcs reflects the Weyl cone band structure and particularly the bulk Weyl nodes energy. The Berry curvature associated with Weyl and Dirac nodes has been shown to result in a chiral anomaly in magneto-transport (Son and Spivak, 2013; Huang et al., 2015a; Zhang et al., 2016) and various non-local transport effects (Parameswaran et al., 2014; Baum et al., 2015).

The formation of Fermi-arcs is guaranteed by the existence of bulk Weyl nodes, and therefore provides a direct way to classify the bulk topology via surface probes. Nevertheless, some of the properties of Fermi arcs, such as their momentum space contour and their connectivity among the Weyl nodes, are not predetermined by the bulk distribution of Weyl nodes but rather by the details of the surface termination (Sun, Wu, and Yan, 2015; Souma et al., 2016). These properties, which can be controlled by surface manipulation, have direct implications for the magnetoelectric dynamics of the Weyl electrons that combine bulk and surface conduction, such as quantum oscillations (Potter, Kimchi, and Vishwanath, 2014; Moll et al., 2016). Nevertheless, the level of susceptibility of the Fermi-arc bands to varying surface potentials has not been studied thoroughly in experiment.
Here we study the compound \(\text{Co}_3\text{Sn}_2\text{S}_2\) that was classified recently as a candidate time reversal symmetry broken Weyl semimetal (Liu et al., 2018; Xu et al., 2018). In contrast to many material candidates (Weng et al., 2015; Sun et al., 2015) and several experimental realizations (Yang et al., 2015; Xu et al., 2015a; Lv et al., 2015a; Zheng et al., 2017) of the inversion symmetry broken Weyl semimetals, time reversal symmetry broken Weyl semimetals were predicted in only a few magnetically ordered material candidates, including \(\text{GdPtBi}\) (Hirschberger et al., 2016), \(\text{Y}_2\text{Ir}_2\text{O}_7\) (Wan et al., 2011), \(\text{HgCr}_2\text{Se}_4\) (Xu et al., 2011), and certain \(\text{Co}_2\)-based Heusler compounds (Kushwaha et al., 2018). In a few, including \(\text{Co}_3\text{Sn}_2\text{S}_2\) (Liu et al., 2018; Muechler et al., 2017), a large anomalous Hall conductivity was reported (Suzuki et al., 2016), an indication of a magnetic Weyl phase; however, a spectroscopic verification remains challenging.

In \(\text{Co}_3\text{Sn}_2\text{S}_2\) (Fig. 4.1A), the magnetic properties arise from the kagome-lattice cobalt (Co) planes (Yin et al., 2018), whose magnetic moments order ferromagnetically out of plane below 175 K (Weihrich and Anusca, 2006; Liu et al., 2018). These Co planes are interleaved with buffer planes of triangularly ordered tin (Sn) and sulfur (S). Ab-initio calculations (Liu et al., 2018; Xu et al., 2018) find six Weyl nodes in the bulk Brillouin zone (Fig. 4.1B). Their projection on the (001) surface identifies three Fermi-arc bands that connect the six surface projected Weyl nodes. We show that the Fermi arc connectivity in \(\text{Co}_3\text{Sn}_2\text{S}_2\) varies with the surface termination, which also affects the intricate Weyl semimetal magnetoelectric response (Fig. 4.1C).

### 4.2 Results

#### 4.2.1 Diverse surface band structure

The layered crystal structure of \(\text{Co}_3\text{Sn}_2\text{S}_2\) enabled us to study spectroscopically all three terminations of the (001) surface. Single crystals of \(\text{Co}_3\text{Sn}_2\text{S}_2\) were cold cleaved at 80 K under ultrahigh vacuum conditions, and measured at 4.2 K. The energetically favorable cleave plane is between the Sn and the S monolayers (Fig. 4.1A). Indeed, most of the cleaved surface exhibits a triangular atomic structure, indicative of the Sn or S terminations, shown in Fig. 4.1C and D, respectively. Rarely, however, we detect the less probable Co termination, hallmarked by the characteristic Kagome crystal structure as shown in Fig. 4.1E. Each termination imposes a distinct surface potential that results in a distinct surface band structure. This diversity is captured by the characteristic \(dI/dV\) spectra we find on the Sn, S and Co terminated surfaces, shown in Fig. 4.1F, G and H, respectively.
4.2. Results

**Figure 4.1:** Diverse surface band structure. (A) The layered crystal structure with Co moments (arrows) ordered ferromagnetically. (B) Bulk Brillouin zone hosting three pairs of Weyl nodes and their (001) surface projection. (C) Atomically resolved Sn, S and Co surfaces, showing triangular, triangular and kagome crystal structure, respectively. Insets show atomic lattice sites indicated by black dots. (F-H) Typical $dI/dV$ spectra on the different terminations.

4.2.2 Fermi arc connectivity and time reversal symmetry breaking

To visualize the Fermi-arcs and explore their energy evolution on the various surface terminations we carried out measurements of the QPI patterns that scattered electrons embed in the local DOS. Topographic image of the Sn terminated surface decorated by moderate concentration of adatoms is shown in Fig. 4.2A. The QPI patterns on the Sn surface appear as weak ripple modulations in the corresponding $dI/dV$ map (Fig. 4.2B). We trace these QPI to originate from subsurface impurities that appear fainter in Fig. 4.2A.

The QPI patterns on the Sn surface assume particularly sharp polygon shapes. Representative QPI maps taken at 7.5 meV (Fig. 4.2C) and at -10 meV (Fig. 4.2D) capture the main QPI patterns with scattering processes that involve the Fermi-arc bands. The QPI patterns at 7.5 meV (Fig. 4.2C) consist of a hexagon shape QPI around $q = 0$ and replications of it centered on Bragg peaks. To associate these QPI patterns with particular scattering processes, we consider the calculated surface density of states $DOS(k)$ of the Sn termination, plotted in Fig. 4.2E along with a simplified schematic diagram of it (left and right panels, respectively). The observed hexagonal QPI pattern originates from scattering processes (pink arrow in Fig. 4.2E) between a hexagonal electron pocket around $\Gamma$ and the adjacent edges of triangular electron pockets at the K and K’ corners of the Brillouin zone. The edges of the triangular pockets around K are open-contour Fermi-arc bands, each of which connects a pair of Weyl cones within the
Brillouin zone. The hexagonal QPI patterns therefore originate both from scattering among surface states as well as from scattering between surface to Fermi-arcs states.

With decreasing energy the hexagonal QPI patterns increase in size and towards -10 meV an additional QPI pattern appears in the form of straight lines along $\Gamma$-M that connect the adjacent corners of the QPI hexagons (Fig. 4.2D). Indeed we find that the additional connecting lines in the QPI pattern originate from quasi-nesting conditions, marked by the vertical pink arrow in Fig. 4.2F, brought about by approximate co-linearity of edges of adjacent triangular pockets at K and $K'$. These scattering processes necessarily involve Fermi-arc bands. The shape, size and orientation of the connecting line QPI patterns indicate that the Fermi-arcs involved in these scattering process connect between pairs of adjacent Weyl nodes within the Brillouin zone.

The full energy evolution of the QPI patterns we find on the Sn surface is given by the momentum cut in Fig. 4.2G. The most prominent dispersing line (left pointing pink arrow), which curves towards $q = 0$ with increasing energy, represents the hexagonal QPI patterns whose size shrinks with energy. As shown in Fig. 4.2E, the size of the hexagonal QPI is determined by the scattering vector between the edges of the hexagonal and triangular pockets. These pockets expand with energy (shown Fig. 4.2H), hence the inter pockets scattering vector and its associated hexagonal QPI decrease with energy.

Close inspection further reveals that, around zero bias, an inner fainter scattering mode appears along the $\Gamma - K$ direction. It is hardly visible since it overlaps with the central $q = 0$ broad peak that originates from inevitable long-wavelength inhomogeneities in the $dI/dV$ map. To recover it we use the fact that these inhomogeneities are isotropic (see central disc shapes in Fig. 4.2C and D) thus rendering the broad $q = 0$ peak symmetric along $\Gamma$-K and $\Gamma$-M. In the inset to Fig. 4.2G we subtract the $\Gamma$-M QPI cut from the $\Gamma$-K one. This subtraction completely eliminates the $q = 0$ peak, which allows us now to resolve two dispersing scattering modes. We naturally attribute this doubled mode to scattering processes from the edge of the hexagonal pocket at $\Gamma$ to the near edge of the triangular pockets at both K and $K'$. The distinct extent in momentum space of the triangular pockets is a direct manifestation of broken time reversal symmetry and the appearance of the second inner QPI mode is a direct measure of it. These signatures of the time reversal symmetry broken surface band structure and the Fermi-arcs it hosts on the Sn termination provide striking experimental confirmation for the existence of a magnetic Weyl semimetal phase in $Co_3Sn_2S_2$. 


4.2. Results

Figure 4.2: Intra Brillouin zone Fermi arc connectivity and time reversal breaking on the Sn termination. (A) Topographic image of Sn terminated (001) surface ($V_{\text{bias}} = 95\text{meV}$, $I_{\text{set}} = 175\text{pA}$) featuring adatoms and subsurface impurities. (B) $dI/dV$ measurement taken on the region shown in A at $V_{\text{bias}} = 7.5\text{meV}$ ($V_{\text{AC}} = 2.1\text{meV}$ RMS, $f = 733\text{Hz}$) showing clear interference patterns. (C-D) Fourier transform of two $dI/dV$ maps taken at different energies showing sharp QPI patterns. (E-F) DFT calculated DOS($k$) and a simplified schematic diagram highlighting the important bands and scattering processes among them involving Fermi arc or surface bands (pink or red arrows, respectively). (G) Energy-Momentum cut of the QPI along K-Γ-M directions. Identified scattering processes are marked with arrows. The inset shows background eliminated two parallel modes obtained by subtracting the Γ-M cut from the Γ-K cut. (H) Energy-momentum cut of the calculated DOS along K-Γ-K’ capturing the energy evolution of the various scattering processes marked by arrows.
4.2.3 Inter Brillouin zone Fermi arc connectivity

We now show that the Co terminated surface exhibits distinct configuration of Fermi-arcs with different connectivity than the one observed on the Sn terminated surface. A topographic image of a Co terminated surface along with its corresponding $dI/dV$ map are shown in Fig. 4.3A and B, respectively. The corresponding QPI linecut taken along the high symmetry line K-$\Gamma$-M (Fig. 4.3C) presents a rich QPI pattern with replications that span up to the second order Bragg peaks. To recognize the main scattering processes, in particular the ones involving Fermi-arcs, we first identify the three main regimes in the energy evolution of the band structure. At low energies (Fig. 4.3D) we find again six closed electron pockets around the Brillouin zone corners at K and K'. Upon increasing the energy, all pockets widen until the three pockets around the K points, disconnect and flatten (Fig. 4.3E), revealing the Fermi-arc connectivity. In contrast to the Weyl node connectivity observed on the Sn termination, where the Fermi-arcs connect a pair of Weyl nodes within the same Brillouin zone, here the Fermi-arcs connect a pair of Weyl nodes from adjacent Brillouin zones. Upon further increase in energy the surface pockets around K' split by hybridizing with the Fermi-arc bands around K, forming C-shaped bands that are partially composed of Fermi-arc states (Fig. 4.3F).

