A COMPLETE ANALYSIS OF THE INTRINSIC PROPERTIES OF THE TOPOLOGICAL STATES IN Bi$_2$Se$_3$ BY ANGLE RESOLVED PHOTOEMISSION SPECTROSCOPY

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Eric J. Reichwein

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Abstract

A Complete Analysis of the Intrinsic Properties of the Topological States in Bi$_2$Se$_3$

By Angle Resolved Photoemission Spectroscopy

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We have conducted Angle Resolved Photoemission Spectroscopy (ARPES) data on Bi$_2$Se$_3$ using Stanfords Synchrotron Radiation Lightsource (SSRL). We used energy distributed curves (EDC) and momentum distributed curves (MDC) to analyze the ARPES intensity spectrum. The Fermi velocity was temperature dependent $v_f \approx 4.0 \text{ eV/Å-3.5 eV/Å}$, that leveled off at $3.5 \text{ eV/Å}$ once temperature increased to $\approx 175K$. By comparing MDC and EDC extremely correlated Fermi liquid theoretical imaginary part of the complex self-energy (with fixed experimental parameters), we observed a sharp increase in the MDC’s self-energy below the bottom of the bulk conduction band. Hence, these quasiparticle interactions were not electron-electron interactions. They could only by impurity or electron-phonon interactions, but since we had low energy Dirac points and high-quality data we assume the increase to be caused by phonon coupling. To our knowledge, this is the first observation of strong intra-surface electron-phonon coupling in Bi$_2$Se$_3$ as well as the lowest reported phonon coupling constant $\lambda = 0.049 \pm 0.007$. 

Contents

Dedication v

Acknowledgements vi

1 Introduction 1
   1.1 Angle Resolved Photoemission Spectroscopy (ARPES) 3
       1.1.1 ARPES Explained 6
   1.2 ARPES Theory: Three Step Model, Sudden Approximation, and Green’s Functions 11
       1.2.1 The Three Step Model 11
       1.2.2 Sudden Approximation 12
       1.2.3 Greens Function Formalism in the Single Particle Spectral Function 12
       1.2.4 ARPES Line Shape Analysis Basics 15
   1.3 Qualitative Topological Insulator Theory 17

2 History and Previous Work 20
   2.1 A Brief History of Topological Insulators 21
   2.2 Previous Work on Bi$_2$Se$_3$ 23
       2.2.1 Quasiparticle Scattering in Bi$_2$Se$_3$ 23
       2.2.2 Weak Electron-Phonon Interaction in Bi$_2$Se$_3$ Surface 26

3 Data and Analysis 29
   3.1 Initial Quasi-Particle Interaction Observation 31
   3.2 EDC Line Shape Analysis 32
       3.2.1 Shastry’s Extremely Correlated Fermi Liquid Model 35
   3.3 MDC Line Shape Analysis 36
   3.4 MDC and EDC Comparison Through $\Sigma_{\text{Im}}$ 37
       3.4.1 Weak Electron-Phonon Coupling of Topological Surface States 40
   3.5 Relation to High Temperature Superconductor Cuprates 41

4 Conclusion 44
   4.1 Future Work 44
To my parents,

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Introduction

In 1887, Heinrich Hertz’ discovered the famed photoelectric effect[1] while investigating the spark-gap generator (earliest known radio receiver). He noticed that the sensitivity increased as the spark-gap generator was illuminated by ultraviolet light, which is caused by the ejection of electrons into the gap. A few years later (after J.J. Thomson’s discovery of the electron), Thomson[2] then offered further insight into the photoelectric effect by suggesting the light “pushed” electrons\(^1\). In 1905, Albert Einstein physically and mathematically explained the photoelectric effect[4] by Equation 1.1.

\[
E_{\text{bind}} = hf - E_k - \Phi
\]  

(1.1)

Where \(E_k\) is the kinetic energy of the photoelectron ejected, \(h\) is Planck’s constant, \(f\) is the frequency of the incoming light, \(\Phi\) is the work function of the material, and \(E_{\text{bind}}\) is the binding energy of the electron. Einstein described this effect purely by en-

\(^1\)Phillip Lenard also made significant contributions to this effect as well, and was awarded the 1905 Nobel Prize for his work on Cathode Rays[3]. He is known for conducting the earliest true experiments on the photoelectric effect.
ergy conservation but neglected momentum conservation. This is because the momentum is not “conserved” in the case of the photoelectron. The simplest way to think of momentum conservation is through symmetry\(^2\) principles of quantum mechanics. The translational symmetry is broken only in the \(z\)-direction at the surface because the potential goes from periodic to a constant zero\(^3\) (for the \(z\)-direction), but remains periodic for the \(x, y\)-directions. See Figure 1.1 for diagram of symmetry breaking in the \(z\)-direction and, hence, “no” momentum conservation in the \(z\)-direction.

---

\(^2\)Translational symmetry is related to momentum conservation, while time symmetry is related to energy conservation, and rotational symmetry is related to angular momentum conservation.

\(^3\)Technically, a constant function is periodic but the potential in a crystal goes from a negative value to negative infinity (at location of ion) which is very much different from a constant zero. This can also be thought of concepts of “least periods”. The potential inside the crystal has a defined period, but the potential outside does not.
It is unclear if Einstein neglected momentum conservation knowingly or merely forgot. The momentum is conserved if you include the crystal momentum $\vec{G}$, but since quantum theory of solids had not yet been developed, we can forgive him for neglecting conservation of momentum in his derivation. In our derivations the lack of momentum conservation will be handled through a term we call the inner potential $V_0$, which has the same affect as the crystal momentum approach.

1.1 Angle Resolved Photoemission Spectroscopy (ARPES)

Angle Resolved Photoemission Spectroscopy (ARPES) is an essential tool for probing the electronic structure of materials. By shooting high energy photons (Ultraviolet to X-Ray) at the sample and analyzing the photoelectrons ejected we are able to probe the electronic structure of the sample. This is done by utilizing the photoelectric effect and momentum conservation. The momentum is determined by knowing the kinetic energy of the photoelectron and the angle which it is ejected from the sample. In Figure 1.2, I present all of the angles needed for the explanation of ARPES. There are two main angles of concern are $\theta$ and $\varphi$, which are directly related to the electron’s momentum. The secondary angles of $\theta_p$ and $\alpha$ are the photons incident angle and the angular resolution of the ejected electrons. These angles are very important in knowing the angular and energy resolution of the apparatus, but for the purpose of the following derivations they will not come into play in this paper. However, they are actually needed for more rigorous derivations and calculations, which are much too involved to present here in this thesis.
The following derivation is guided by multiple sources\cite{6-9}. Let us derive the momentum outside of the sample first. Since the electron is in vacuum (after being ejected from sample), it is considered a free electron (dispersion for free particle is $E = \frac{\hbar^2 k^2}{2m}$). This gives that the momentum is simply

$$\vec{p}_{\text{out}} = \hbar \vec{k}_{\text{out}} \rightarrow \vec{k}_{\text{out}} = \frac{\sqrt{2m_e E_k}}{\hbar} \approx 0.512 \sqrt{E_k}$$ (1.2)

Where the value 0.512, in Equation 1.2, comes from evaluating the constants $\left(\frac{\sqrt{2m_e E_k}}{\hbar}\right)$ in terms of Å and eV. Momentum in this case units of Å$^{-1}$ and energy in the units of eV. Equation 1.2 is entirely in vector form, so we need to use the angles shown in Figure 1.2 to determine each of the components of the momentum (or equivalently wavevectors $\vec{k}$). This is done by switching from Cartesian to polar coordinates produces Equation 1.3.
\[
\begin{bmatrix}
k_{\text{out},x} \\
k_{\text{out},y} \\
k_{\text{out},z}
\end{bmatrix}
= \begin{bmatrix}
\sqrt{2m_eE_k \hbar} \cos \varphi \sin \theta \\
\sqrt{2m_eE_k \hbar} \sin \varphi \\
\sqrt{2m_eE_k \hbar} \cos \varphi \cos \theta
\end{bmatrix}
\] (1.3)

