

Study of the Potential Dirac Material $\text{Pb}_{0.9}\text{Sn}_{0.1}\text{Se}$ by Soft Point-Contact Spectroscopy

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The method of soft point-contact spectroscopy was refined to clearly measure the superconducting gap of Indium, then applied again in an attempt to measure the Landau levels of the potential Dirac material $\text{Pb}_{0.9}\text{Sn}_{0.1}\text{Se}$. Differential conductance measurements were performed on multiple samples of $\text{Pb}_{0.9}\text{Sn}_{0.1}\text{Se}$ at temperatures down to 2 K and magnetic fields up to 9 T using both indium and conductive silver paint as soft point-contacts in order to optimize the measurement parameters. As the method was modified to include a noise-reducing electronic setup, the resulting differential conductance curves gradually showed more distinct features for the indium samples, eventually arriving at a procedure that confirmed indium's superconducting gap at 2 K. For the unexplored samples, the soft-point contacts used for differential conductance measurements revealed intriguing qualities in the sample's differential conductance curves that should be investigated more extensively.

I. Introduction

Scanning tunneling microscopy (STM) has been successfully employed to measure Landau level (LL) spectra, but the technique is limited to relatively low magnetic fields and ambient pressure¹. For this reason, soft point-contact spectroscopy (SPCS) is experimentally employed under high magnetic fields in order to investigate novel materials that often have unique LL spectra². This technique could then be adapted to observe the pressure dependence of these spectra, with hopes of investigating possible pressure-induced Dirac materials such as topological insulators and 3D Weyl semimetals, materials that provide applications to quantum computing and "spintronic" devices^{3,4}. Adapting a new technique for the measurement of gaps in LLs at various pressures and fields promises to aid in the identification of these novel materials.

Landau levels are cyclotron orbits with distinct, quantized energy values³. When subjected to high magnetic fields, electrons move in LLs, and the quantization of LLs causes oscillations of

the electronic properties of the material called quantum oscillations³. The energy of the electron in LL n may be defined as

$$E_n = (n + \frac{1}{2}) \hbar \omega_c \propto (n + \frac{1}{2}) B \quad (1)$$

for normal materials¹, but for materials with a Dirac dispersion, the energy of the electron can often be written as⁵

$$E_n \propto \omega_c (nB)^{\frac{1}{2}}. \quad (2)$$

As the magnetic field increases, the quantum oscillations become more pronounced. Most techniques for observing LLs have been limited to probing only near the Fermi level, but information about LLs at other energies are necessary to definitively determine whether a material shows a Dirac dispersion of electrons³. For this reason, I attempt to observe LL spectra with soft-point contact spectroscopy, which can probe both above and below a material's Fermi level by tuning a bias voltage over the soft-point contact³.

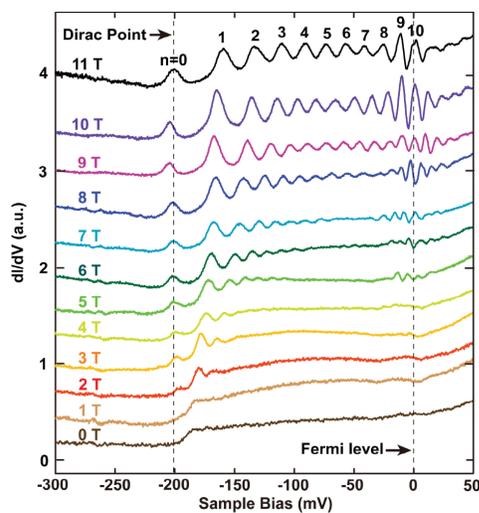


Figure 1: Differential conductance curves obtained through STM at varied magnetic fields, reproduced from Ref. 1.