These three regimes of pocket structures correspond to three regimes we identify in the evolution of the QPI patterns in Fig. 4.3C: At high energies (about 50-80 meV) we find a single non dispersing mode (Fig. 4.3G and H); At intermediate energies (about 20-50 meV) we find a single slightly dispersing mode; at low energies (-10-20 meV) we find several dispersing QPI patterns (Fig. 4.3I and J). Identifying this energy evolution together with theoretical calculations of the joint density of states (JDOS) allowed us to identify the scattering processes that give rise to the QPI patterns that are marked in dotted lines and arrows of corresponding color in Fig. 4.3C and D-F, respectively. Among the scattering processes we have identified we concentrate here on those which involve Fermi-arcs states (pink arrows), and bands which evolve with energy into Fermi-arcs. The high energy scattering between the C-shaped bands generates the slightly dispersing high intensity QPI peaks in Fig. 4.3G, as well as in the corresponding JDOS (circled in both). The low energy scattering within the K pocket (pink arrow in Fig. 4.3D) that splits to Fermi-arc bands at slightly higher energies is identified with the diamond-like QPI pattern replicated beyond the Bragg peak in Fig. 4.3J (pink arrow). The QPI patterns we detect on the Co terminated surface differs dramatically from those we observed on the Sn termination. This allowed us to deduce a distinct surface band structure with distinct shape of Fermi-arcs and Weyl nodes connectivity.
4.2. Results

**Figure 4.3:** Inter Brillouin zone Fermi arc connectivity on the Co surface. (A) Topographic image of Co terminated (001) surface ($V_{bias} = -95\,meV$, $I_{set} = 250\,pA$) featuring adatoms and sub-surface impurities. (B) $dI/dV$ measurement taken on the region shown in A at $V_{bias} = -5\,meV$ ($V_{AC} = 2.1\,meV$ RMS, $f = 733\,Hz$). (C) Energy-momentum cut of the QPI along K-Γ-M direction. Dispersing scattering peaks are marked by dotted lines. Those that involve Fermi-arc bands appear in pink. (D-F) Representative calculated DOS($k$) at different energies, signifying three major regimes in the energy evolution of the main electron pockets in the band structure. Identified scattering processes among them are marked in arrows with corresponding colors to the dotted lines in C. (G) Fourier transform of two $dI/dV$ maps taken at relatively high energies, showing dispersive QPI broad peaks along Γ-M. (H) Calculated JDOS at the respective energy. (I) Calculated JDOS at low energy. (J) Fourier transform of two $dI/dV$ maps taken at relatively low energies showing rich dispersing QPI patterns.
4.2.4 Weyl cone dispersion on the S termination

The S terminated surface band structure presents a third distinct case. In contrast to gapped systems where the topological surface states are protected by the bulk gap, the protection of the surface Fermi-arcs is more subtle. To be protected from scattering into the bulk, the contour of the Fermi-arcs should lie within the Weyl cone’s bulk gap. The local surface potential, however, is able to push the Fermi-arcs contour out of that gap and hence to lift their protection from scattering into the bulk. This is the case for the S terminated surface for which the Fermi-arcs overlap with surface projected bulk bands at almost all energies and momenta away from the Weyl node energy (Xu et al., 2018). Still, the S terminated surface, whose topographic landscape is dominated by vacancies (Fig. 4.4A) exhibits clear signatures of electronic interference in the $dI/dV$ mappings (Fig. 4.4B). The energy dispersion of the QPI pattern on the S terminated surface along the $K - \Gamma - M$ symmetry line is shown in Fig. 4.4C. We find several dispersing patterns (shown in details in chapter B.8), however here we focus our attention to the QPI features that appear along the $\Gamma - M$ direction at around 100 meV (within the white dotted rectangle). In the Fourier transformed $dI/dV$ map taken at 100 meV (Fig. 4.4D) these QPI features correspond to the $\Gamma - M$ broad peaks, located between the Bragg peaks and the centered flower-like pattern. In the calculated DOS(k), at the corresponding energy shown in Fig. 4.4E and H, we identify a single dominant surface band that decorates the rims of the bulk band and terminates at the vicinity of the Weyl nodes. Detailed calculation shows that it indeed hybridizes with the Fermi-arc bands as they emanate from the Weyl nodes (Xu et al., 2018). The calculated JDOS (Fig. 4.4F) is similar to the measured QPI in Fig. 4.4D, showing both the broad peaks and the flower-like patterns. This identifies the broad QPI peaks with inter-band scattering processes (bounded by $Q_a$ and $Q_A$), and the flower-like pattern with intra-band scattering processes (bounded by $Q_b$) of that surface band. Following the energy evolution of the broad peak QPI pattern, highlighted in Fig. 4.4G, we find that it shifts towards smaller momentum transfers with increasing energy and seems to terminate below 200 meV. This dispersion is well captured by the DOS(k) calculation in Fig. 4.4H which shows that the surface band remains bound to the rims of the bulk band. Finally, cutting the dispersion DOS(k) through two adjacent Weyl nodes, as shown in Fig. 4.4I, reveals that the projected bands that the surface state follows, are the bulk Weyl cones that hybridize $\Delta E_{\text{Weyl}}$ above the Weyl node energy. The dispersion of the QPI pattern in Fig. 4.4G accordingly embodies the dispersion of the Weyl cones. In particular, its vanishing slightly below 200 meV corresponds to the merging of the two adjacent bulk Weyl cones and the termination of the Weyl gap about $\Delta E_{\text{Weyl}} = 95\text{meV}$ above the Weyl node energy. The maximal momentum transfer of $Q_A = 9.5\text{ nm}^{-1}$ at the Weyl node energy indicates the Weyl node momentum separation, $\Delta k_{\text{Weyl}}$, in
agreement with our ab-initio calculation (Fig. 4.4E). Although on the S termination the Fermi-arc contours are pushed out of this bulk gap and therefore could not be detected, on the Sn and Co terminations they lie within this gap which protects their hybridization and allows their detection.

4.3 Discussion

The $\text{Co}_3\text{Sn}_2\text{S}_2$ compound provides unique opportunity to perform local spectroscopy on all surface terminations, sharing the same crystal bulk, while allowing us to measure complementary aspects of its surface and bulk band structure: On the Sn termination we visualize the Fermi-arcs and measured direct signature of time reversal symmetry breaking, induced by the magnetic order of the Co atoms. This verifies the magnetic Weyl nature of $\text{Co}_3\text{Sn}_2\text{S}_2$; By comparing the Sn and the Co terminations, we demonstrated that modifying the surface potential may alter the Fermi-arcs dispersion and particularly the Weyl node connectivity. This has significant implication on the magneto-transport properties of the Weyl electrons (Fig. 4.1C). Finally, on the S termination, we extract the Weyl node energy and momentum, and the extent of the Weyl gap, by following the surface state dispersion, providing also a clear demonstration of the surface-bulk correspondence in Weyl semimetals. Our results characterize the time reversal symmetry broken Weyl phase of the semimetal $\text{Co}_3\text{Sn}_2\text{S}_2$ and demonstrate the unprotected aspects of topological semimetals, which can be manipulate and engineered by controlled surface perturbations.
Figure 4.4: Inter Brillouin zone Fermi arc connectivity on the Co surface. (A) Topographic image of the S terminated (001) surface ($V_{bias} = 50\text{meV}$, $I_{set} = 250\text{pA}$) featuring S vacancies. (B) $dI/dV$ map taken on the region shown in A ($V_{bias} = 100\text{meV}$, $V_{AC} = 5.3\text{meV}$ RMS, $f = 733\text{Hz}$), showing electronic interference patterns correlated with the location of the vacancies. (C) Energy-momentum QPI cut along the $K - \Gamma - M$ direction showing several dispersing QPI branches. (D) Fourier transformed $dI/dV$ map at the vicinity of the Weyl node energy showing flower-shaped QPI pattern around $\Gamma$ and broad peaks along $\Gamma - M$. (E) ab-initio calculated DOS(k) at the corresponding energy showing a single sharp surface band and the extremal scattering wave-vectors $(Q_{a,A,b})$ among it, where $Q_A$ corresponds to the largest momentum separation of the Weyl nodes, $\Delta k_{Weyl}$. (F) Calculated JDOS at the corresponding energy agrees well with the measured QPI pattern in D. (G) Highlighted QPI cut (from the dotted rectangle in C) showing inward dispersing peak from which we extract the Weyl cones structure in momentum, $\Delta k_{Weyl}$, and energy, $\Delta E_{Weyl}$. (H) Energy-momentum cut of the DOS(k) along the dash-dotted line in E showing the evolution of the $Q_{a,A}$ scattering processes. (I) Energy-momentum cut of the DOS(k) along the dashed line in E showing the extent of the inter-Weyl cone bulk, $\Delta E_{Weyl}$, gap as projected to the S surface termination.
Chapter 5