Where \(m_e\) is mass of the electron and \(E_k\) is the kinetic energy of the photoelectron. The angles \(\varphi\) and \(\theta\) are measured with respect to the \(z\) and \(x\) axis respectively. From conservation of momentum and energy principles we can determine the values of the electron momentum inside the sample. The \(x\) and \(y\) components of the momentum are conserved as the photoelectron is ejected from the sample. However, the \(z\)-component is not conserved. Due to diffraction at the surface \(k_z\) is altered by we call “inner potential” \(V_0\). To model the potential function responsible for the inner potential we treat it as a step function, negative potential in the material and zero outside, which can be seen in Figure 1.1. The change in potential only occurs in the \(z\)-direction (normal to surface) and hence there is only a retarding force in the \(z\)-direction. According to Newton’s Law this means that the momentum of the electron will be altered, given by \(F = -\frac{\partial V}{\partial z} = \frac{dp}{dt}\). However, it is easier to analyze it in terms of the inner potential instead Newton’s 2nd Law. This effect causes the measured \(k_{z,\text{out}}\) to be less than the \(k_{z,\text{in}}\), which is easily modified by the following term in Equation 1.4.

\[
\begin{bmatrix}
k_{\text{in},x} \\
k_{\text{in},y} \\
k_{\text{in},z}
\end{bmatrix}
= \begin{bmatrix}
k_{\text{out},x} \\
k_{\text{out},y} \\
\sqrt{k_{\text{out},z}^2 + \frac{2m_eE_k \hbar}{\hbar} V_0}
\end{bmatrix}
\] (1.4)

Equation 1.4 is the main result we need because this tells us the momentum of the electrons in the sample from simply knowing the kinetic energy and ejection angles of the
photoelectron.

1.1.1 ARPES Explained

The light is produced by accelerating electrons in the undulator of the synchrotron light source\(^4\). It then gets focused by a series of mirrors, gratings, and apertures as seen in Figure 1.3. The light then enters the ultra high vacuum chamber and is aimed at the sample. We ideally would like the beam to make a spot as small as possible on the sample, but this is not the case in reality. Our beam spot is on the order hundreds of micrometers, but recent developments have been able to get beam size down to the nanometer scale\(^5\).

![Figure 1.3: The experimental schematic of the ARPES instruments at Stanford’s Synchrotron Radiation Lightsource\(^8\).](image)

As the high energy photons bombard the sample, the photoelectrons fly out in a spread of angles. This is a very complicated process with many interactions and parameters, so we take a three step model and sudden approximation approach, which will be explained further later in this paper. We first focus these photoelectrons to the hemispherical detector (or electron spectrometer) entrance slit, as seen in Figure 1.4. They then enter the hemispherical analyzer for momentum and energy determination. The electrons then exit

\(^4\)This is a non-thermal radiative process. Thermal radiation is like a blackbody spectrum where photons get produced by electrons changing energy levels.

\(^5\)This is referred to as nanoARPES, or nARPES.
the hemisphere and get amplified for the coupled device detector (CCD) detector.

Figure 1.4: The experimental schematic of hemispherical electron spectrometer[6]. See text for discussion.

The spectrometer is made of two metal hemispheres held at different potentials. The resulting electric field in between the hemispheres separate the electrons of different kinetic energies and momenta (or equivalently angles). Depending on the potentials on each hemisphere, we can determine the pass energy of the electrons. Higher energy electrons end up closer to the larger hemisphere, while slower electrons end up closer to the smaller hemisphere. The pass energy is the allowed kinetic energy range that can pass the end detector. If they are not in the pass energy range then they will be absorbed by either of the two hemispheres. The electrons get multiplied by a micro-channel plate (similar to night vision), then those hit a phosphor screen which create photons. Once the photons are created they excite a single bin (of specific energy and angle/momentum) on the CCD.
The result is a beautiful 2D image of the electron dispersion. Our energy resolution was 11 meV and angular resolution was approximately 0.1°. We can stack these to obtain 3D images or movies through different binding energies.

Figure 1.5: Energetics indicating the photoelectric effect and subsequent detection by spectrometer[6].

In Figure 1.5 we see the process of ARPES explained in terms of energetics. The spectrometer and sample are in electrical contact which puts there chemical potential (or equivalently the Fermi energy) at the same level. However, in general, the spectrometer and the sample have different work functions. This produces a discrepancy in the kinetic energies measured, since it is relative to the spectrometer, not the sample. Therefore, we must determine the work function of the spectrometer prior to taking measurements using a known sample i.e. gold or platinum have very well known work functions[6]. Once we
know the work function of the spectrometer we can determine the binding energy of the electrons. Since we know the photon energy $h\nu$, the samples work function and the kinetic energy we can apply Equation 1.1 and obtain the binding energy.

Figure 1.6: Density of states of the spectra. The solid is for the bottom left figure is the ideal curve matching the top right graph. The dashed line is for the nonzero temperature distribution, which allows for electrons be above the Fermi edge[6].

The electrons inside the sample will have some binding energy and associated momentum, which make up an individual state. We average the spin values ($\vec{\sigma}$) so we don’t need to take this extra parameter into account$^6$. When the electron with a specific

$^6$Spin-and-ARPES is a photoemission technique which the spins are inherently tied to momentum, such as magnetic materials and topological insulators, can be probed and analyzed. This is implemented by
momentum and energy gets excited by the photon it makes a transition to higher energy level, as shown in Figure 1.6. At high enough energies, the levels are sufficiently close together to consider the available states as continuous[6]. Due to many reasons including non-zero temperature and many-body effects like electron-electron and electron-phonon interactions, as illustrated in Figure 1.7, we see a spread in energies (and crossing the Fermi edge). If there were no many-body effects or other interactions (purely single particle physics) then we would see infinitely sharp peaks for each momenta (or equivalently angle $\theta$). In Figure 1.6, each peak corresponds to a different angle and corresponding energy in the map.

![Figure 1.7: Two common electronic interactions during photoemission.](image)

attaching a large magnetic material at the detector which will deflect spin up electrons one way and spin down electrons in the opposite directions.
1.2 ARPES Theory: Three Step Model, Sudden Approximation, and Green’s Functions

We will now present the most basic treatment of the mathematical formalism behind ARPES and photoemission in general. There are three main topics in ARPES theory that we will show: The three step-model, sudden approximation, and Green’s Functions.

1.2.1 The Three Step Model

The three steps of the single electron photoemission model are excitation, transport, and escape. This can be seen in the photoelectric effect equation (Equation 1.1) by the three terms.

1. **Excitation**, electron absorbs a photon and transitions to an unoccupied state: \(+h\nu\)

2. **Transport**, with the absorption of the photon and subsequent excitation, the electron has overcome the binding energy and is free to move to the surface: \(-E_{\text{bind}}\)

3. **Escape**, electron leaves the material by surmounting the work function: \(-\Phi\)

The resulting left over energy (when electron escapes into vacuum) is purely kinetic energy as shown in Equation 1.1. This is the basic idea of the three step model. The momentum of the photon is much less than that of the electrons, so the momentum in the transition to an unoccupied state is essentially unchanged[7]. A illustration is shown in Figure 1.8 to show the single electron three step model. This model was thoroughly developed by C.N. Spicer and W.E. Berglund in 1958[10] and 1964[11], but can be dated back to H. Fan in 1945[12]. This model is especially useful for dealing with 2D (or surface states) of materials, such as TI’s and graphene.
1.2.2 Sudden Approximation

The sudden approximation is essentially saying that the final state of the photo-electron does not interact with the remaining system. So the three step process can be assumed to happen instantaneously (or suddenly) before the remaining electron system has time to react[7]. If we assume this then our calculations become significantly easier. For convenience, I will be setting $\hbar \equiv 1$ so that $\omega$ is the energy and $\vec{k}$ is momentum (which I have previously referred to as momentum).

