Introduction:

Topological insulators are a newly discovered class of narrow-gap semiconductors with strong spin-orbit coupling and a novel type of metallic surface state. Materials like Bi$_2$Se$_3$ and Bi$_2$Te$_3$ have recently been identified as topological insulators [1], but already have a long history as industry-standard thermoelectric materials. The topologically protected metallic surface state arises because the s-orbital conduction band and p-orbital valence bands anticross to form the bulk gap. As a result, spin-orbit coupled Dirac-like (linear-dispersion) 2D surface states bridge the bulk gap, leaving sheets of spin-orbit coupled surface currents to flow around a bulk-gapped intrinsic material. The thermopower of such surface states has not yet been conclusively measured, and at low temperatures, a novel spin-polarized Seebeck effect may arise when the surface states dominate electron transport. The thermoelectric figure of merit for such nanolayers should increase with vanishing layer thickness due to phonon scattering and may have useful thermoelectric cooling properties, particularly below room temperature when these topological surface states will dominate charge transport. The goal of this work, therefore, was to measure the Seebeck coefficient of the surface state of an exfoliated thin film. Although Hall characterization successfully measured a p-type concentration of $8.8 \times 10^{18}$ cm$^3$, Seebeck characterization was inconclusive, due to film-thickness-dependence of the gold leads.

1. Exfoliation of Bi$_2$Te$_3$ thin flakes

Bi$_2$Te$_3$ has a quintuple layer sandwich structure (Te-Bi-Te-Bi-Te) with weak Van der Waals forces between quintuple layers. Thus we were able to exfoliate thin flakes from the bulk material and transfer them onto a Si substrate with 300 nm SiO$_2$ top insulating layer. The exfoliation method is detailed in our published paper[2] supported by this grant. Atomic force microscopy (AFM) tests showed that flake thickness varied from less than 10 nm for small flakes (less than 1 µm $\times$ 1 µm) to over 400 nm for larger flakes. For electrical and thermal transport measurements, flakes with a minimum size around 20 µm $\times$ 20 µm and no overlapping with other flakes are of particular interest. Those flakes have a typical thickness of 100 - 250 nm and relatively uniform flat top surface. Figure 1 shows the AFM results of a typical flake.

![FIGURE 1](image1.png)

**FIGURE 1.** (a) AFM result for a typical Bi$_2$Te$_3$ flake around 20 µm $\times$ 20 µm. (b) The height profile along line 1 in (a).

2. Measurement design

A photolithography pattern with eight large pads and eight small pads was designed, as in Fig. 2(a). The large pads were designed for easy wire bonding and connecting to macroscopic testing equipment. The small pads are designed as contact pads as well as alignment marks in consequent e-beam patterning. 5 nm of Ti and 45 nm of Au were deposited for this optical lithography pattern.

Two different e-beam patterns are needed for electrical and thermal transport measurement. For electrical transport characterization, Van der Pauw setup with 8 contacts as in Fig. 2(b) was used for Van der Pauw characterization of the conductivity as well as Hall characterization of the density and mobility. For thermoelectric characterization, a series of parallel contacts perpendicular to the thermal gradient was used for measuring Seebeck voltage coming from
thermal gradient, as shown in Fig. 2(c). To ensure good contact with the 100 nm to 200 nm thick flakes, 5 nm of Ti and 195 nm of Au were deposited for e-beam patterns.

![Image of photolithography pattern, Van der Pauw contacts, parallel strip contacts, and thermoelectric measurement apparatus](image)

**FIGURE 2.** (a) Design of the photolithography pattern. (b) Example of Van der Pauw contacts made by E-beam lithography. (c) Example of parallel strip contacts made by E-beam lithography. (d) Design of the thermoelectric measurement apparatus, in which A is the Si substrate with thin film sample on front, B is the 8 Ω resistor used as heat source, C consists of two thermistors on the back of the substrate, and D is the copper strip connecting to the cold sink.

Figure 2(d) shows the design of a microscopic thermoelectric measurement apparatus we built. This apparatus consists of two thermistors attached on the back side of the substrate to measure temperature gradient, a resistor to provide heat, and a cold sink to dissipate heat. The device applies heat to one end of the substrate with an 8 Ω resistor. The other end of the substrate is attached to a copper strip that connects to the cold sink. This creates a noticeable thermal gradient across the sample. All thermal contacts were made using thermally conductive but electrically insulating silver epoxy. By measuring the temperature difference ΔT and distance in thermal gradient direction Δx between the two attached thermistors, and Seebeck voltage ΔV and distance d between the parallel contacts on sample, we will be able to deduce the Seebeck coefficient $S = \frac{\Delta V \cdot \Delta x}{\Delta T \cdot d}$. We can also calculation the substrate thermal conductivity $k = \frac{P \cdot \Delta x}{A \cdot \Delta T}$ by measuring the supplied power $P$ and cross-sectional area $A$ of the substrate to check the functionality of the apparatus. This whole apparatus was built on a 16-pin header so that it can also fit into our Oxford Variable Temperature helium flow cryostat which allows sample characterization from 1.5 K to 300 K.

3. **Electrical measurement results**

Van der Pauw and Hall measurements were made at 300 K and 3.5 K on a sample with thickness $d = 140$ nm, using perpendicular magnetic field up to 15 T.