Construction of VT-STM

5.1 Introduction

During my research I have been constructing a versatile and highly accessible VT-STM (Stipe, Rezaei, and Ho, 2002) system (Fig. 5.1A), following similar design guidelines inspired by a dilution fridge STM head from Yazdani’s lab in Princeton (Misra et al., 2013), combined with our current commercial Unisoku STM transfer mechanism. The custom-built STM features a variable temperature environment ranging from 15K to room temperature using a continuous flow cryostat. The scanner is a “Pan walker” type (Pan, Hudson, and Davis, 1999), and is suspended by eddy current damped springs inside two radiation shields which are cooled by the cryostat. Our STM design provides extremely high accessibility to the tip-sample space paving a path to a variety of designated capabilities -

- *In situ* retractable commercial Orsay Physics UHV SEM, that can be positioned up to 50 mm from the sample granting a resolution better than 10nm
- Access to molecular beam epitaxy (MBE) grown samples transferred *in situ*.
- Possibility of performing a tip exchange without having to remove the sample
- Access to copper crystal for tip treatment without having to remove tip or sample
- Four-pocket e-beam evaporator allowing both warm and cold deposition conditions via manually controlled slits on the radiation shields
- Three sample contacts with an optional upgrade to six (1-bias, 1-gate, 4-probe)
- *In situ* annealing and ion sputtering of the sample surface
- Compatibility with our current lab’s STM tips and sample mounting mechanism

The VT-STM system set of capabilities gives rise to two major relative advantages. The first one, is the combination of the excellent spatial resolution of the STM and
the SEM. This grants our new system the ability to resolve areas of orders millimeters down to angstroms, giving us a unique and versatile magnification range. Two recent experiments carried out in the system exemplify this. The first experiment showcases a $WP_2$ crystal, which takes the form of a thin elongated “whisker” and spans few tens of $\mu m$ in width. Such thin $WP_2$ crystals were recently found to host hydrodynamic electron flow (Gooth et al., 2017). Exploring the interesting physics of this compound in STM requires access to its narrow facet, by cleaving the crystal standing up. Indeed as illustrated in Fig. 5.1B, we managed to cleave such a crystal, and by virtue of the STM and SEM - further managed to approach and measure step edges on the narrow facet. The second experiment depicts (Fig. 5.1C) $InAs$ nanowires, which were transferred to a $Si$ based device under UHV. This device allows tunneling current to be drained from its $Au$ contact, as well as applying a gate voltage to the $Si$ substrate, thus allowing to tune the chemical potential of a nanowire during an STM experiment. To that end, one needs to locate a nanowire on top of the pre-fabricated trenches. Using the SEM one can easily approach to such location, especially because the flatness of the substrate allows the user to further image the electronic shadow of the tip (Fig. 5.1C), which grants a controlled and precise approach. Finally, upon approach we validated (Fig. 5.1D) the effect of the gate on the $InAs$ band-gap, by measuring its shift in energy over the spatial region correlated with the trench.

The second advantage, which is still a work in progress, is the UHV connection of the VT-STM to our existing Omicron commercial MBE unit (Fig. 5.2A), which notably involves the vibration decoupling between the two systems, and the STM scanning test of the plate holder (Fig. 5.2B). We recently managed to grow nanowires and micro-crystals of $SnTe$ (Fig. 5.2C), a very interesting crystal combining the novel physics of topological crystalline insulator (Fu, 2011; Okada et al., 2013) and higher order topological insulators (Schindler et al., 2018). Ultimately the system will offer an all-in-one solution which combines MBE growth, fabrication via e-beam evaporator and STM scanning abilities, thus establishing an experimental basis for the exploration of novel topological phases in low dimensions.

5.2 Microscope Assembly

5.2.1 Electro-polishing

Before assembly, it is advantageous to polish parts which reside in vacuum. The shiny facets allow for overall better vacuum performance by reducing the surface to area ratio. It is in principle possible to outsource the electropolish procedure to local (Odem) or foreign (Russamer Lab) companies. Do note that for small parts (relevant scale are
5.2. Microscope Assembly

Figure 5.1: Latest achievements of the VT-STM. (A) Main chamber (left) featuring a top loading design, variable temperature cryostat (blue), UHV SEM unit (yellow), E-Beam evaporator (red) and at the center of the spherical chamber the VT-STM head (green). VT-STM head (right) blown up and shown in its constructed form. (B) SEM micrograph (left) taken when the STM tip is few hundreds of nanometers away from tunneling conditions. STM topography (right) taken on the narrow facet of $WP_2$ crystal (C) SEM micrographs (left) of a silicon based device capable of gating $InAs$ nanowires. Note the electronic shadow resulting from the tip. STM topography (right) of a nanowire sitting on a trench. (D) $dI/dV$ line measurement taken on the nanowire shown in C, illustrating the effect gating has on the energy shift of the semiconducting band gap. (C,D adapted from unpublished work of Tom Koren)
beakers sizes), the setup is very simple. The following are guidelines we successfully employed for oxygen-free high thermal conductivity copper (OFHC), beryllium copper and molybdenum. Aluminum and titanium were found to be a bigger challenge, independent of what recipe we used, and were left unpolished.

Irrespective of the work-piece material, the setup (Fig. 5.3A) will require the following:

- High current power supply
- Adjustable height platform
- Magnetic stirrer
- Beaker
- Stainless steel cathode
- Wires and crocodile clips
- Copper wire

In the polish process it is preferable to use a cathode which respects the symmetry of the desired polish. For example, we spot-welded a stainless steel shim, to create a cylindrical cathode of diameter fitting a standard size beaker, which was found very useful for most small parts. In contrast, for the radiation shields, a different type of cathode was made (Fig. 5.3B), allowing both an inner and outer surface polish. Tailoring the cathode for the part in hand allows for a more uniform result. The anode may
consist of a copper wire wrapped around few parts, or simply the part itself. For small parts the former may be more efficient. The important thing to note is a very good electric contact is needed through-out the anode for satisfactory polish results, and so a dense and tight wrapping of the copper wire is required. Finally the general protocol is the following:

1. Prepare electrolyte according to the work-piece (see below). Always start with $H_2O$, and note some acids are diluted out of the box.

2. Stir using magnet for 3-5 minutes.

3. Place cathode in beaker, and connect using a crocodile to power supply. Make sure crocodile remains above solution level, as they are not made from stainless steel.

4. Place work-piece in solution. Confirm it is not touching the cathode.

5. Fill an extra Beaker with $H_2O$.

6. Close hood, turn on power supply, confirm bubbles forming around parts ($H_2$ gas). Typical currents 3-7A.

7. Turn off power supply, dip parts in $H_2O$ beaker.

<table>
<thead>
<tr>
<th>Material</th>
<th>Electrolyte ratio by volume</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Cu, BeCu$</td>
<td>$H_2O : H_3PO_4$ 1:1</td>
<td>For oxide removal $CH_3OH : HNO_3$ 9:1 (No current required)</td>
</tr>
<tr>
<td>$Mo$</td>
<td>$H_2O : H_3PO_4 : H_2SO_4$ 4:4:1</td>
<td>If black coating appears, increase current or connect leads directly to part</td>
</tr>
</tbody>
</table>

5.2.2 Gluing piezo stacks

The VT-STM has two sets of motors, which allow it’s tip holder to move in XY directions (parallel to sample) and along Z. The motors employ a set of piezo stacks purchased from PI (model Pic 255) which are pressed against a polished sapphire rated at S/D of 60/40 (Gavish). In the following I will describe the procedure we performed in the assembly of the VT-STM, describing separately the XY and Z componets.

The XY stage (Fig. 5.4A) consists of three main parts molybdenum base plate, copper plate and a beryllium copper spring. In the original design of the VT-STM the
**Figure 5.3:** Electropolish setup and results (A) General setup for small parts, includes wrapping of the parts with a conductive wire or (B) with an electrode that respects the symmetry of the work piece. (C) Practice run result on a small BeCu work piece. (D) Results on the different copper parts that assemble the STM microscope.
copper plate had guide holes for the wires connecting the piezo for thermal anchoring. During assembly these holes were found to be very tricky to use. Future design might benefit from a more flexible setup. Ultimately, the sapphire assembly is sandwiched (Fig. 5.4C) in between the plates, which then transfers the translation to the tip assembly. Following the overall symmetry of the microscope, we installed three pairs of piezo stacks along cartesian XY axes. Firstly, we used a thin layer of UHV epoxy (EPO-TEK H74F) to glue the first three stacks on the molybdenum plate, which were aligned according to machined groves. Care must be given to ensure the proper piezo direction response. We have chosen to set the X direction in proximity to the sapphire, which therefore means we have glued the Y facing side of stacks. The plate was then cured for two hours at 80°C. Blank flange can be used to apply pressure on the piezo stacks during curing to ensure a parallel end result. To find the exact position for the counter copper plate it is advisable to assemble the three parts (molybdenum base plate with glued stacks, copper plate and spring). The beryllium copper spring needs to be positioned and tightened gently on top of three sapphire balls (purchased from Swiss Jewel), while confirming the balls reside in their dedicated positions. This allows to properly tune the gluing position of the remaining stacks to the copper plate and achieve a more symmetric result. Finally the whole assembly was cured for two hours at 80°C. Refrain from performing a faster cure at higher temperatures, as that will facilitate an oxide layer on the freshly electro-polished parts.

The Z stage (Fig. 5.4B) is more straightforward. It too consists of three pairs of piezo stacks which are glued directly to three molybdenum rectangular plates. Prior to gluing, the stacks electrodes were spot welded using copper electrodes by applying 4 joules energized current, and trimmed to allow an easy positioning. We have ensured the proper polarization of the stacks, by simply having each electrode polarity face its opposite one. Finally the pizeo stacks were aligned according to groves, glued and cured for two hours at 80°C.