1.2.3 Greens Function Formalism in the Single Particle Spectral Function

Taking the single particle Greens Function approach[6–9] the intensity of the spectra is (Pulled from the Green’s function formalism by[14–16])
\[ I_\vec{k}(\omega) \propto \sum_{i,f} |\langle \psi_f | M | \psi_i \rangle|^{2} A_\vec{k}(\omega) f(\omega) \] (1.5)

Where, in Equation 1.5, \( M \) is the perturbing component of the complete Hamiltonian, \( \psi \) is state of a single photoelectron, \( \omega \) is the energy of the photoelectron\(^{6-9}\). For notation simplicity I will use the convention of \( \langle \psi_f | M | \psi_i \rangle = M_{if} \). The value for \( f(\omega) \) in Equation 1.5 is given by Equation 1.6,

\[ f(\omega) = \frac{1}{e^{\omega/k_B T} + 1} \] (1.6)

and is called the Fermi-Dirac distribution, which tells us the average number of particles in state and ranges from 0 to 1\(^{7}\). The value for \( A_\vec{k}(\omega) \) in Equation 1.5 is given by Equation 1.7,

\[ A_\vec{k}(\omega) = \frac{1}{\pi} \text{Im} G(\vec{k}, \omega) \] (1.7)

where \( G(\vec{k}, \omega) \) is the single particle Green’s function. The simplest way to express this Green’s function is arguably in the form of Dyson’s equation

\[ G(\vec{k}, \omega) = \frac{1}{\omega - \epsilon(\vec{k}) - \Sigma(\vec{k}, \omega)} \] (1.8)

where \( \Sigma(\vec{k}, \omega) \) is the electrons self-energy and \( \epsilon(\vec{k}) \) is the standard dispersion relation (or band energy). The self-energy is way of accounting for the electrons energy due to the interactions of its surrounding system. Andrea Kamimski et. al.\(^{17}\) showed how the self-energy can be extracted directly from ARPES data, which we use in our analysis.

\(^{7}\)This is with the convention assuming that the Fermi energy, \( E_f \) or \( \mu \), is taken to be zero. If not the case, then we would have to change \( \omega \longrightarrow \omega - \mu \)
The best way to understand the self-energy is by utilizing Feynman Diagrams, but it is unnecessary for the point of this thesis, so it will be omitted. Refer to Quantum Theory of Many-Particle Physics by A.L. Fetter and J.D. Walecka[18] for further mathematical insight into this many body effect. The main thing to get from \(\Sigma(\vec{k}, \omega)\) is that it is a complex value \((\Sigma = \Sigma_{\text{Re}} + i\Sigma_{\text{Im}})\). The real part tells you the change of the group velocity while the inverse of the imaginary part tells you the lifetime of the electron, and is related to the full width half maximum (FWHM) of the line shape distributions[6]. \(\Sigma_{\text{Im}}\) is the more important quantity for our purposes. It is often referred to as the imaginary part of the complex self-energy, but I will refer to it simply as self-energy because I do not refer to the real part for the entirety of this paper.

Getting back to the original question: explaining Equation 1.5 in terms of ARPES. Now that we have all individual parts explained, we can make some approximations to simplify our expression. Taking into account all the interactions in an electron system, the value of interaction Hamiltonian \(M\) is given by[7]

\[
M = -\frac{e^2}{2m_e c} \left( \vec{A} \cdot \vec{p} + \vec{p} \cdot \vec{A} \right) - eV + \frac{e^2}{2m_e c} \vec{A} \cdot \vec{A} \tag{1.9}
\]

Where, in Equation 1.9, \(\vec{A}\) is the vector potential, \(\vec{p}\) is the electrons momentum, \(V\) is the electric potential, and \(m_e\) is the electrons mass. Ignoring the inhomogeneity of the electron system\(^8\) we find that we are actually working in the Coulomb gauge \((\nabla \vec{A} = 0)\) which means \(V \rightarrow 0\) (for more information of this subject please see David J. Griffith’s phenomenal textbook: Introduction to Electrodynamics). By neglecting multi-photon processes\(^9\) we

---

\(^8\)The inhomogeneity of the electron system causes the need for local field corrections. By taking the average we significantly improve the efficiency of our computations.

\(^9\)Multi-photon processes are when multiple photons interact with the same electron. This phenomena has an extremely low probability of occurring. Refer further to references[19]
make $\vec{A} \cdot \vec{A} = 0$. The last simplification we make is that the wavelength of the photons ($\sim 1000\text{Å}$) is much larger than the atomic spacing ($\sim 0.1 \text{Å}$), such that $\vec{A}$ is constant (independent of $\vec{r}$). This means that $\vec{A}$ and $\vec{p}$ commute ($[\vec{A},\vec{p}] = 0$). Combining all of these approximations we find our interaction Hamiltonian gets reduced down to

$$M = \frac{e}{m_e c} \vec{A} \cdot \vec{p}$$

(1.10)

With the simplification from Equation 1.9 to Equation 1.10 we now know the ARPES spectrum intensity $I(\vec{k}, \omega)$. This will be the keystone of our analysis of our ARPES data on Bi$_2$Se$_3$ for the rest of this thesis. The two types of line shape analysis we do is Energy Distributed Curves (EDC) and Momentum Distributed Curves (MDC). For EDC we take fixed momentum values, $\vec{k}$, and look at how the electrons energies are distributed. Conversely, for MDC we fix the energy values, $\omega$, and look at how the electrons momenta are distributed. In most cases, researchers have found it sufficient to just look at MDC explicitly for analysis because EDC analysis is often more difficult to extract information from. In this paper we will take into account both MDC and EDC analysis equally and try paint a more coherent picture of the electronic structure of TI’s for further use to explore other more complicated materials like high $T_c$ superconductors.

1.2.4 ARPES Line Shape Analysis Basics

The following calculations have been guided by Professor G. H. Gweon, as well as references[7, 13]. Both the EDC and MDC are based on Lorentzian line shapes. Without loss of generality, we shall provide a basic MDC analysis mathematical framework. When looking at the distribution of momenta at certain energy values we assume that $\vec{k}$ is independent of
the self-energy $\Sigma(\vec{k}, \omega)$ and $|M_{if}|$. The dispersion is also approximated to be linear\textsuperscript{10} in $\vec{k}$, which means $\epsilon(\vec{k}) \approx v_g k$. The value of $k$ is assumed to be along the ARPES-cut direction.

Now we can rewrite out Intensity spectrum $I(\vec{k}, \omega)$ (Equation 1.5) as follows,

$$I(\vec{k} = k, \omega = \omega_0) = |M_{if}|^2 f(\omega_0) \frac{1}{\pi} \text{Im} \frac{1}{\omega_0 - v_g k - i \Sigma \text{Im}(\omega_0) - \Sigma \text{Re}(\omega_0)}$$  \hspace{1cm} (1.11)

This does not take on the conventional form of a Lorentzian, but by taking the imaginary part of Equation 1.11 we get

$$I(\vec{k} = k, \omega = \omega_0) = |M_{if}|^2 f(\omega_0) \frac{1}{\pi} \frac{\Sigma \text{Im}(\omega_0)}{[\omega_0 - v_g k - \Sigma \text{Re}(\omega_0)]^2 - [\Sigma \text{Im}(\omega_0)]^2}$$  \hspace{1cm} (1.12)

From this we can see that the FWHM is just $\Sigma \text{Im}(\omega_0)$ and is has a peak centered at $\omega_0 = v_g k + \Sigma \text{Re}(\omega_0)$. The EDC fits are based on Fermi Liquid Theory (FLS), developed by Professor Sriram Shastry, which account for the electron-electron interactions of the quasi-particles. However, at its core it too is a Lorentzian Distribution so we can assume the same basic mathematics for the EDC line shape analysis.

So, how does this analysis give us insight into the quasi-particle scattering? The key is the relationship between the width of the Lorentzian peak $\Delta k(\omega)$ and the quasiparticle scattering rate $\Gamma(\omega) = 2|\Sigma \text{Im}| = \Delta k(\omega) v_0(\omega)$, where $v_0$ is the group velocity\textsuperscript{20}.

Since we can extract the $\Sigma \text{Im}$ from the MDC analysis, we are able to determine the scattering rate through the previous equation.