STM has already been used to determine the spacing between LLs and the superconducting gap of materials with Dirac electrons under moderate magnetic fields¹ (Figure 1), revealing the Dirac nature of the electronic states of Mi_2Se_3 and Cd_3As_2 by observing that the $n=0$ LL was independent of magnetic field³. The topological insulator Bi_2Se_3 was also investigated in two experiments using LLs detected by STM^{1,6}. STM measurements, however, are

limited in the field and pressures under which they can be conducted. Point-contact spectroscopy (PCS) has been employed by fabricating a tunnel junction with an insulating layer to show

spectroscopic information when STM methods and other such technically demanding techniques are not feasible². This method has been successfully used to observe the Fermi surface of URu₂Si₂ beyond the confines of STM⁷.

PCS measured nonlinearities in the differential conductance are a consequence of Andreev reflection, a quantum phenomenon that occurs when a point contact is made between a superconductor and a normal material². When a voltage is applied over the contact junction, the electron in the normal state material “retroreflects” off the superconducting interface as a quasihole, but momentum and charge are conserved and carried across the junction by a Cooper pair⁸. Thus, the current from the normal material is converted to supercurrent as charge passes into the superconducting material⁹. If the applied voltage is much greater than the superconducting gap, all the electrons with energy lower than the gap undergo Andreev reflection, but only the electrons with energy higher than the gap are transmitted through the interface in response to the applied voltage, contributing to the voltage-dependent current².

Like PCS, SPCS is also used to provide information on the spectrum of the superconducting gap through nonlinearities that give rise to atypical structures in the “differential conductance,” or dI/dV . SPCS differs from PCS mainly in the way in which the contact to the sample surface is formed, employing a 20 μm drop of conductive paint or a small indium flake rather than the electrode and metallic tip used in the “needle-anvil” PCS technique^{2,8,10}. When measured through SPCS in particular, the SPCs are stable over a wide temperature range, can be observed at high pressure and field, and do not require the presence of large single-crystal samples².

After first being tested with well-known superconductors, an electronic setup directly measured the differential conductance of my samples. This circuit was applied to SPCS on the known superconductor Indium to confirm the superconducting gap before moving on to

exploring Landau levels of the potential Dirac material $\text{Pb}_{0.9}\text{Sn}_{0.1}\text{Se}$. To the best of my knowledge, the SPCS technique has never been successfully employed to measure gaps between LLs. However, since SPCS is a sensitive technique often used for measuring superconducting gaps on the order of a few meV, it is reasonable to hypothesize that SPCS might also be sensitive enough to investigate LL spectra with similar energy gaps without the limitations of current STM techniques.

II. Method

For a first attempt at obtaining differential conductance curves with the Physical Property Measurement System (PPMS) at 2 K, soft point-contacts were prepared using 1 mil. gold wire was pressed gently to a triangular indium sample to create the four-wire resistance circuit shown in Figure 2. The goal of assembling such fragile connections is to make the contact area between the indium sample and the gold wire smaller than the electron's mean free path. Both gold wires were then soldered to a sample platform and cooled to 2 K, and the IV curve was measured over a DC range of ± 0.005 mV.

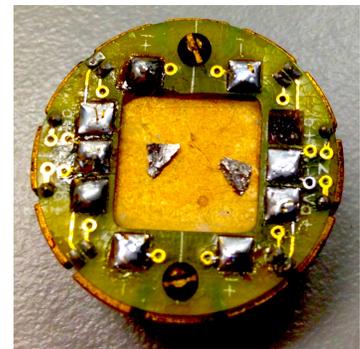


Figure 2: Two 2mm indium samples and gold wire

In the next attempt, the method was modified by pressing the indium between two glass slides, flattening the sample as much as possible. The flattened indium was cut using a razor blade to achieve a very thin indium strip. Annealed 1 mil.