5.2.3 Gluing sapphire prism

The scanner tube piezo of the VT-STM resides on a molybdenum wire feed-through rod (known as the “crown”), which is glued to the Z piezo stacks sapphire prism (Fig. 5.5A). The gluing of these two parts was quite challenging, as the surface area of gluing is large, which therefore increases the probability of trapped air bubbles (Fig. 5.5B). Such bubbles are notoriously dangerous for UHV requirements, as they tend to leak very slowly. The gluing process might warrant second (and thirds) trials, and in between it is important to properly clean the prism and the crown by sonicating both in acetone.
for at least 5 minutes. The protocol that proved to work for us (Fig. 5.5C) was the following:

1. Gluing top part of prism to the crown, by stamping the prism on a thin layer of UHV epoxy.

2. Filling a syringe with UHV epoxy and putting it in a centrifuge spinning at 6000RPM for 5 minutes. This will allow the air to separate from the epoxy. The syringe may look full, but after phase separation it will be evident it is roughly half empty.

3. Inserting four pre-electropolished wires at the exact dimension of the wires feed-through of the crown.

4. Wrapping the bottom part of the crown and prism assembly with plastic wrap.

5. Puncturing a small hole in the plastic wrap at the fifth hole position and injecting the epoxy using the syringe until epoxy comes out from the top.

6. Inserting a fifth wire

7. Hold for 15 hours (overnight) in order for the epoxy to become more viscous.
5.2. Microscope Assembly

8. Taking out the wires slowly, while avoiding the creation of bubbles

9. Clean prism with wipers rinsed with very little acetone followed by isopropanol.

10. Cure for three hours at 80°C

5.2.4 Electronic wiring

The following table lists the wires connections for the STM head.
### Contact Role | Wire type | Remarks
--- | --- | ---
Sample 1 | Coaxial | 32 AWG
Sample 2 | Coaxial | 32 AWG
Sample 3 | Coaxial | 32 AWG
Tip | Coaxial | 32 AWG
Coarse X | Solid core | 30 AWG, Twisted-pair
Coarse X (GND) | Solid core | 30 AWG, Twisted-pair
Coarse Y | Solid core | 30 AWG, Twisted-pair
Coarse Y (GND) | Solid core | 30 AWG, Twisted-pair
Coarse Z1 | Solid core | 28 AWG, Twisted-pair
Coarse Z1 (GND) | Solid core | 28 AWG, Twisted-pair
Coarse Z2 | Solid core | 28 AWG, Twisted-pair
Coarse Z2 (GND) | Solid core | 28 AWG, Twisted-pair
Coarse Z3 | Solid core | 28 AWG, Twisted-pair
Coarse Z3 (GND) | Solid core | 28 AWG, Twisted-pair
Fine X+ | Solid core | 34 AWG
Fine X- | Solid core | 34 AWG
Fine Y+ | Solid core | 34 AWG
Fine Y- | Solid core | 34 AWG
Fine Z | Solid core | 34 AWG
Fine AUX | Solid core | 34 AWG
Outer-capacitor | Solid core | 34 AWG
Inner-capacitor | Solid core | 34 AWG

In total, 22 connections feed the STM (coaxials purchased from Lakeshore - Type SC, and solid core from Accuglass). To avoid crosstalk, the current driving wires (sample and tip) are directed to the top of the head and the rest of the wires are directed to the bottom.

The scanner tube (Fig. 5.6A) is soldered with lead-free solder (97% Sn 3% Cu) to a kapton coated copper wire (34 AWG). During soldering the solder iron was held at temperature 275°C. The contact surface was treated with minimal flux (Castolin). Following a connectivity check, the solder connections were reinforced with epoxy (Fig. 5.6B). Finally, the tip holder was wired to a coaxial cable (Fig. 5.6C), with a 34 AWG kapton wire connected to its shield, providing a possible grounding shield for the tip-sample region.

The XY coarse piezos were contacted (Fig. 5.6E) along their respective planes (i.e above and below the sapphire) separately, and were later shorted on the bottom STM...
connectors plate. We found using a 30 AWG wire, gives a nice trade-off between rigidity and flexibility. The protocol of wiring we employed was the following -

1. Clamping the wires in place on the ribs, and marking the electrodes positions with a marker.
2. Removing the wires from microscope.
3. Cutting the starting and ending position of the marker using a 28 AWG cutter.
4. Exposing the kapton with razor blades.
5. Clamping wires back, and spot welding with 6 joules.
6. Reinforce contact with epoxy

During this procedure for all eight wires (four wires per plane) some wires were disconnected after spot welding in which case we spot welded them again. Moreover there were some incidents of sparks being generated while spot welding, this could be because current was driven through the kapton. The spot welder is a bit bulky for this compact area, however soldering is not an option for the tantalum electrodes. In a similar fashion one can also contact the Z coarse piezos. Their sparse geometry makes the spot-welding much easier. The VT-STM was designed to walk using using the stick-slip protocol (Besocke, 1987). However, the Z motors have additional contacts which allows different biasing schemes as well.

Upon completion of the STM construction, initial ambient tests were taken. Specifically we analyzed the piezoelectric positioners range, in terms of coarse movement, by recording a reference point via an optical microscope as shown in Fig. 5.7A-B. We then proceeded to calibrating the scanner tube by visualizing the atomic structure of HOPG (5.7C) and terraces on the surface of copper (5.7D). By fitting the lattice constant, and by averaging several atomic steps heights, the XY and Z piezo tube response can be calibrated.

Finally, for easy troubleshooting of possible disconnections in the wiring scheme, the following lists typical capacitance values of the piezo contacts. In general, these values will vary depending on temperature of the STM and the measuring method. However, as a general rule, if the charging is unstable or the reading is zero, an in-depth investigation is needed.

- Capacitance of a single X or Y piezo stack: 4.4 nF
- Capacitance of a single Z piezo stack: 4.5 nF
Figure 5.6: **Scanner tube and piezo stack wiring** (A) Z electrode of scanner tube contact - solder is coated with UHV epoxy. (B) Scanner tube assembly ready for mounting. 34 AWG wires contact the different electrodes and are channeled down. (C) Titanium tip screw holder with a coaxial wire connected to it. The shield of the coaxial has an extension 34 AWG wire for possible future shielding of the tip-sample region. (D) View of (B) and (C) before gluing the two. (E) STM head partially assembled, XY stage is wired plane-wise using 30 AWG wires.

- Capacitance of the inner tube fine Z electrode with respect to one of the quadrants: 1.5-2 nF
- Capacitance of the AUX electrode with respect to Z: 2.2 nF

### 5.3 System Grand Assembly

The VT-STM grand Assembly is shown in Fig. 5.8. In the following chapter important sub-assemblies will be discussed in a bottom up fashion.

#### 5.3.1 Suspension springs

The microscope assembly described in chapter 5.2 is suspended on three springs. This additional vibration isolation stage allows for a significant reduction of vibration, as was previously discussed by Julian Chen (Chen, 2007) in chapter 10.

The springs used in the VT-STM are made from Inconel X750 (Suhrm spring works), each hosting 97 coils with an outer diameter of 0.157" and coil thickness of 0.012". The spring constant of a helical spring is given by

\[ k = \frac{Gd^4}{8D^3n} \]  

(5.1)
5.3. System Grand Assembly

Figure 5.7: **Scanner tube and piezo stack calibration** (A) Optical microscope setup used for tracking tip location. (B) Max scan range, as determined by the optical images taken along Y (top) and X (bottom) movement. (C) Atomic scale image of HOPG, used for calibrating the XY scanner tube response. (D) Terraces on Cu(111) which can serve for calibrating the Z scanner tube response.

Figure 5.8: **VT-STM grand assembly** (A) Top view of system, inset shows the CAD version, highlighting the role of the different components: piston supporting frame (red), load-lock (yellow), UHV suitcase (orange) and main STM chamber (gray). (B) View of STM microscope assembly, showing the aluminum outer radiation shield. (C) a PhD student and his two babies.
Chapter 5. Construction of VT-STM

Figure 5.9: STM head without radiation shields The STM is suspended from three springs (top) and is damped using a set of six magnets (bottom).

where $G$ is the modulus of elasticity, $d$ is the mean diameter, $D$ is the outer diameter and $n$ are the number of coils. By calculating $k$ and weighing the microscope head we can extract the natural frequency of the STM. Doing so gives $\omega_0 = \sqrt{\frac{k}{m}} = 8.1$ Hz for the VT-STM. In general, one will want to decrease this number even further, however this usually means having longer extending springs, which is problematic in most designs.

The springs are then damped using a set of six (Fig. 5.9) rare earth samarium–cobalt magnets (HKCM), by the eddy currents induced in the copper inner radiation shield.

5.3.2 Radiation shields

Fig. 5.10A shows the VT-STM and its two radiation shields, termed inner and outer. The inner radiation shield is made from OFHC for its high thermal conductivity, and the outer is made from aluminum (alloy 6061) for its durability and light-weight. The versatile capabilities of the VT-STM require tunable radiation shields, namely they need to allow the different instruments in the system to have access to the tip sample region. For that purpose we have designed rotating slits, which are maneuvered using two gears using the vertical top-loading manipulator. Indeed the radiation shields feature four working modes -

1. Mode I: single window reveals left most marking hole (Fig. 5.10B) - radiation shield is fully closed. Used for base temperature setup.
2. Mode II: double window reveals middle two marking holes (Fig. 5.10C) - radiation shield hole for electron beam, SED and evaporation holes are open. Used for SEM scanning and hot evaporation setup.

3. Mode III: quad window reveals four marking holes (Fig. 5.10D) - radiation shield evaporation holes are open. Used for cold evaporation setup.

4. Mode IV: large window open (Fig. 5.10E) - radiation shield main window is open. Used as a visual aid for top loading samples and tips, and as an access port for side loading of MBE samples.

The inner radiation shield is equipped with four SMP coaxial feedthroughs (PE) on its top flange (Fig. 5.11B), used for the sample and tip connections. They are then connected to four pre-cooled coaxials wrapped around the cryostat. The bottom flange of the inner radiation shield interfaces the non-current wires of the microscope. The feedthrough (Fig. 5.11A) is based on a similar design principle described by Vershinin (Vershinin, 2004), in which a 1mm OFHC wires are glued to a sapphire cylinder (Swiss Jewel) which in turn are glued to the bottom flange. This setup allows a spacious and cheap interface, while maintaining satisfactory cooling anchor. The wires are then soldered to a springy gold connector (TE) which act as plugs for the copper wire. We found these plugs to be the weak point of this feedthrough setup, and indeed the majority of disconnections occurred at the soldering point of the wires and the gold connectors. Fig. 5.11C surveys the convention of the wiring scheme, where the red arrow marks the location of a scratch made on the bottom flange, and it is assumed the flange is observed from below. Finally, the outer radiation shield features a similar bottom flange feedthrough. In order to decouple heat transfer, the wires connecting the outer shield and the chamber are made from manganin (Allectra). The wires terminate at two commercial PEEK plugs (Accuglass) and are wired according to the diagram shown in Fig. 5.12.