\textsuperscript{10}This is only true when speaking of the momentum in the ARPES-cut direction, meaning in the direction that $\vec{k}$ data is taken.
1.3 Qualitative Topological Insulator Theory

The following section has been summarized after reading much literature on TI's, specifically references [21–25]. The topology in topological insulator refers to the topology of the Hamiltonian describing the electronic system, not to do with the real space geometry of the material. This is because the wavefunction is mapping from $k$-space to a Hilbert space, or a manifold in a Hilbert space. If we do a mapping to a complicated manifold in the Hilbert space then we are dealing with non-trivial topology, which is the case for TI's. For TI's the non-trivial topology of the Hamiltonian produces a characteristic band gap in the bulk of the material. On top of this, there is a topological invariant associated with the Hamiltonian (and the band gap) which cannot change if there is a band gap. That is why only at the surface does the band gap disappear.

Figure 1.9 (a) and (c) show the difference between topological insulators and conventional insulators, and their corresponding band diagrams. The key to the TI's surface state electron propagation mechanism is the following: The vacuum also has a band gap, but with trivial topology, where the TI has non-trivial topology. This means the Hamiltonian (or mapping from $k$-space to a manifold in the Hilbert space) must change topologies at the interface between the material and the vacuum. This causes the surface states to have no band gap and is the cause of the “Dirac cone” shape of the dispersion relation. The linear dispersion means the electrons have an effective mass of zero and have to be treated relativistically by the 4D Dirac equation. Remember that in the MDC analysis (Equation 1.11) we treated $\epsilon(k)$ as linear in $k$, this was due to the aforementioned fact that the theory predicts the dispersion to be linear. Although, in reality this is not the case for Bi$_2$Se$_3$, especially near the “Dirac point” (the point where the two surface state dispersions
meet in phase space, as seen in Figure 1.9. These “massless” electrons are referred to as Dirac fermions, and have very unique properties. Recall that massless particles like photons have linear dispersions because of Einsteins famous equation \( E = \sqrt{p^2c^2 + m^2c^4} \) becomes \( E = pc = \hbar k/c \) when \( m \to 0 \), where \( m, \hbar, c \) are the mass, Planck’s constant, and the speed of light, respectively, and are all constants.

Now, the topological transition produces electron states that have the momentum locked (orthogonally) to the spin. Spin is locked to the momentum by strong spin-orbit coupling in the material. The key here is that the spin-orbit coupling must be on the order of the size of the band gap for the locking to take place. Due the spin being locked to the momentum there are only spin polarized currents on the surface. Note, that this also means there is no net charge (conventional) current, only net spin current. Not only that, the surface states are also time reversal invariant usually (if there is no magnetic impurities) and for more mathematically rooted\(^ {11} \) reasons (such as time-reversal symmetry, berry phase and the Rashba effect), the currents are protected from perturbations that would usually cause current dissipation. This is why TI’s are front runners for applications in Spintronics and quantum information.

In Figure 1.9 (a) we see (left figure) shows a caricature of the electrons in a insulator, which are depicted as being in tightly bound orbits. In the right figure we see a band diagram of an insulator, which shows a large energy gap between the valence electrons and the conducting states. Figure 1.9 (b) depicts the QSH state. Due to spin-momentum locking and time reversal symmetry there are two directions of current (no net charge current, just spin current). All spin up electrons have positive momentum and all spin

\(^ {11} \)The mathematics are far above the undergraduate level, so I will omit the gory details, but please refer to references \([21, 24–26]\).
down electrons have negative momentum (or vice versa). This is also depicted in the band diagram with bulk band gap and two surface states. Where the surface states intersect (in $k$ space) is called the Dirac cone. The reason the that the quantum hall effect does not have spin polarized currents is due to the Rashba effect and momentum dependent spin-orbit coupling\[21\]. This effect is produced when an electron is traveling in an asymmetric potential with strong applied magnetic field. The asymmetric potential comes from the skewing of the periodic potential at the surface of the crystal. Now, there is an asymmetric potential in quantum hall insulators, but there is not the momentum dependent spin-orbit coupling (via a magnetic field), which separates the quantum hall effect from the QSH insulator.

![Figure 1.9: Different states of matter. (a) Standard insulator (b) A topological insulator (QSH insulator). Figure is taken from ref. [26].](image)

This was a very, very basic explanation of TI’s, and I encourage the reader to learn more through ref. \[21, 23, 26, 27\] as well as the numerous other resources online and in texts. Now that we have the basics of ARPES and TI’s, let us now move on to historical perspective of TI’s and the work leading up this study.
Table 2.1 is a summary of all topological insulator materials discovered so far, and was compiled by Yoichi Ando[23]. I have highlighted Bi$_2$Se$_3$ to show the relative quality to other topological insulators. We see that Bi$_2$Se$_3$ has largest bulk band gap with simple surface states. It is also one of the easier compounds to grow and ours were grown in Brookhaven National Laboratory in Dr. Gu’s Laboratory. Please note that there are quite a few question marks placed in Table 2.1 because TI’s are relatively new materials and there is still a lot of room for discovery and innovation. Also, notice that all of the TI’s are made of very heavy elements such as bismuth, mercury, lead etc. This is because of the need for strong spin-orbit coupling which is the mechanism for spin polarized surface currents, instead of applying a strong magnetic field. We will discuss this further in the following sections. Please refer to Yoichi Ando’s wonderful paper[23] discussing all the basics and history of TI’s for a more in depth discussion of TI’s in general.
Table 2.1: Summary of TBI found experimentally to date. Found in Yoichi Andos ARXIV paper, will be published in Journal of Japan Applied Physics. SS=Surface states, PT=Phase Transition, SM=Semi-Metal.