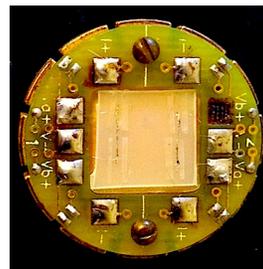


Figure 3: Two 5mm indium samples and annealed platinum wire contacts

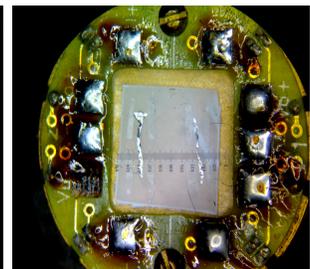


Figure 4: Two 5mm indium samples and annealed platinum wire contacts, with only one SPC per sample

platinum wire rather than gold wire was then pressed gently to both ends

of the indium sample, and the resistance of the sample junction at 300 K was measured at 10-20

Ω with a multimeter, an ideal contact resistance range providing the desired bias voltage without excessive current. This sample was soldered to the PPMS sample platform and cooled to 2 K, where the IV curve was measured over a DC range of ± 5 mV (Figure 3). In a slight variation of this procedure, another sample was prepared with a strip of indium and platinum wires, as previously, but a large square of indium was then pressed over the platinum wire on one end of the sample (Figure 4). This created a sample with only one soft point-contact and one good contact instead of a sample with two soft-point contacts, one at each end. The resistance of this sample was measured to be 1-2 Ω at 300 K, and the dI/dV curve was then taken with the PPMS at 2 K.

A technique called fritting was subsequently used to tune the resistance of the contacts to the appropriate range for Andreev reflection, destroying some of the existing electron pathways or creating new tunnels by piercing the thin oxide layer on the surface of either electrode at the point of contact. To accomplish this tuning, a Keithley 6221 was used to send brief 1 mA pulses through the sample, and the PPMS dI/dV measurement was performed over a bias voltage of ± 3 mV.

With the differential conductance curves tested on the known superconductor indium, I then used the refined SPCS method to take the dI/dV curves of samples with unexplored properties. The first such sample investigated was $Pb_{0.9}Sn_{0.1}Se$. Blocks of the sample were plated with gold on one side using a sputtering machine, and platinum wires dipped in DuPont 4929N conductive paste were attached to

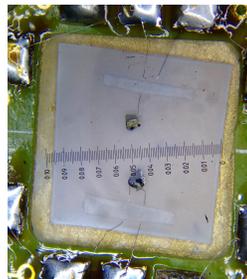


Figure 5: $Pb_{0.9}Sn_{0.1}Se$ and Ag paste contacts were attached to the PPMS puck. The gold-plated side was used for the good contact. The bare side was used for the SPCs, so the annealed Pt wire was attached with a small drop of Ag paste.

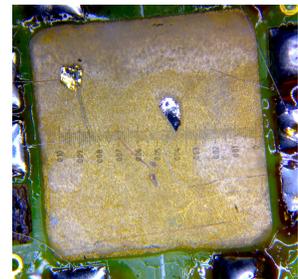


Figure 6: $Bi_{1.01}Te_{0.99}I$ and Ag paste contacts were attached to the PPMS puck after being prepared using the same method of the $Pb_{0.9}Sn_{0.1}Se$ contacts.

both the gold-plated side and the bare side^{2,7,8} (Figure 5). The dI/dV curves were measured from the PPMS data at 2 K and 2 T with a bias voltage ranging from 0-15 mV. The second sample investigated was BiTeI. A block of the sample was plated with gold on one side with the sputtering machine, and the tip of an annealed platinum wire was dipped in conductive paste and attached to the gold-plated side (Figure 6). On the bare side, a platinum wire was attached using a small piece of indium². The wires of the sample were then soldered to the PPMS sample platform, and the dI/dV curve was calculated from the data collected by the PPMS at 2 K and 2 T with a voltage range of ± 400 mV. For this BiTeI sample, the differential conductance curves were repeated with various range settings.

To reduce noise, an electronic setup using four negative feedback op-amps (OP07s) and a summation amplifier (OP551) was borrowed from Prof. Amlan Biswas (Figure 7).