5.3.3 Cryostat

The Helitran LT3B from ARS is a UHV liquid helium flow cryostat, allowing working temperature ranging from 4.2K to room temperature, with sub-angstrom level vibrations. This is made possible due to a special large-surface-area matrix heat exchanger (Fig. 5.13A), and a fine controllable steady flow rate. It is fed from a triaxial transfer line (Fig. 5.13D) carrying liquid helium, and helium exhaust through an adjustable needle valve onto the cold stage, which is rigidly bolted to the top flange of the inner radiation shield.
Chapter 5. Construction of VT-STM

Figure 5.10: **Radiation shield working modes** (A) STM head assembly without radiation shield covers, the top aluminum flange connects to outer radiation shield, and the bottom copper flange connects to inner radiation shield. (B-E) The different working modes I-IV described in text. (F) The outer radiation shield gears, partially assembled. These will transfer the top loading manipulator rotation to the slit.

Figure 5.11: **Radiation shield wiring scheme** (A) The feedthrough which interfaces the radiation shields - gold connector plugs connect OFHC wires which are glued to a sapphire cylinder. (B) SMP Coaxial connectors, glued to a sapphire cylinder (C) Wiring scheme of the two bottom radiation shields flanges, as observed from the bottom. Pink symbols refers to coarse motors, blue refers to scanner tube.
The transfer line is mounted on a 350 liter dewar (Cryofab), whose top neck features two half-inch ports, allowing up to two transfer lines connections. This setup allows a continuous cooling process, however requires careful pressure maintenance regimes, and was yet to be perfected in the system. Nevertheless, more than 10 days of stable base temperature (about 10K, 15K and 60K for both radiation shields closed, only inner closed and both open respectively) are currently achievable (Fig. 5.13B).

### 5.3.4 Scanning Electron Microscope

The VT-STM is equipped with a compact, UHV compatible, electrostatic SEM (e-CLIPSE from Orsay Physics). The SEM column is designed and optimized to work at large working distance compared to typical electro-magnetic SEM columns, which allows its positioning in close proximity to the outer radiation shield (Fig. 5.14A) of the VT-STM.

Micrographs are acquired by a Secondary Electron Detector (SED) located on the side. Due to its narrow form factor, it is possible to allow its penetration to both radiation shields, thus allowing maximal absorbance of emitted electrons. This side detection further allows to observe the tip’s shadow (Fig. 5.1C) thus directly probe the tip sample distance and spatial approach location.
In the setup of our system the column is mounted on a 55 degree tilted retractable port-alligner. This has led to quite a dominant vibration noise on the scale of 20 Hz, which hampers the performance of the SEM. Our tests shows that this noise is irrespective of the microscope assembly, and in particular its eddy current damping magnets. Adding additional support to the column seems to reduce the amplitude of the vibrations, however not sufficiently. More rigid anchoring of the column to the frame is required and is currently under design.
5.3. System Grand Assembly

Figure 5.14: SEM chamber setup (A) View of the STM UHV chamber showing the SEM column on the left, the outer radiation shield on the center and the SED on the right. Both SEM and SED are in retracted position. (B) Plasma cleaning of the SEM column with $N_2$ gas following a system vent.


Huang, Shin Ming et al. (2015a). “A Weyl Fermion semimetal with surface Fermi arcs in the transition metal monopnictide TaAs class”. In: Nature Communications 6.1, p. 7373.
Huang, Xiaochun et al. (2015b). “Observation of the chiral-anomaly-induced negative magnetoresistance: In 3D Weyl semimetal TaAs”. In: Physical Review X 5.3.
Liu, Enke et al. (2018). Giant anomalous Hall effect in a ferromagnetic kagome-lattice semimetal.


Okada, Yoshinori et al. (2013). “Observation of dirac node formation and mass acquisition in a topological crystalline insulator”. In: Science 341.6153, pp. 1496–1499.


Parameswaran, S A et al. (2014). “Probing the chiral anomaly with nonlocal transport in three-dimensional topological semimetals”. In: Physical Review X 4.3.


Yin, Jia Xin et al. (2018). Giant and anisotropic many-body spin–orbit tunability in a strongly correlated kagome magnet.


Appendix A

Supplementary information TaAs

The following chapter contains supplementary information to the results shown in chapter 3.

A.1 Sample synthesis

The single crystals of TaAs were grown using the chemical vapor transport method in a two-zone furnace on the basis of the precursor of polycrystalline samples, which were prepared by mixing high-purity (> 99.99%) Ta and As elements. Both the polycrystalline TaAs powder and 0.46 mg cm$^{-3}$ of iodine were loaded into a 24-mm-diameter quartz tube and then sealed under vacuum. Two ends of the tube were kept at 1150°C (charged part) and 1000°C for 21 days. The synthesized single crystals can be as large as 0.5 to 1 mm in size.

A.2 Extended q-space map

To examine whether there are significant replication of QPI features to Bragg peaks higher than the first, we have taken a high resolution $dI/dV$ map (pixel per 0.5 Å, corresponding topography in Fig. A.1a). The q-space field of view we obtain contains 3rd order Bragg peaks. We indeed confirm that replications of QPI features are detected around $q = 0$ and the 1st Bragg peak only (Fig. A.1b).

A.3 $dI/dV$ maps: Raw data and symmetrization

We make use of the mirror symmetry of the crystal in order to enhance the signal to noise ratio of the QPI data, in which the mirror symmetry is commonly diminished due to small anisotropies in the rastering mechanism. Fig. A.2-A.3 shows the same map of Fig. 3.1, in other energy values, after mirroring (and rotation). For each data set used we confirm that no features are added or eliminated by the mirroring procedure. We
Figure A.1: Extended q-space map. (a) Topographic image taken during measurement of a high-resolution $dI/dV$ map (pixel per 0.5Å). (b) Fourier transform of a $dI/dV$ map measured over the region in left panel. No QPI signatures detected around Bragg peaks higher than the 1st order.

note that a leaf-like pattern about q 0 does not repeat at all energies (eg Fig. A.3-c,d), as would be the case had it originated from some long-wavelength modulations due to an underlying inhomogeneous potential.

A.4 Fermi arc scattering signature

Fig. A.4 presents an additional data-set taken over a different region (with respect to the one shown in Fig. 3.1) on the surface of the same sample show the leaf-like patterns that we associate with scattering involving Fermi-arcs. This demonstrates that this feature is not due to a specific realization of a vacancy distribution or any other local parameter, but rather a property of the interfering electrons.

A.5 Agreement between vacancy- and step edge–induced QPI

The step edge that we measure is oriented 49° relative to the $\Gamma - X$ crystallographic direction (see Fig. 3.2a). The Fourier transform of the 1D interference pattern that we measure due to scattering off this step edge at the Fermi energy ($E_F$) perfectly agrees (Fig. A.5) with 2D QPI pattern (along the same cut) that we measure off vacancies at the corresponding energy.
Figure A.2: **Raw $dI/dV$ maps.** (a-o) Raw Fourier transformed $dI/dV$ maps at different energies. All features discussed appear in the raw data.
Figure A.3: Mirroring of QPI maps. (a-o) To enhance the symmetric features based on the mirror symmetry of the crystal, the raw QPI maps were first rotated to align the Bragg peaks, and mirrored around a mirror axis taken along two opposite Bragg peaks. No features were artificially added or eliminated during this procedure. Mirror symmetry broken mainly by minute anisotropy in the scanning mechanism is restored.
A.5. Agreement between vacancy- and step edge–induced QPI

Figure A.4: QPI pattern involving Fermi-arc scattering from a different vacancy distribution. (a) Topographic image showing the local distribution of As vacancies over which the presented $dI/dV$ map was taken. (b) Fourier transform of that $dI/dV$ map showing a leaf-like pattern similar to that of Fig. 3.1f. Momentum resolution is somewhat lower due to smaller spatial field of view imaged.

Figure A.5: Agreement between vacancy and step-edge induced QPI. (a) 3D plot presenting the line cut’s QPI data in the vertical plane crossing the horizontal 2D vacancy induced QPI map at the right angle (49°) and energy (0 meV). It shows the correspondence between the identified features in the 2D QPI pattern – namely, ellipse at $q = 0$ and squares at zone edges – and their dispersion. (b) corresponding SSP.
A.6 Fermi arc dispersion

The extent of the Fermi-arcs in momentum space decreases as the Weyl node is approached (Fig. A.6. At the energy of $W_2$ the arc locus becomes almost a straight line connecting the pair of nodes. As a result, at the Weyl energy, the shortest $q$ vector in the QPI pattern involving the Fermi arcs results from scattering between $k$ values in the vicinity of the Weyl nodes. As a consequence it represents the distance between the Weyl nodes.

A.7 Correlation between scatterer-free dI/dV modulations and replications of QPI patterns

The QPI patterns that we observe near scatterers show clear replications of several patterns around adjacent Bragg peaks, while the spatial maps of local density of states in scatterer-free regions show oscillations on the scale of a unit cell. Both of these phenomena reflect the interaction of the electrons with the periodic lattice potential.

The Bloch theorem constrains a state with a crystal momentum $k$ to be a superposition of momenta $k + G$, where $G$ is the reciprocal lattice vector, $\Psi_k(r) = \sum_G C^*_G e^{i(k+G)\cdot r}$. Consequently, the local DOS in a vacancy-free region becomes $\sum_g A_g \exp^{ig\cdot r},$ where $p = \sum_{G,k} C^*_G C^s_G \delta(E - F_k)$ is the amplitude of the Bragg peak that corresponds to $g = G - G'$, $r$ is the position, $E$ is the energy, and $F_k$ is the energy of the state with momentum $k$. A state with multiple significant coefficients $C^s_G$ is generally a state with a fine structure within the unit cell.