<table>
<thead>
<tr>
<th>Type</th>
<th>Material</th>
<th>Band gap</th>
<th>Bulk transport</th>
<th>Remark</th>
</tr>
</thead>
<tbody>
<tr>
<td>2D, (n = 1)</td>
<td>CdTe/HgTe/CdTe</td>
<td>(&lt; 10) meV</td>
<td>insulating</td>
<td>high mobility</td>
</tr>
<tr>
<td>2D, (n = 1)</td>
<td>AlSb/InAs/GaSb/AlSb</td>
<td>(~4) meV</td>
<td>weakly insulating</td>
<td>gap is too small</td>
</tr>
<tr>
<td>3D (1:111)</td>
<td>Bi(_{1-x})Sb(_x)</td>
<td>(&lt; 30) meV</td>
<td>weakly insulating</td>
<td>complex S.S.</td>
</tr>
<tr>
<td>3D (1:111)</td>
<td>Sb</td>
<td>semimetal</td>
<td>metallic</td>
<td>complex S.S.</td>
</tr>
<tr>
<td><strong>3D (1:000)</strong></td>
<td>Bi(_2)Se(_3)</td>
<td>0.3 eV</td>
<td>metallic</td>
<td>simple S.S.</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>Bi(_2)Te(_3)</td>
<td>0.17 eV</td>
<td>metallic</td>
<td>distorted S.S.</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>Sb(_2)Te(_3)</td>
<td>0.3 eV</td>
<td>metallic</td>
<td>heavily (p)-type</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>Bi(_{1-x})Te(_x)</td>
<td>(&lt; 0.2) eV</td>
<td>reasonably insulating</td>
<td>(\rho_{xx}) up to 6 (\Omega) cm</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>(Bi(_2)Sb(_2))Te(_3)</td>
<td>(&lt; 0.2) eV</td>
<td>moderately insulating</td>
<td>mostly thin films</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>Bi(_{2-x})Sb(_x)Te(_2)</td>
<td>(&lt; 0.3) eV</td>
<td>reasonably insulating</td>
<td>Dirac-cone engineering</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>Bi(<em>{2}Te</em>{1.6}S_{1.4})</td>
<td>0.2 eV</td>
<td>metallic</td>
<td>(n)-type</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>Bi(_{1-x})Sb(_x)Te(_x)S</td>
<td>0.2 eV</td>
<td>moderately insulating</td>
<td>(\rho_{xx}) up to 0.1 (\Omega) cm</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>Sb(_2)Te(_3)Se</td>
<td>?</td>
<td>metallic</td>
<td>heavily (p)-type</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>Bi(_2)(Te(_2)Se(_2))(Se(_2))</td>
<td>0.3 eV</td>
<td>semi-metallic</td>
<td>natural Kawazakite</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>TlBiSe(_2)</td>
<td>(&lt; 0.35) eV</td>
<td>metallic</td>
<td>simple S.S., large gap</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>TlBiTe(_2)</td>
<td>(&lt; 0.2) eV</td>
<td>metallic</td>
<td>distorted S.S.</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>TlBi(S(<em>x)Se(</em>{1-x}))</td>
<td>(&lt; 0.35) eV</td>
<td>metallic</td>
<td>topological PT.</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>PbBi(_2)Te(_4)</td>
<td>(&lt; 0.2) eV</td>
<td>metallic</td>
<td>S.S. nearly parabolic</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>PbSb(_2)Te(_4)</td>
<td>?</td>
<td>metallic</td>
<td>(p)-type</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>GeBi(_2)Te(_4)</td>
<td>0.18 eV</td>
<td>metallic</td>
<td>(n)-type</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>PbBi(_2)Te(_7)</td>
<td>0.2 eV</td>
<td>metallic</td>
<td>heavily (n)-type</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>GcBi(_{1-x})Sb(_x)Te(_7)</td>
<td>0.1(-0.2) eV</td>
<td>metallic</td>
<td>(n ,(p) type at (x = 0)</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>(PbSe(_2))(Bi(_2)Se(_3))(_6)</td>
<td>0.5 eV</td>
<td>metallic</td>
<td>natural heterostructure</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>(Bi(_2))(Bi(_2)Se(_3))(_6)</td>
<td>semimetallic</td>
<td>metallic</td>
<td>((Bi(_2))(Bi(_2)Se(_3))(_m) series</td>
</tr>
<tr>
<td>3D (1:000)</td>
<td>(Bi(_2))(Bi(_2)Te(_3))(_2)</td>
<td>?</td>
<td>?</td>
<td>no data published yet</td>
</tr>
<tr>
<td>3D TCI</td>
<td>SnTe</td>
<td>0.3 eV (4.2 K)</td>
<td>metallic</td>
<td>Mirror TCI, (n_M = -2)</td>
</tr>
<tr>
<td>3D TCI</td>
<td>Pb(_{1-x})Sn(_x)Te</td>
<td>(&lt; 0.3) eV</td>
<td>metallic</td>
<td>Mirror TCI, (n_M = -2)</td>
</tr>
<tr>
<td>3D TCI</td>
<td>Pb(<em>{0.77})Sn(</em>{0.23})Se</td>
<td>invert with (T)</td>
<td>metallic</td>
<td>Mirror TCI, (n_M = -2)</td>
</tr>
<tr>
<td>2D, (n = 1)?</td>
<td>Bi bilayer</td>
<td>(&lt; 0.1) eV</td>
<td>?</td>
<td>not stable by itself</td>
</tr>
<tr>
<td>3D (1:000)?</td>
<td>Ag(_2)Te</td>
<td>?</td>
<td>metallic</td>
<td>famous for linear MR</td>
</tr>
<tr>
<td>3D (1:111)?</td>
<td>SmBi(_5)</td>
<td>20 meV</td>
<td>insulating</td>
<td>possible Kondo TI</td>
</tr>
<tr>
<td>3D (0.001)?</td>
<td>Bi(<em>{14})Rh(</em>{10})</td>
<td>0.27 eV</td>
<td>metallic</td>
<td>possible weak 3D TI</td>
</tr>
<tr>
<td>3D (1:000)?</td>
<td>RBiP(_R) ((R = Lu, Dy, Gd))</td>
<td>zero gap</td>
<td>metallic</td>
<td>evidence negative</td>
</tr>
<tr>
<td>Weyl SM?</td>
<td>Nd(<em>2)(Ir(</em>{1-x})Rh(_x))(_2)O(_7)</td>
<td>zero gap</td>
<td>metallic</td>
<td>too preliminary</td>
</tr>
</tbody>
</table>

2.1 A Brief History of Topological Insulators

The mechanism for the interesting surface state behavior in topological insulators have been studied since 1980 when the quantum hall effect was first discovered by von Klitzing[28] and his colleagues. They showed that in large magnetic fields the Hall conductance is quantized by integer values in high-mobility 2D semiconductors. The quantum hall
The effect is shown in Figure 2.1, where we see that the bulk has the same insulating electronic states (as a typical insulator) but the edge (or surface) of the material have very peculiar electronic states. It shows that they move along the surface one way, which is dependent on the magnitude and direction of the applied magnetic field. The band diagram shows the same bulk band gap, but with a single connecting surface state indicating a single direction of current propagation[21]. They were awarded a Nobel Prize in 1985 for this revolutionary work. Two years later Thouless, Kohmoto, Nightingale, and den Nijs showed that the quantizing effect was purely quantum mechanical, but also topological[29].

Figure 2.1: Depiction of the quantum hall effect. Figure is taken from [26].

The next major idea in the TI history came from the prediction of the QSH effect and eventual discovery of it by Kato in 2004[30]. Figure 1.9 (b) shows the band diagram and the simple model of the mechanism QSH effect. QSH materials existed with band gaps for their bulk and arbitrarily low energy gaps for their surface states. Now, the QSH effect occurs in the presence of large external magnetic fields. However, researchers wanted to see if there was a such a class of materials that did not need external fields. Murakami, Nagaosa, and Zhang proposed this in 2004[25]. Shortly thereafter, Kane and Mele had the ingenious idea to construct a model based on graphene and incorporate spin-orbit coupling[31]. Their calculations showed that there was indeed the topological invariant $Z_2$ that governed the bizarre features of graphene and other potential QSH insulators. However, the spin-orbit
coupling in graphene is very weak so experimental observation is very difficult, and did not provide conclusive evidence of QSH insulators.

In 2006, Bernevig, Hughes and Zhang [24] then showed that this type of QSH insulator could be observed in heavy metals and proposed CdTe/HgTe/CdTe quantum wells should exhibit characteristic signs of the QSH insulators. Only a year later, Konig et. al. experimentally verified this prediction in 2007[32]. From there, Moore and Balent were able to predict the existent of QSH insulator in 3D systems[33]. They also coined the term topological insulator.

2.2 Previous Work on Bi$_2$Se$_3$

The two main projects we have followed were Park et. al. 2010 and Pan et. al. 2012. The former used EDC only to determine the bottom of the bulk conduction band, and they both used standard MDC analysis to describe the quasi-particle interactions in Bi$_2$Se$_3$. The following two sections will summarize their studies, as both will be drawn upon in the presentation of our findings.

2.2.1 Quasiparticle Scattering in Bi$_2$Se$_3$

In this study, Park et. al.[34] describes the first evidence of the protected nature of the topological states in Bi$_2$Se$_3$. They note that previous experiments have verified the existence of spin chiral states in Bi$_2$Se$_3$ topological metallic regions, but have not discussed the general protection of topological states. This can be observed directly in sharp peaks of the ARPES data due to the long quasi-particle lifetime ($\tau$) of the protected states, refer to Figure 2.2 for peak shapes and scattering mechanisms. They tested one sample, at
T=15K, when it was freshly cleaved and four days after cleaving. Aged samples were used to observe the strength of the protected surface states to physisorbed atoms and molecules that caused charge transfer to the surface, among other effects. This was seen in the change of the Dirac point. They used several photon energies to determine the best probing energy. They determined it was 8 eV because the bulk state spectral function is suppressed due to the $k_z$-selection rules (more defined topological states for certain energies, since less weight in bulk state). Please refer to back Section 1.1, specifically Equation 1.4 and Figure 1.6 for the discrepancy between $k_z$ inside and out side the sample of the photoelectron, and its dependency on the photon energy.