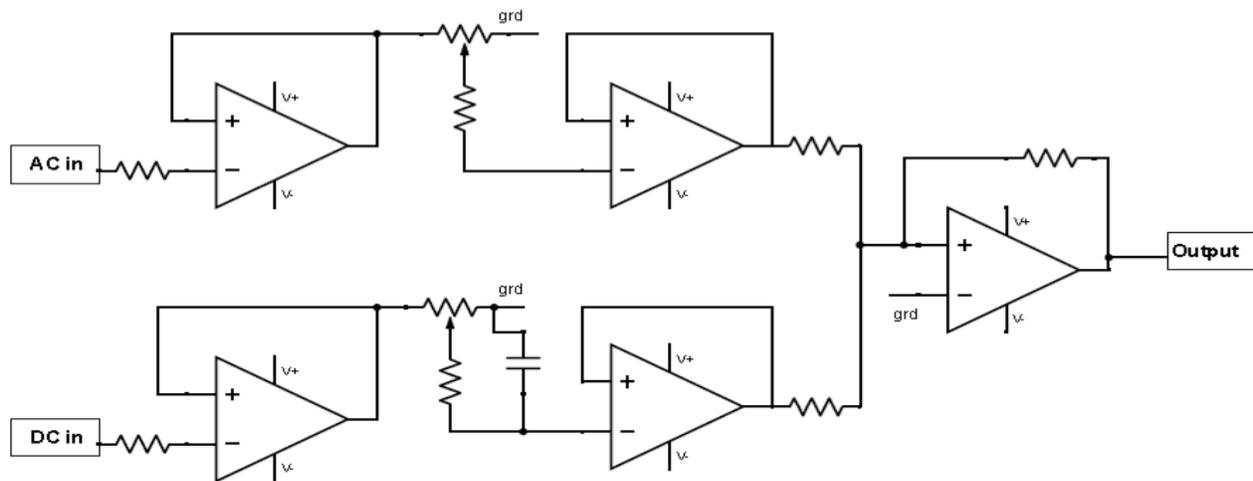


Figure 7: Circuit diagram of original op-amp setup, consisting of four negative feedback circuits and a summation circuit, reproduced from Ref 10. The electrical setup combines a triangle wave with a sine wave before feeding it into the sample.

I employed the op-amp circuit to combine a triangle wave with a small sine wave, and the combined wave was then sent through the sample¹⁰ (Figure 8). The AC voltage drop (dV) over the sample was measured by a SR830 lock-in amplifier, and the AC voltage drop over a known resistance was measured with another to be used in calculating the current (dI) through the

sample. The DC voltage drop across the sample was measured using a Keithley 2182A nanovoltmeter to serve as the bias voltage for the differential conductance graphs. This circuit was used to obtain a graph of the differential conductance that I hoped would match up with the corresponding PPMS dI/dV curve, only with less noise¹⁰, but the summed wave from the op-amp circuit proved to be inexact and riddled with anomalous behavior, necessitating a few modifications.