A vacancy violates the periodicity and adds a potential $V(r)$, whose Fourier transform is $V_q$. A realistic Gaussian-like impurity potential with a scale $\lambda < a$ (where a
is the lattice constant) would yield a QPI pattern that is gradually attenuated with increasing momentum, with a characteristic decay momentum of $2\pi/\lambda$. While we do not know the details of the potential, it is reasonable to expect that it allows momentum transfer that extends beyond $2\pi/a$. The vacancy may scatter an electron from a state $\Psi_k(r)$ to a state $\Psi_{k'}(r)$ through a series of momentum transfers $q_g$ satisfying $q_g = k - k' + g$. The amplitude for each such process is proportional, within the Born approximation, to $\sum G' V_q C^*_k G + C^k_{G'}$. Hence, multiple significant coefficients $C^k_G$ result in multiple replicas of the QPI around several Bragg peaks, limited by the ability of the potential to provide the required momentum transfer. The QPI pattern at momentum $q$, which is the Fourier transform of the local density of states, sums all the scattering processes that involve a momentum $q$, provided that both $E_k$ and $E_{k'}$ are identical to the energy being probed.

The QPI pattern that we measure for the non-topological states is highly anisotropic in a band selective manner (bowtie strongly replicated in $\Gamma - X$, ellipse in $\Gamma - Y$), highly dispersive, non-monotonic in $q$ and sharply changing at high $q$ (note ellipse pattern at Bragg peak half intense, half faint). All these are consistent with the wave functions having several significant Bloch components. The spatial modulations in the scatterer-free regions lend further support to this interpretation. An alternative explanation by which the replicated structure of the QPI emerges from properties of the scattering potential requires an elaborate fine tuning of the potential. As the next subsection shows, the wave function structure that we extract from the QPI pattern is also consistent with the theoretical calculations.

**A.8 Correspondence between QPI patterns and Bloch wave function**

Fig. A.7a shows that at low energies the bowtie QPI appears in comparable intensities at $q=0$ and around the Bragg peak. At somewhat higher energies ($-70 \text{meV} < E < 80 \text{meV}$) the Bragg peak pattern dominates, while at yet higher ones the $q=0$ pattern becomes dominant. The ellipse appears with comparable intensities around these two $q$ values at most energies at which this band exists. We further note that around Bragg peaks both the ellipse and the bowtie appear relatively intense at $q < G$ and fainter at $q > G$. We have not detected any signature of replications for the Fermi arcs, which probably means their wavefunction contains a single dominant Bloch term, or several components whose momentum difference is larger than the scale of momentum of the potential.
We now demonstrate how the calculated structure of the Bloch wavefunction of the different bands can reproduce these features. We first show in Fig. A.7c calculated Bloch coefficients of the three bands at energy -100meV and at a single momentum (point 6 in inset). Remarkably, we indeed find that both the ellipse and the bowtie wavefunctions are constructed from two main Bloch terms of adjacent $G$ values. For the ellipse these are $G_{0,0}G_{0,-1}$ (the subscripts refer to the number of reciprocal basis vectors in the x and y directions), while for the bowtie we find $G_{0,0}$ and $G_{-1,0}$. Indeed in the vacancy-free $dI/dV$ map we find modulations in the $\Gamma - Y$ at energies where the ellipse band dominates, and modulation in the $\Gamma - X$ direction when the bowtie band dominates the $dI/dV$ map. In contrast, for the Fermi arc we find a Bloch wavefunction which is strongly dominated by a single term - $G_{0,0}$. Hence, it is never replicated to higher Bragg peaks, and unlike dangling-bond derived trivial bands, it is weakly modulated by the surface potential.

From this distribution of coefficients of the ellipse, for instance, one can construct a rough picture of the wave functions in the range of momenta that covers nine Brillouin zones as shown in Fig. A.8 (left), where color represents the intensity of Bloch coefficient in Fig. A.7c with matching color (i.e. gray – strong, orange – intermediate, pink – weak but finite). The intensity of the various instances of the QPI pattern can be read off from this information by multiplying the intensities of the initial and final wave numbers. Accordingly the strongest QPI signal originates from scattering between states for which $k_x \approx \pm |\pi/a - \delta k|$ (with a small $\delta k$) and $k_y \approx 0$. This scattering appears around the $\Gamma - Y$ Bragg peak. Since only the internal halves of the ellipse band have strong Bloch coefficients only half an ellipse will appear strong in that QPI pattern, as indeed seen at the QPI pattern Fig. A.8 (right). The next dominant QPI pattern would arise from scattering between $k_x \approx \pi/a \pm \delta k$ (with a small $\delta k$) and $k_y \approx 0$ (gray to orange) and would generate the slightly weaker QPI pattern at $q = 0$. Next contribution comes from scattering between momenta at the remote zones on either side of the central one (orange to orange) which contributes the faint external halves of the QPI pattern at the $\Gamma - Y$ Bragg peak. These are hardly seen in the QPI shown in Fig. A.8 (right), but detected at other energy cuts as in the Fermi energy $E_F$ (Fig. 3.1c). The faintest ellipse QPI pattern we still track would come from momentum transfer along the y axis, between states where $k_x \approx \pi/a \pm \delta k$ (with a small $\delta k$) and $k_y \approx 0$ to states where $k_x \approx \pi/a + \delta k$ (with a small $\delta k$) and $k_y \approx \pm 2\pi/a$ (gray to red). The QPI signal of this event appears at the $\Gamma - X$ Bragg peak (barely observed in Fig. 3.1c). A similar analysis may be carried out for QPI patterns of other bands.
A.8. Correspondence between QPI patterns and Bloch wave function

Figure A.7: Structure of the Bloch wave function of the different bands. (a) Energy dependence of the Bragg peaks’ intensities in the vacancy-free region. Inset: Fourier transform of the $dI/dV$ map from which the intensities are extracted. (b) Energy dependence of the QPI intensities of the bowtie and ellipse patterns (orange and blue, respectively) around $q = 0$ and around the Bragg peaks (see inset for legend). The ellipse is correlated with the Bragg peaks along $\Gamma - Y$, whereas the bowtie is correlated with Bragg peaks along $\Gamma - X$. (c) Coefficients of the Bloch wave function calculated by DFT for the ellipse, bowtie and Fermi arc bands (left to right) at $E = -100\text{meV}$ and $k$ marked by points 6,8,1, respectively, at inset. It shows the anisotropy of the Bloch wave function of the ellipse and bowtie bands along $\Gamma - X$ and $\Gamma - Y$ respectively as they have dominant $G$’s along these directions in addition to the dominant $G = 0$ terms. In contrast, the Fermi-arc wavefunction is composed of a single dominant $G$, and several evenly distributed subdominant coefficients reflecting its extended isotropic nature.
Appendix A. Supplementary information TaAs

Figure A.8: Correspondence between calculated Bloch wavefunction and the QPI pattern. Left: layout of momentum space, centered around zero momentum, where color of the different zones mark the intensity of their associate coefficient in the Bloch wave function of the ellipse band shown in Fig. A.7c. The intensity of the QPI pattern around a given Bragg peak $g = G - G'$ is correlated with the intensity of the multiplication of the Bloch coefficients of the relevant scattering process $C_k G C_{k+G'}$. Right: QPI pattern at corresponding energy (-100meV). Replications of the ellipse (as well as bowtie) band appear with matching intensities.

A.9 Band structure calculations

The ab-initio density-functional theory calculations were performed in the generalized-gradient approximation level with spin-orbital coupling, which is implemented in the Vienna ab initio simulation package (VASP). To simulate the surface, a slab model with a thickness of seven unit cells was constructed, in which the top and bottom surface are terminated by As and Ta, respectively. The surface band structures and the Fermi surface were projected (Fig. A.9 to the first unit cell of the As-terminated side, which fits the previous experimental band structure well (Yang et al., 2015).

A.10 Extracting the intensity of QPI features

In Fig. A.7b we plot the average intensity across the ellipse and bowtie around $q = 0$ and Bragg peaks. To extract these intensities we have manually defined the maximal bounding box around a quarter of each feature such that will not contain neighboring features. We then averaged over the 30 highest maxima (Fig. A.10) within the bounding box making sure that these maxima indeed fall on the feature of interest and not on a spurious high intensity pixel. From this value we have subtracted the average background in the vicinity of the box (note that the background level is more than an order of magnitude weaker than the intensity of the features, and therefore had little effect on the resulting value). We have repeated this procedure around the Bragg peak by translating the bounding box by a reciprocal wave vector such that the bounding box is defined once per feature.
A.11. Splitting the line-cut $dI/dV$ into submaps

In Fig. 3.2 we split the measured $dI/dV$ line-cut map (Fig. 3.2b) into two separate sub-maps, one contains the $dI/dV$ features measured on As sites (Fig. 3.2e) and the other on Ta (Fig. 3.2f). Fig. 3.2b already shows the $dI/dV$ after the DC term has been removed (by subtracting the average $dI/dV$ value at each energy). As a result false color shows position with intensity larger than the average (blue) and smaller (red). We find perfect correlation between the atomic sites in the topographic profile displayed at the bottom and the large/small intensity pattern. Accordingly the sign of the flattened $dI/dV$ corresponds to being on/off atomic sites. We then split this map gathering locations with low and high intensities separately, which forms Fig. 3.2e,f. Since the distribution of these points is regular this amounts to lowering the sampling resolution for each sub-map. The lower spatial resolution results in smaller field of view in q-space. However, it allows to separate overlapping low-q features into those residing on/off As sites. Note, that the sum of the Fourier-transformed sub-maps (Fig.

Figure A.9: Wavefunction distribution. Ab-initio calculation of the wavefunction equal density surface calculated for the ellipse band (left) and Fermi arc (right) at -100meV and arbitrary k-value. The Fermi arc’s density profile is smoother and isotropic compared to the ellipse’s distribution.