Figure 2.2: Taken from ref. [34]. (a) We see an illustration of the interactions and scattering mechanisms in a 3D topological insulator. Notice how most interactions produce final states in the bulk. This has a direct effect on the lifetime of these electrons, and hence an effect on the self-energy. (b) Shows the effect we can observe, through sharp peaks, in the self-energy, excluding electron-hole creations. We see the electron-electron interactions (electron-hole creation) have significant consequences for high binding energies, while impurities and electron-phonon interactions turn on at the bottom of the bulk conduction band and have significant effect near the Fermi surface. This indicates large scattering to the bulk states as expected.

In Figure 2.2, they show an illustration of the scattering mechanisms to the bulk conduction band, and it’s resulting effect on the self-energy ($\Sigma_{\text{Im}}$). In Figure 2.3 they present their data retrieved from high resolution ARPES data. They see some electron-hole(for high binding energy), phonon and impurity scattering (for low binding energy),
but not the characteristic sharp phonon peaks expected, as seen in Figure 2.2 (b).

Figure 2.3: Figure is taken from ref. [34].

In terms of both the fresh and aged surfaces the they observed a monotonically increasing $\Sigma_{\text{Im}}$, until a certain point (near the bottom of the bulk conduction band), then monotonically decreasing, which is shown in Figure 2.3. This is unlike ordinary metallic cases, where the $\Sigma_{\text{Im}}$ should only increase monotonically. They believe this is due to the phonon/impurity scattering channels where the bottom of the bulk band starts. The lifetime of the surface state electrons were calculated to be approximately 40 fs, which gives a mean free path of $\ell_m = \tau \times v_g = 0.02\mu\text{m}$. The long mean free path (compared inter adsorbate distances) are another indication that the surface states are topologically protected.

The last result they present is that the Fermi surface volume increased between the fresh and aged samples by 2.3%. This means there should be an increase in the $\Sigma_{\text{Im}}$ near the Fermi edge ($E_f$). However, they did not actually observe this which could mean the surface states are topologically protected, especially from the weak disorder from the potential induced by the adsorbates.
2.2.2 Weak Electron-Phonon Interaction in Bi$_2$Se$_3$ Surface

This study demonstrated there was “exceptionally” weak electron phonon interactions between the surface states in Bi$_2$Se$_3$. This claim goes against that of Pan’s study, which stated the surface states are topologically protected and that electron phonon interaction occurs mainly between the bulk and surface conduction bands.

First we present their ARPES spectra in Figure 2.4 (a) and (b), which depicts the change in spectra due to increasing temperature from $T = 18$K to $T = 255$K. The Dirac point shifted from 270meV to 230meV when the temperature was changed, respectively.
Since there were only a few hours between cleaving and reaching both of these temperatures. This means the change in spectra was caused by intrinsic temperature relation to the quasi-particle dynamics instead of “spurious” effects caused by adsorption/desorption of residual gases[20].

Figure 2.4 (c) shows a contradiction to what Figure 2.3 depicts, by showing increased $\Sigma_{\text{Im}}$ for higher energy values. This could be, however, simply due to electron-electron interactions (electron-hole creation) as seen in Figure 2.2. From their analysis they proved this was in fact electron-phonon coupling. This was shown by determining the electron-phonon constant $\lambda$ from the slope of the $\Sigma_{\text{Im}}(0)$. At higher temperatures ($k_B T < \Omega_0/3$),

$$|\Sigma_{\text{Im}}(\omega, T)| = \pi \int_0^\infty d\nu \alpha^2 F(\nu) \left[ 2n(\nu) + f(\omega + \nu) + f(\omega - \nu) \right] \quad (2.1)$$

is roughly linear which turns in to[20].

$$\Sigma_{\text{Im}}(\omega = 0, T) \approx \lambda \pi k_B T \quad (2.2)$$

In Equation 2.1, $\alpha^2 F(\nu)$ is Eliashberg coupling-function, $f(\omega)$ and $n(\omega)$ are the Fermi and Bose-Einstein functions\(^1\), $\Omega_0$ is energy of the strongest phonon[35] The values of the phonon coupling constants were reported as $\lambda = 0.088 \pm 0.009$ for the first sample and $\lambda = 0.076 \pm 0.007$ for the second sample. They claim these are the lowest values, even lower than the theoretically lower limit value presented by S. Giraud and R. Egger [36]. This however, could be underestimated due to the finite resolution of detector. They also noted

\(^1\)Fermi-Dirac statistics for the electrons, which are Fermions, and Bose-Einstein for the phonons, which are Bosons.
the absence of the typical ARPES “kink”, which is in agreement with Park et. al. [34] and all other studies as well. This again shows the low energy surface states are well protected from scattering/coupling due to phonons.

These studies have offered tremendous insight and guidance to our research on the interactions in Bi$_2$Se$_3$. I commend S. R. Park and Z.-H. Pan and their colleagues for writing such a wonderful paper describing there experiments. We, however, will build upon their work and go further to say we observed a significant amount of quasi-particle interactions between surface states in Bi$_2$Se$_3$. 
The Bi$_2$Se$_3$ samples were grown at Brookhaven National Laboratory by Dr. Genda Gu. Gu reports his method has placed the Dirac point at approximately 150meV. This is extraordinarily well grown sample and we are very grateful for his contribution to this project. Gu reports the method of the main sample we use in this study as the following:

The single crystals of Bi$_2$Se$_3$ were grown by a self-flux solution with 97% high purity Se (99.9999%) and 3% high purity Bi (99.9999%). The 100 grams raw materials with 97 %Se and 3% Bi were sealed in a quartz tube and were heated up to 700°C and kept for 12 hours. The sealed quartz tube was rocked for 2 hours at 700°C before the solution at the one end of the tube was cooled down to 455°C at 0.5°C per hour. The sealed quartz tube was kept at 455°C for two hours before the liquid solution at the one end of the tube with Bi$_2$Se$_3$ single crystals was moved to another end of the tube. The tube was cooled down to room temperature at 10°C per hour. A typical single crystal is 2 mm x 2 mm x 0.05 mm.

The ARPES data was taken on beamline 5-4 at Stanford’s Synchrotron Radiation Lightsource. The Bi$_2$Se$_3$ samples were cleaved \textit{in situ} in a ultra high vacuum chamber of pressure \(~5 \times 10^{-11}\) Torr. We explored many photon energies between 7.5 eV to 25 eV, but have found that 11 eV produced the cleanest data. We produce the photon energies of 7.5eV, 11eV, and 25eV below in Figure 3.1.
Figure 3.1: Shows the effect of the ARPES spectra with various photon energies. Note that the bulk conduction band extends approximately 40meV under the Fermi edge.

The energy resolution is 15meV and the angular resolution is 0.3°. We took data on three samples on three different occasions and found that our second experiment produced the cleanest data. We took data from 300K all the way down to 50K, in steps of 25K. The aim of this project is to combine the Energy Distributed Curves (EDC) and Momentum Distributed Curve (MDC) analysis of the samples into one coherent picture. We will use Lorentzian based Fermi Liquid Theory (FLS) for the EDC’s and purely Lorentzian analysis for the MDC’s. The reason for the discrepancy in fit models for EDC and MDC is because many-body interactions, such as electron-electron and phonon coupling, effect the distribution in energy more than the distribution in momentum.

In Figure 3.2 we see the effect of changing the temperature on the ARPES spectra, by increasing the energy of the Dirac point with decreasing temperature. We also see broadening in the spectra, indicating a stronger electron-electron and phonon coupling (should be little to none increase broadening from impurities). Comparing our spectra to Pan et. al. and Park et. al.’s reported spectra, we see that our sample has the lowest energy Dirac point. This indicates are samples are in fact of better quality than theirs. The ideal sample would point the Fermi edge at the Dirac point, as would be needed to produce
a semiconductor\textsuperscript{1}. The reason there is a gap is due to quasi-particle interactions, which is the main focus of this thesis.

![Graph showing the increase in binding energy of the Dirac point with respect to temperature. Notice how for each $\Delta T = 100$K we see an approximate increase of binding energy of 50meV.]