At 300 K, Van der Pauw measurement showed sheet resistance $R_{\|} = 136$ Ω/□ at zero magnetic field, while the two-point resistances for most contact pairs were around 10 kΩ. Hall measurement showed a hole density $p = 1.6 \times 10^{19}$ cm$^{-3}$ and hole mobility $\mu_p = 210$ cm$^2$/Vs. This thin film mobility is lower than reported bulk Bi$_2$Te$_3$ samples by a factor around 2$^{[3]}$. The linear transverse resistance $R_{xy}$ shows that bulk doping conduction is dominating for this 140 nm sample at room temperature.

At 3.8 K, a curvature was observed in transverse resistance $R_{xy}$ versus magnetic field $B$ curve, implying both n-type and p-type carriers exist at low temperature. By fitting the measured total
longitudinal resistance $R_{xx}$ and transverse resistance $R_{xy}$ into the parallel conduction model\cite{4}, we extracted the carrier density to be $1.2 \times 10^{14}$ cm$^{-2}$ for p-type carriers and $6.5 \times 10^{8}$ cm$^{-2}$ for n-type carriers. As a topological insulator, this Bi$_2$Te$_3$ thin film is expected to have an n-type conducting surface state induced by exposure to air on the top and bottom layers. Assuming that all p-type carriers come from the bulk and all n-type carriers come equally from top and bottom surfaces, the bulk hole density is $8.8 \times 10^{18}$ cm$^{-3}$ and the electron density from each surface is $3.2 \times 10^{8}$ cm$^{-2}$. The mobility of holes and electrons are $5.7$ cm$^2$/Vs and $2040$ cm$^2$/Vs respectively.

4. Thermoelectric measurement results

In the thermoelectric characterization of the Bi$_2$Te$_3$ thin film, the Seebeck voltage showed a very linear relation versus thermal gradient, as shown in Fig. 4(a). Considering the fact that radiation and ambient air also contributed in heat conduction, the measured substrate thermal conductivity $180$ W/(m·K) was reasonably close to that of silicon $149$ W/(m·K). Those evident showed that our thermoelectric testing apparatus was working properly. However, the measured Seebeck coefficient from different contact pairs varied significantly.

FIGURE 3. Transverse resistance $R_{xy}$ versus magnetic field $B$ at (a) $300$ K and (b) $3.8$ K, dashed lines are the fitted value using parallel conduction model. Inset: Optical microscope image of the sample used for electrical characterization.

FIGURE 4. (a) Seebeck voltage $\Delta V$ versus temperature gradient $\Delta T/\Delta x$ on a sample with parallel contacts connecting Bi$_2$Te$_3$ thin film. (b) Temperature difference $\Delta T$ between thermistors versus supplied heating power $P$. 
Other than the sample itself, one possible source of Seebeck voltage is the gold contacts. To rule out that portion of Seebeck signal, we fabricated a “NULL” sample with a gold line instead of the Bi$_2$Te$_3$ flake connecting those parallel contacts. Seebeck voltage measured on this “NULL” sample was at the same order as measured on samples with flakes on them.

Three kinds of contact junctions are possible for this Seebeck voltage, as illustrated in Fig. 5. Since Seebeck voltage is always proportional to temperature different between the junctions, we plotted the “integrated Seebeck coefficient” $\Sigma$, defined as Seebeck voltage $\Delta V$ per unit thermal gradient $\Delta T/\Delta x$, versus junction distance in Fig. 6. Among them, $X3$ shows the best linearity. Combining the fact that we deposited gold with different thickness for photolithography pattern and e-beam lithography pattern, the thickness dependence of gold thin film Seebeck coefficient$^{[5]}$ is likely be the source of this Seebeck voltage. So in order to extract an accurate Seebeck coefficient, it is necessary to use only one step of large-scale e-beam lithography and deposition to make both the fine contacts on Bi$_2$Te$_3$ flakes as well as the microscopic contacts pads. This conclusion marks the status of the experiment thus far.

**FIGURE 5.** Possible kinds of junction pairs that cause the Seebeck voltage in “NULL” contacts, the distances (in direction of thermal gradient) are defined as $X1, X2, X3$ respectively.

**FIGURE 6.** Integrated Seebeck coefficient $\Sigma$ versus $X1, X2, X3$ (defined in Fig. 5) respectively.

**Reference:**
Submitted External Proposals:

1) PI: Matthew Grayson  
Title: “Thermoelectric Characterization of Topological Insulator Surface States”  
Agency: DARPA, Young Faculty Award  
Amount: $300,000  
Period: 06/01/2011 – 05/31/2013

2) PI: Matthew Grayson, (co-PI’s – John Ketterson, Mercouri Kanatzidis)  
Title: “NEB: Topological FET’s -- Topological Insulator Surface States as Field-Effect-Transistor Channels”  
Agency: NSF – Nanoscale Electronics for the year 2020 and Beyond  
Amount: $2,000,000  
Period: 08/01/2011 – 07/31/2015

3) PI: Matthew Grayson  
Title: “Mobility and Carrier Lifetime in Type II Broken Gap Superlattices and Heterostructures”  
Agency: NSF  
Amount: $450,000  
Period: 06/01/2011-05/31/2014

Awarded External Proposals:  
(none)

List of Publications from ISEN funding:  

List of Presentations from ISEN funding:  