For the subtraction of the Bragg peak QPI from the q=0 one we have constructed the multiple zone boundaries based on the atomic Bragg peaks, and subtracted the average of two zones centered about opposite Bragg peaks, normalized to the average intensity of the feature to be eliminated in the central q=0 zone.
**Figure A.10:** Extraction of QPI feature intensities. Average over 30 pixels of maximal intensity within a predefined bounding box.

3.2g,h) does not yield that of the complete map since the relative phase information is lost when displaying the magnitude of the Fourier decomposition.
Appendix B

Supplementary information $Co_3Sn_2S_2$

The following chapter contains supplementary information to the results shown in chapter 4.

B.1 Single crystal growth of $Co_3Sn_2S_2$

Single crystals were grown by a self-flux method with the congruent composition. The stoichiometric ($Co : Sn : S = 3 : 2 : 2$) samples were put in a graphite crucible sealed in a quartz tube. The samples were heated to 1000°C over 48 hours and kept there for 24 hours. The samples then were cooled to 600°C over one week and kept there for 24 hours. The compositions of the crystals were checked by energy dispersive X-ray spectroscopy. The structure and orientations were determined by a single-crystalline X-ray diffraction.

B.2 Ab-initio calculations

We calculated the electronic band structures based on the density functional theory by using the code of Vienna ab-initio simulation package (VASP) with the projected augmented wave method (Kresse and Furthmüller, 1996). The exchange and correlation energies were considered in the generalized gradient approximation (GGA) level of Perdew-Burke-Ernzerhof function (PBE). The cutoff energy was 400 eV, and the $k$ mesh for self-consistent calculation was $10 \times 10 \times 10$. To investigate the surface state, we projected the Bloch wavefunctions into maximally localized Wannier functions (MLWFs) (Mostofi et al., 2008). The surface states were calculated with a semi-infinite open boundary condition by using a tight-binding model Hamiltonian constructed from the MLWFs and Green’s function method (Lopez Sancho, Lopez Sancho, and Rubio, 1984). The joint density of states (JDOS) were calculated by using the Fourier-transformed density of states (Kourtis et al., 2016). The Fermi energy of the different surface termination projections was fitted to the QPI data, by both comparison to constant energy
Fourier transformed maps and high symmetry directions momentum transfer linecuts. Following this procedure for each of the terminations, we have shifted the Fermi energy by 55 meV, 45 meV and 40 meV for the Sn, Co and S surfaces, respectively. This uncertainty is well within the accuracy of the ab initio calculation which is not better than ±10 meV.

### B.3 Identification of Co,S,Sn terminations

Below we describe the considerations that led us to determine the chemical identity of the three different cleave planes we measured: Most measured surfaces had a triangular atomically resolved structure (Fig. 4.1D,E), while rarely we found an hexagonal-like structure (Fig. 4.1F). We could only find a single well ordered instance of the hexagonal structure amidst a highly steppy region (Fig. B.1A). We identify the hexagonal atomic structure as the Co layer. The kagome structure formed by the Co surface atoms appears hexagonal in topography as we could not resolve individual atoms within the corner-shared triangles. This is not due to lack of sufficient spatial resolution but due to the strongly hybridized nature of these Co triplets (see (Yin et al., 2018) as another example). The triangular structure terminations were identified with either the Sn or the S triangular layers, which needs further resolution. The triangular surfaces are found with one of two dominant defect structures: either adatoms (Fig. 4.1D) or vacancies (Fig. 4.1E). The distinct defect character repeatedly corresponds to distinct $dI/dV$ spectra (different also from Co spectrum, all shown in Fig. 4.1G-I). We resolved the identity of the triangularly ordered surfaces with the vacancies by imaging a crystallographic step edge between it and the Co terrace shown in Fig. B.1B. Aligning the upper terrace identified as Co with the Co layer in the crystal structure and comparing the height of the step edge (1.72 nm) shows that the adjacent lower terrace falls between Co and S terminations (Fig. B.1B). Accordingly, S is the closest non-Co layer that identifies the triangular termination with vacancies as the S termination (and single atomic layer above the Co layer). The other triangular lattice with adatoms is therefore identified as Sn terminated surface. Importantly, this topographic termination identification scheme is backed by the detailed QPI study of all respective surface terminations. Intriguingly, this Co-S step edge was the only step edge we found separating two terraces of distinct atomic character within a single field of view. The height of all other step edges encountered were of a multiple of a third of a unit cell, thus separating the same atomic terminations. Fig. B.2A depicts S terminated region hosting a few atomic vacancies. It displays four terraces, separated by 1/3 of a unit cell height (Fig. B.2B). Average $dI/dV$ measurements (Fig. B.2C) taken on the different terraces indeed show the typical spectra (Fig. 4.1H) associated with the S termination. The marked cleave planes...
B.4 Raw Fourier transformed $dI/dV$ data measured on Sn, Co and S terminated surface

**Figure B.1:** Co-S step height analysis. (A) Topography taken on a rough terrain revealing a Co terminated terrace. Inset: blown up topography of the flat Co terrace. (B) 3D Topographic image ($I_{set} = 150\text{pA}, V_{bias} = 70\text{meV}, 32 \text{nm} \times 32 \text{nm}$) taken on $\text{Co}_3\text{Sn}_2\text{S}_2$ surface which includes a step between Co and S surfaces (red dashed square in A). Top surface: typical $dI/dV$ point spectrum of the Co terminated surface measured at the blue dot. Bottom surface: typical $dI/dV$ point spectrum of the S terminated surface, measured at the yellow dot. Superimposed crystal structure of $\text{Co}_3\text{Sn}_2\text{S}_2$ reveals the cleave planes (blue, yellow, red indicate the Co, S and Sn atoms, black solid line indicates the unit cell of the crystal).

illustrated in Fig. B.2B are based on two findings. The first is related to the step height analysis performed in Fig. B.1B, revealing a cleave plane for the S atoms below the Sn layer. The second is related to the scarcity of Co terminated surfaces, implying it is an energetically unfavorable cleave plane, in consistence with theoretical calculations (Xu et al., 2018). Note, that this further implies it is less likely to cleave along the plane of the S atoms above the Sn layer.

**B.4 Raw Fourier transformed $dI/dV$ data measured on Sn, Co and S terminated surface**

The Fourier transformed $dI/dV$ data shown in the main text was symmetrized to represent the $C_3$ symmetry of the crystal, and in order to enhance the signal to noise ratio of the QPI data. For each used data set, we validated that no extra features are added or erased by the applied symmetry analysis. Fig. B.3 illustrates the raw Fourier
Figure B.2: S terraces. (A) Topography showing four S terraces (B) Height profile along the red line in (A) revealing step edges of 1/3 unit cell high. (C) Average $dI/dV$ taken on the four different terraces in vacancy free regions (see colored squares in A). Measurements are shifted for clarity.
B.5 Spin and orbital character from ab initio calculation

The QPI data shown in the text was compared to JDOS calculation, and not to spin-selective scattering probability (SSP). This is because in this compound within the ferromagnetic phase the spins of all relevant bands are fairly polarized, as seen in ab-initio calculation in Fig. B.4. As a result, the spin degree of freedom does not add selection rules by limiting scattering processes. This can be seen in the band structure (Fig. B.4B) where indeed near the Fermi energy the bands are composed mostly from a single spin along $S_z$. Our ab-initio calculations also resolve the orbital character of the different surface bands of the different surface terminations. Representative such band structures at energy cuts corresponding to prominent QPI patterns are shown in Fig. B.5. It is evident that the surface bands consist of a hybridized mixture of different

![Figure B.3: Raw QPI data on Sn, Co and S surface.](image)

(A-B) Sn terminated surface QPI at V=-10 and 7.5 meV. (C-F) Co terminated surface QPI at V=70, 55, -5 and 10 meV. (G) S terminated surface QPI at V=100 meV.

transformed $dI/dV$ data of the corresponding symmetrized results appearing in Fig. 4.2,4.3,4.4.
Figure B.4: **Band structure spin texture.** (A) Band structure without Spin-orbit coupling (SOC). The black and red lines represent spin up and down channel, respectively. (B-D) Band structures with SOC when spin projected into z (B), x (C) and y (D) directions ($S_z$, $S_x$ and $S_y$). The red and blue lines represent spin up and down. Bands near Fermi energy are mostly composed by $S_z$ with spin up.

Orbitals both on the Sn surface (Fig. B.5A-C) and on the CoSn surface (Fig. B.5D-I), apart from the S termination which is singly dominated by p-wave orbitals (Fig. B.5J and K). In particular, the DOS contributed by the p-orbital of the Sn termination (Fig. B.5B) is as inclusive as the one obtained by integrating the contributions of all atomic orbitals in the unit cell (Fig. B.5C). The same is true for both the Co and Sn s-orbitals (Fig. B.5D and G, respectively) compared to the full unit cell calculation (Fig. B.5I) of the CoSn surface termination. Consequently, the orbital composition does not provide a straight forward scattering selection rules that would limit available scattering processes either. We thus use the full JDOS calculation for comparison with QPI and use simple intensity thresholding for singling out the dominant scattering processes among all available ones.