\textbf{3.1 Initial Quasi-Particle Interaction Observation}

We see that there is a much better fit when we convolve the Lorentzian with a Gaussian which indicates electron-phonon interactions. It manifests itself in the distribution of the electrons in $k$-space. Here is an EDC slice sample of our $T = 149$K data. We did this for all of our temperatures and compiled the Gaussian convolutions needed to fit the data here in Figure 3.3.

We first observed the possible quasi-particle scattering when doing line shape analysis on sub-par data. In Figure 3.3 (b) we show the data (obviously a very broad spectra). We show our near Fermi edge fits in the dotted circles, and direction of the EDC curves in the vertical solid red line. We saw our fits improve when incorporating a Gaussian into our FLS EDC. Figure 3.3 (a) shows the FWHM of the Gaussian convolution against the

\textsuperscript{1}Theoretically there is no gap in the surface states, however, in reality there is. The definition of a semiconductor is the chemical potential (or Fermi energy) being in between the conduction and valence bands. If the Fermi energy was at the Dirac point in TIs, we would have a semi-conducting surface and insulating bulk.
momentum (near the Fermi surface) for temperatures $T = 300\text{K}, 298\text{K}, 294\text{K}, 287\text{K}, 275\text{K}, 190\text{K}$ and $149\text{K}$, with respectively decreasing FWHM values. From here we knew we had to obtain sharper data to perform a more thorough analysis to get any sensibly definitive results.

Figure 3.3: Temperature dependence of Gaussian convolution in Bi$_2$Se$_3$. The Gaussian convolution is measured in terms of it FWHM, so it has units of (eV).

3.2 EDC Line Shape Analysis

EDC give us information on how the energy of electrons is distributed at certain $k$ values. The reason there is a spread in energy values is because of the many body effects and interactions and non-zero temperature, and as mentioned previously alter the distribution of energies more than it does to the spread in momenta. Due to these consequences, we must use the more sophisticated FLS theory for the EDC’s. There are three main (different than those established in section 1.2) parameters that are introduced by using the FLS theory which are: $\omega_0$, $Z$, and $\eta$. The value of $\omega_0$ tells you the position of the ARPES kink, $Z$ is the
quasi-particle residue and tells you how the spectral function \( A(k, \omega) \) gets affected (It’s dominant effect scales the dispersion \( \epsilon(k) \) by the amount \( Z \)), and \( \eta \) is related to the lifetime of the quasi-particle. The results of the EDC fits are shown in Figure 3.5 for \( T = 50K, 150K \) and 300K.

![Figure 3.4: FLS EDC results. We present the fits for \( T = 50K, 150K \) and 300K. The columns represent the EDC fits for the three temperatures. The first row is the fitted dispersion \( \epsilon(k) = v_f(\sqrt{|k - \delta k|^2 + \Delta^2_v - \Delta E}) \) (peak positions are circular dots) over the ARPES spectra. The second row shows the EDC line shape fits for the dispersion \( \epsilon(k) \).](image)

In Figure 3.4 we see that the dispersion fit function for \( T = 50K, 150K \) and 300K is

\[
\epsilon(k) = v_f(\sqrt{|k - \delta k|^2 + \Delta^2_v - \Delta E}) \tag{3.1}
\]
Where $v_f$ is the Fermi velocity, $k$ is the momentum, $\delta k$ is Dirac point momentum shift from $\Gamma$, $\Delta_v \equiv \frac{\Delta}{v_f}$, $\Delta$ is the gap formed where the Dirac cone should be and $\Delta E$ is to fix the Fermi crossing point. Since $\Delta_v$ is small, we see that in the limiting case of $k \to 0$, Equation 3.1 becomes parabolic and for $k$ large Equation 3.1 becomes approximately linear.

We determined this by first fitting the dispersion with free parameters then determine the parameters of the fit function for the $\epsilon(k) = a(\sqrt{|k| - b^2 + c^2} - d)$. As seen in Figure 3.4, the determined dispersion is actually a very good fit. We kept the $T=50K$ FLS parameters ($\eta, Z, \omega_0$, Gaussian convolution, etc.) fixed for the rest of the experiments. The Fermi velocities for the fit function (Equation 3.1) of $T=50K$, $150K$ and $300K$ were $\approx 4.0$, $3.85$ and $3.5$, respectively. These values are almost a perfect match for the EDC and MDC self-energy comparisons. The $T=150K$ EDC determined $v_f$ is slightly higher than the $T=150$ MDC $v_f$, which we believe is due the robustness of the FLS theory. To obtain more detail from the EDC line shape analysis we re-fit the dispersion in three different regions with the same FLS parameters in Figure 3.5.

We applied the values of $Z = 0.8$, $\omega_0 = 0.99$ and $\eta = 0.015$ (along with some Gaussian convolution, baseline, spectral weight function etc.) and constrained $\epsilon(k)$ for three different regions. The first two regions (first and last row of Figure 3.5) are the linear dispersion regions. These are lower energy and have these electrons have the Dirac fermion characteristics. The last region (middle row of Figure 3.5) is the parabolic region, which is not exactly parabolic. This is why we could not get perfect fits with the Equation 3.1 as our fit function. However, we see that there is some discrepancy for the high energy electrons. This is due to the fact that the FLS theory does not fully take into account the electron-phonon coupling. As we will see later in Figure 3.7, both the MDC and FLS
self-energies are in agreement except for a sharp peak in the MDC self-energy (FWHM) at high binding energies.

3.2.1 Shastry’s Extremely Correlated Fermi Liquid Model

Professor Shastry has developed a Fermi Liquid model for high $T_c$ superconductors, and we have implemented it in our analysis of the EDC of Bi$_2$Se$_3$. This model is mathematically too complicated to present in this thesis, but I will offer a qualitative explanation of its core concept. The idea is that electrons in a Fermi liquid interact with each other (as well as phonon’s and impurities), and Professor Shastry has developed a highly sophisticated and
elegant model taking into account the electron-electron interactions. However, due numerical computation limits this model can only incorporate electron-electron interactions, but models including other interactions are currently being developed. The net effect of these interactions is to cause electrons to be affected by all other electrons and create what we call quasi-particles. They are called “quasi-particles” because they are actually a collection of electrons that “move” around the crystal together, just as atoms in water balloon move as a single unit but are only being held together by the balloon (or crystal lattice in the case of quasi-particles). In essence they share the same wavefunction and are very much considered an emergent phenomenon.

3.3 MDC Line Shape Analysis

As can be seen in Figure 3.6, the MDC fits are very good, except for some asymmetry in the intensities, as seen in Figure 3.6. The asymmetric intensities should be expected because positive momentum corresponds to a different crystallographic direction than the negative momentum. The effect is slight and can be noticed in others data, but is neglected due to minimal effect on the actual analysis.

As expected the MDC analysis, shown in Figure 3.6, can be fit to very good accuracy with just simple Lorentzian’s. Although, we don’t take into account the fact that TI’s are strongly correlated materials we believe this is adequate model because of the excellent fits, as compared to the EDC’s without FLS theory applied (even with $\epsilon(k)$ as a free parameter). From our MDC’s we can determine the FWHM (constrained to be equal for both peaks), which has units of momentum. Multiplying the FWHM by $v_g/2$ will give us the true imaginary part of the complex self-energy (please refer back to Section 1.2.4
and Equation 1.12). The dispersion’s ε(k) fits the ARPES spectra quite nicely as well. The MDC analysis we employed is quite standard to TI's to extract pertinent information, such as the self-energy, which is further discussed in the following section.

![Double Lorentzian MDC results](image)

**Figure 3.6:** Double Lorentzian MDC results. We present the fits for T = 50K-300K in increments of 50K.

### 3.4 MDC and EDC Comparison Through Σ_{Im}

All the interaction information is stored in the imaginary part of the complex self-energy. To determine the types of interactions in Bi₂Se₃, we have to compare the self-energies of the EDC and MDC analysis. In Figure 3.7, we present the imaginary part of the complex self-energy which was extracted from the MDC data and are comparing it to
the FLS theoretical self-energy with the parameters from our EDC analysis.

Notice in Figure 3.7 the bulk ends around the 50meV point (exact position of the bottom of the bulk conduction band depends on the temperature). Referring back to Section 2.2.1, we see that impurities and phonon interactions should show a spike in the $\Sigma_{\text{Im}}$ near the Fermi edge, indicating scattering from surface states to bulk and electron-electron interactions dominating at high energies. However, we do not see this exactly, which is very intriguing. Our results do agree with the electron-electron interactions dominating at high energies by matching an overall increase in $\Sigma_{\text{Im}}$ for increasing binding energy. However, there is no spike in $\Sigma_{\text{Im}}$ for low energy electrons (near Fermi edge), rather there is one for high binding energies. This spike is characteristic of electron-phonon scattering as seen in Figure 2.2, but it is not in the same region as the bulk. Since we know the relative change in
energy of the initial and final states of these interactions is relatively small, but has to have a large change in momentum, we can conclude that the interacting phonon’s are relatively small excitations. However, seeing that the spike is quite large, we predict that there is a large number of these small amplitude phonon’s that produce coupling between surface states. These intra-surface interactions are concentrated well below the bulk conduction band and have sharp kinks leading into the spike of self-energy.

There is a kink in which the slope decreases in the region of the bulk conduction band, indicating that there is indeed electron-phonon interactions near the Fermi edge. The strange aspect is that there is not a sharp peak as mentioned in Section 2.2.1 and Figure 2.2 for low energies. The data strongly suggests that there is strong intra-surface electron-phonon coupling (and impurity scattering) for the high energy states, since the bulk has lower energy states.

Please note that the Fermi velocity \( v_f \) is decreasing for increasing temperature, until roughly 200K, then it remains constant. We believe that the decrease can be explained by having the same semi-parabolic \( \epsilon(k) \), except with decreasing \( k_f \) values, which would push the semi-parabolic dispersion upwards. Since the Fermi velocity is the proportional to the gradient of the \( \epsilon(k) \), the shifting upwards of the dispersion would in essence decrease the slope of \( \epsilon(k) \) at the Fermi edge. If this were a perfect model of the sample’s dispersion, then we would expect the decreasing change in \( v_f \) to become larger, rather than leveling off. This simple fact proves that the dispersion is truly non-trivial. We can keep all fit model parameters the same, but we must slightly adjust the \( \epsilon(k) \) constraint function for different temperatures and, potentially, the binding energies. The change in the dispersion constraint would need to be fairly insignificant to remain physically consistent, but also take into
account the FLS theory only handles the electron-electron interactions. As stated previously in Section 3.2, the EDC fits were not perfect because FLS neglects electron-phonon and impurity interactions. If we were able to build a model that takes these interactions into account the computational complexity would be astronomical.

3.4.1 Weak Electron-Phonon Coupling of Topological Surface States

Using Equation 2.2, as an approximation of Equation 2.1, we can determine the coupling between the topological surface states and phonons in Bi$_2$Se$_3$[20]. Pan et. al. reported the lowest electron-phonon coupling constant as $\lambda = 0.076 \pm 0.007$, which is significantly lower than the theoretical value[36]. The value is observed by plotting the imaginary part of the complex self-energy at the Fermi level as a function of $T$, which should be linear, with slope given in Equation 2.2. We plot our values of $\Sigma_{\text{Im}}(\omega = 0, T)$ in Figure 3.8.

![Figure 3.8: A plot of $\Sigma_{\text{Im}}(\omega = 0, T)$. The slope tells us the approximate electron-phonon coupling constant $\lambda$.](image)

From the slope of the data in Figure 3.8, we have determined the electron-phonon coupling to be an astonishing low value of $\lambda = 0.049 \pm 0.007$. This value is nearly half as much
as the previously reported phonon coupling constant value, and within the same margin of error. This agrees with lack of the “kink” in the phonon energy range we mentioned earlier and shown in Figure 2.2 and the minimal temperature broadening of the ARPES spectra[20]. However, to determine the temperature broadening accurately would require significantly higher energy resolution than we have now. The small value of the electron-phonon coupling constant and evidence of strongly protected topological states is extremely important for developing new technologies that require precise control of spin properties in materials, as well as new insight into high $T_c$ superconductors.

3.5 Relation to High Temperature Superconductor Cuprates

Topologically protected states of Bi$_2$Se$_3$ have similar properties of Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ (Bi-2212) in the superconducting state at very high temperatures when Bi-2212 is optimally doped. We can compare these properties through the use of EDC analysis with FLS theory.

In Figure 3.9, we see the EDC taken at the Fermi level for both Bi-2212 and Bi$_2$Se$_3$ at $T=300K$. There are three regions for both materials: above the Fermi level ($BE>0eV$), the peak ($-0.1<BE<0eV$), and the background/bulk ($BE<-0.1eV$). There is excellent agreement in the EDCs for above the Fermi level and going into the peak region. There is a slight deviation due to peak values being off by $\approx 7meV$. For the higher energy side of the peaks we see greater differences. The main difference is caused by the inherit background of the high temperature superconductor as compared to the simple bulk of the topological insulator.
Figure 3.9: A comparison of the EDC’s taken at the Fermi level for T=300K data. The Bi-2212 and Bi$_2$Se$_3$ Fermi momenta values are $k_f = 0.075\,\text{Å}^{-1}$ and $k_f = 0.037\,\text{Å}^{-1}$, respectively.

Since the main difference in the EDCs is the background we can assume this is due to some strong scattering effects in Bi-2212. As we have presented in previous sections that the Bi$_2$Se$_3$ has highly protected topological states which could, in theory, prevent any background from being observed. Where as Bi-2212, is not topologically protected from certain scattering events because the electronic transport mechanism is different in Bi-2212 (and other high temperature superconductors) than that of TIs. However, an interesting fact is that superconductivity can be induced in TIs by proximity effects and is currently being studied to aid in the development of quantum computers. Now the thing here is we have concise descriptions TIs through very accurate theories, but we cannot describe high temperature superconductors. So by looking at the peaks in the EDCs of the ARPES spectrum we can use TIs to understand the peak, and to help determine the cause of the background, which is the main difference between TIs and high temperature superconductors. In Matsuyama et. al.’s paper[37] describing the ARPES spectra of Bi-2212 (among others) with both MDC and EDC treated equally, they were able to perfectly fit
the ARPES spectra using extremely correlated Fermi-liquid theory (ECFL). Since ECFL
theory was able to fit MDC and EDC for multiple materials it indicates that it is possible
to establish a phenomenological model capable of describing large classes of high temper-
ature superconductors. This could ultimately lead to breakthroughs in high temperature
superconductor theory and experiment which could lead to the ever sought after room tem-
perature superconductor and open up numerous industries such as spintronics and quantum
information.
Conclusion

We have conducted ARPES experiments on Bi$_2$Se$_3$ at various photon energies and temperatures. We first noticed hints of inter surface quasi-particle interactions in some dirty data. After obtaining very clean and superior data we performed line shape analysis of both MDC and EDC. By investigating how the electrons momentum and energies are spread out we can determine insightful and valuable information about the strongly correlated electronic system of topological insulators. Specifically, in this study, we were able to observe intra-surface interactions in the topological insulator Bi$_2$Se$_3$ by comparing the self-energies extracted from both MDC and EDC analysis. From the imaginary parts of the complex self-energies at Fermi edge we were able to determine the electron-phonon coupling constant to be $\lambda = 0.049 \pm 0.007$, which is the weakest coupling constant reported to date.

4.1 Future Work

We need more computationally sophisticated machines to be able to model all aspects of these extremely correlated systems. By simply taking into account the electron-
electron interactions we are neglecting important scattering mechanisms in these materials such as phonon’s importance in superconductivity. The FLS theory is revolutionary in-and-of itself and I praise Professor Shastry for his contributions to it. Without his model we could not have obtained the results presented here. However, FLS is just the starting point to much more complex models which handle all other scattering mechanisms. More complicated models could offer further insight into more novel materials and complicated physical systems, and potentially lead to more breakthroughs in the sciences and, of course, revolutionary technological devices and materials that could vastly improve the quality of living around the world.
Bibliography