### B.6 The electronic structure of the Sn terminated surface

The QPI features shown in Fig. 4.2C,D can be compared to JDOS calculations. Figure B.6A describes a JDOS calculation using the DOS(k) plotted in Fig. 4.2E. The hexagon feature we identify in the QPI, both near q=0 and the Bragg peaks, agrees quantitatively with the one observed in the JDOS. We were further able to identify in our JDOS
B.6. The electronic structure of the Sn terminated surface

**Figure B.5:** Band structure orbital composition (main panels show summed contributions of individual orbitals that are shown in the insets). (A-C) Sn terminated surface calculation at $E=7.5\text{meV}$, Sn $s$ and $p$ orbitals shown in A-B and full projected unit cell shown in C. (D-I) CoSn terminated surface calculation at $E=10\text{meV}$, Co $s$, $p$, and $d$ orbitals shown in D-F, Sn $s$ and $p$ orbitals shown in G-H and full projected unit cell shown in I. (J-K) S terminated surface calculation at $E=100\text{meV}$, S $p$ orbitals shown in J and full projected unit cell shown in K.

calculation (Fig. B.6B) the connecting line feature appearing in Fig. 4.2D. Its existence confirms the identification of the relevant bands (pockets connected with an open pink arrow in Fig. 4.2F) associated with such a feature. Our JDOS calculation indeed finds the appearance of the additional connecting lines pattern due to accidental quasi-nesting conditions. However, it appears at about 40 meV above the energy at which they appear in our QPI measurement. Nevertheless, as the schematic illustration in Fig. 4.2F shows, these arise from quasi-nesting conditions that are highly susceptible to the exact curvature of the triangular pockets, and can be easily met at a range of energies by slight modifications of the pocket curvature, which is beyond the accuracy of our ab-initio modeling. We also provide in Fig. B.6C an additional analysis method to the one described in Fig. 4.2I for highlighting the second dispersing QPI mode that we attribute to time reversal symmetry breaking. Here we fit a Lorentzian of the form $L(q) = P1/((q - P2)^2 + P3) + C$ to every energy slice in the $\Gamma\sim\bar{K}$ QPI linecut. By subtracting the fit from the raw data we again identify two branches corresponding to the two different scattering events (Fig. 4.2I), marked in the pink and red arrows. Representative raw QPI profiles are plotted in Fig. B.6D, clearly showing the two dispersing peaks. Lastly, we show an additional measurement taken on a remote Sn terminated surface, different than the one described in the main text, and find both dispersing features are indeed reproduced. Fig. B.6E shows a constant energy Fourier transformed
**Figure B.6:** Sn electronic structure. (A) JDOS calculation for DOS(k) at E=7.5 meV. Pink arrow corresponds to the hexagon feature observed in the QPI (Fig. 4.2C). (B) JDOS calculation for DOS(k) at E=30 meV revealing the connecting line observed in the QPI (Fig. 4.2D). (C) Lorentzian subtracted $\Gamma K$ QPI linecut showing the energy dispersion of the two different scattering events described in Fig. 4.2I. (D) Intensity profiles extracted from Fig. 4.2G from QPI data prior to background substraction. Two visible peaks correspond to the two dispersing scattering processes from time reversal symmetry broken K and K' valleys shown in Fig. 4.2I. (E) Fourier transform of a $dI/dV$ map taken at a different location on the Sn terminated surface (V bias = −7.5 meV) showing two hexagon features, intense one around $q = 4 nm^{-1}$ and a fainter one around $q = 3 nm^{-1}$. (F) Energy-Momentum cut of the QPI along $K - \Gamma M$ direction for the map taken at E.

$dI/dV$ map, and nicely finds two sharp concentric hexagon edges along the $\Gamma K$ direction, corresponding to the two different scattering processes. We further show the energy evolution of these processes (Fig. B.6F) by following their dispersion along the $\Gamma K$ QPI linecut. We find this different measurement to be in good agreement with the data shown in Fig. 4.2G.

**B.7 The electronic structure of the Co terminated surface**

The QPI evolution of the less probable Co termination presents a complex structure illustrated in Fig. 4.3. Here we provide our detailed resolution of it. The QPI patterns appear both within and beyond the momentum space region bounded by the
Bragg peaks and respectively involve both intra- and inter-Brillouin zone scattering processes, providing a complementary view of the allowed scattering. We decompose the QPI in Fig. 4.3H into four energy evolving patterns (marked by arrows of different colors in Fig. 3.4(i)). The first is a honeycomb like structure, consisting of one central hexagon around the $\Gamma$ point, and six encircling hexagon replications centered on the first order Bragg peaks (orange hexagon in Fig. 4.3H). The second feature is the broad spot at each vertex of a Hexagon (blue triangles in Fig. 4.3H). Both features are nicely reproduced in JDOS calculations (Fig. 4.3G). We attribute the hexagonal QPI to inter-pocket (orange arrow, Fig. 4.3D(i)) scattering and the broad spot to scattering between the central hexagon band at $\Gamma$ and the $\mathbf{K}$ pockets (blue arrow, Fig. 4.3D(i)). The third feature is the high intensity spot around the Bragg peak (Fig. 4.3H, $E=-5$ meV), which we track to intra-hexagon band scattering (green arrow, Fig. 4.3D(i)). Lastly, the fourth feature is the diamond like shape beyond the Bragg peak (Fig. 4.3H, $E=10$ meV). This scattering event is attributed to intra-$\mathbf{K}$-pocket scattering (pink arrow, Fig. 4.3D(i)). We now follow the QPI energy evolution by comparing it to the energy evolution of the band structure along $\mathbf{K}'\rightarrow\mathbf{\Gamma}\rightarrow\mathbf{K}$ direction presented in Fig. B.7A. Three relevant trends occur with increasing energy:

1. The central hexagonal hole band shrinks, and thus the broad spots shift to higher momentum (green and blue dotted lines in Fig. 4.3C respectively). In principle, we expect the dotted green lines to appear around $q=0$ and to shrink in momentum with increasing energy. In Fig. 4.3C, however, we do not follow the features around $\Gamma$ (as they are masked by the long wavelength peak around $q=0$), but rather their replications around the Bragg peaks (see chapter 3.2.4, for details about replications in QPI). Hence, their size evolution in momentum transfer is defined with respect to the Bragg peak momentum, which is shifted by a reciprocal lattice from $q=0$. This higher momentum transfer scattering event is illustrated in the calculated DOS(k) (Fig. B.7B) by multiple green arrows.

2. The electron pockets around $\mathbf{K}$ expand with increasing energy, hence the intra-pocket scattering grows (pink dotted line within low energy range (i) in Fig. 4.3C and pink arc in Fig. B.7C and D). Above a certain energy, this scattering process becomes larger than intra-hexagon scattering. This allows the visualization of the diamond like QPI feature (pink arrow in Fig. 4.3H and Fig. B.7B) above $E=7.5$ meV.

3. The electron pockets around $\mathbf{K}'$ expand with increasing energy, and therefore do not allow nested inter-pocket scattering along $\Gamma - \mathbf{M}$ (orange dot in Fig. 4.3C and orange arc in Fig. B.7C and D).
Figure B.7: Co electronic structure. (A) Projected band structure evolution on Co terminated surface along the $K'\Gamma K$ direction (B) Calculated DOS(k) at E=10meV featuring several Brillouin zones. Arrows of same color represent scattering to higher Brillouin zones, and reflect scattering events observed in Fig. 4.3C. (C-D) Calculated JDOS and measured QPI at E=10 meV. Color shapes reflect corresponding colored arrows in DOS(K) shown in B. (E) JDOS linecut along the $\Gamma M$ direction. Dotted lines correspond qualitatively to dotted pink lines in Fig. 4.3C.

The expansion of the K and K’ pockets persists at higher energies (Fermi-arcs and K’ pockets open up thus no longer visible in the momentum cut presented in Fig. B.7A, see also 2D slices in Fig. 4.3D(i)-(ii)). This gives rise to the dispersing pink dotted line within the intermediate energy range (ii) in Fig. 4.3C describing two equal momentum transfer scattering - intra-Fermi-arcs (pink arrow in Fig. 4.3D(ii)) and intra-K’-pocket . Upon the opening of the K’ pockets and their apparent merging to the Fermi arcs in the form of the C-bands, a rather non-dispersing branch appears, depicted in the pink dotted line within the higher energy range (iii) in Fig. 4.3C. The flat segments of the C band give rise to the broadening of the feature shown in Fig. 4.3E. Both these scattering processes are qualitatively traced in a $\Gamma M$ JDOS linecut shown on Fig. B.7E.
B.8 The electronic structure of the S terminated surface

In addition to the QPI patterns discussed in Fig. 4.4, we also observed QPI patterns in other energy slices that occur due to the scattering of electrons from surface bands. Fig. B.8A describes the low energy QPI cut along $\Gamma - M$, in which we recognize three main features: slightly dispersing band marked by green dotted line, continuum of scattering vectors marked by a red dot and a swiftly dispersing band marked by an orange dotted line. We first identify the origin of the QPI marked in green. At energy -60 meV, shown in Fig. B.8B, we observe an arrow-shaped QPI feature near the Bragg peak. The scattering processes related to this feature have been identified as the scattering between the petals of a flower-like band located at K to the inner triangle of the other flower-like band located at the other K point (green arrow in Fig. B.8C). We confirm these scattering processes by comparing it with the corresponding JDOS (Fig. B.8D). Indeed both the triangles and flower bands slightly shrink with decreasing the energy. Decreasing of the energy further gives rise to the merging of the circular band (gray arrow in Fig. B.8C) with its higher Brillouin zone replications. At energy -130 meV (Fig. B.8E), we observe an elongated feature which we associate with the merging of the aforementioned bands (gray arrow in Fig. B.8F), and the resulting nesting along the $\Gamma - M$ direction (red arrows in Fig. B.8E and F, and red dot in Fig. B.8A). This QPI feature is also apparent in the corresponding JDOS (Fig. B.8G). Intriguingly, the appearance of these QPI patterns resembles the one we found on the Sn surface termination at a different energy (Fig. 4.2D). Lastly, we observe a high intensity (orange dotted line in Fig. B.8A) band near the Bragg peak. At energy -360 meV (Fig. B.8H) we observe an extended QPI pattern near the Bragg peaks (orange arrow from Bragg peak), resulting from the scattering processes between the inner small circular band near the $\Gamma$ point and a hexagon band next to it (orange arrow in Fig. B.8I). Calculated JDOS (Fig. B.8J) agrees well with our results. We confirmed the dispersion of this QPI feature to be consistent with the expansion nature of the hexagon band with decreasing energy.
**Figure B.8: S electronic structure.** (A) Energy-momentum QPI cut along the $\Gamma - M$ direction. (B-D) Measured QPI at $V=\text{-60 meV}$ with corresponding DOS($k$) (C) and calculated JDOS (D) representing green band in A. (E-G) Measured QPI at $V=\text{-130 meV}$ with corresponding DOS($k$) and JDOS (F,G respectively) at red dot (A) energy value. (H-J) Measured QPI at $V=\text{-360 meV}$ with corresponding DOS($k$) (I) and calculated JDOS (J) portraying the orange band in A.