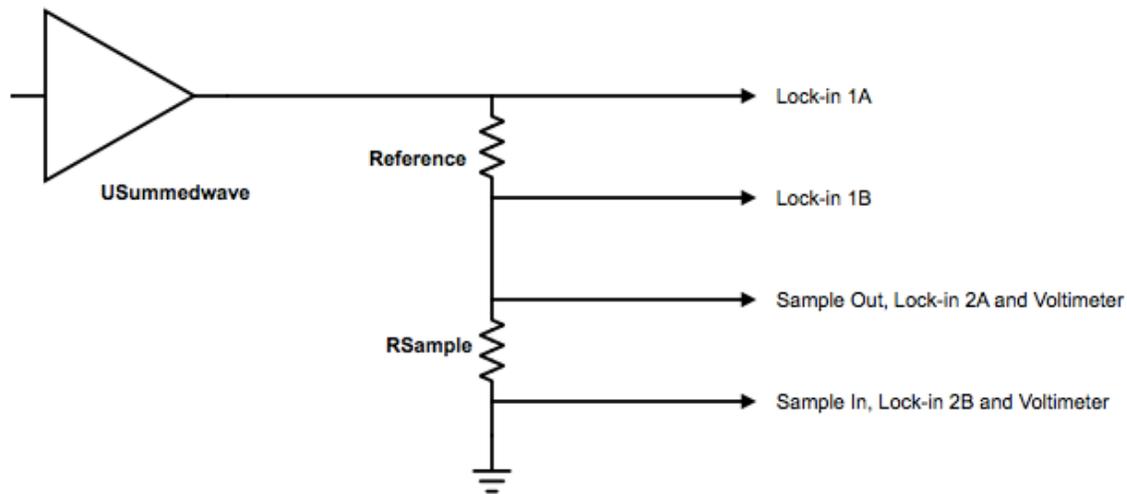


Figure 8: After the triangle wave and sine wave were combined by the op-amp circuit, the summed wave was fed across a known resistor (100 ohms), allowing the current to be calculated. The wave was then fed into the sample, where the drop in AC and DC voltages were measured by the lock-in 2A and the voltmeter respectively.

The first AC and DC negative feedback op-amps being used to buffer the incoming voltages were removed, and the OPA551 summation amplifier was switched with an OPA741. Also, the triangle wave generated was offset by 0.01V to achieve symmetric bias voltages. These adjustments were tested by taking a dI/dV curve over a 1.5 Ω resistor, and the data was shown to match that of the PPMS dI/dV curve of the 1.5 ohm resistor taken with the same conditions. The op-amp circuit, once operating correctly, was applied to measuring the differential conductance of an indium sample connection with a large indium square pressed to one end (Figure 3).

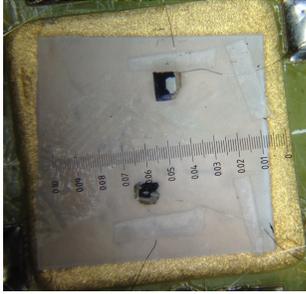


Figure 9: $\text{Pb}_{0.9}\text{Sn}_{0.1}\text{Se}$ and Ag paste contacts were attached to the PPMS puck. One side of the sample was gold-plated, and a large drop of silver paste was used to attach the annealed Pt wire to this side. The bare side was used for the SPC, so the wire was only attached with a 20 micron drop of Ag paste.

Additional soft point-contacts were made using $\text{Pb}_{0.9}\text{Sn}_{0.1}\text{Se}$ as the sample (Figure 9). Again, one side was plated in gold and attached to an annealed platinum wire using a large drop of silver paint. The other side was left bare and attached to an annealed platinum wires using 20 μm drops of silver paint thinned with 2-butoxyethyl acetate to provide an appropriate consistency, creating the soft point-contact. A quick preliminary measurement of the resistance of this sample junction revealed it to be 94Ω . A differential conductance curve shown was taken at 2 K and an applied field of 9 T using the op-amp summation circuit measurements.

Due to the uncertainty of the voltage range in which any unique features of the dI/dV curve might be observed, a new $\text{Pb}_{0.9}\text{Sn}_{0.1}\text{Se}$ sample was prepared with one good contact and eight soft point-contacts (SPCs) using the aforementioned method⁷ (Figure 10).

Measurements of the differential conductance were continued over various magnetic fields,

temperatures, and voltage ranges, and a four-probe resistivity measurement of this sample using

a LS370 illustrated the expected quantum oscillations in the magnetoresistance. Another

$\text{Pb}_{0.9}\text{Sn}_{0.1}\text{Se}$ sample was prepared, this time with one good contact of silver paste and eight SPCs of annealed platinum wire connected to the sample with a 100 micron piece of indium (Figure 11).



Figure 10: $\text{Pb}_{0.9}\text{Sn}_{0.1}\text{Se}$ and Ag paste contact was attached to the PPMS puck, with one good contact on the Au plated side of the sample and eight SPCs on the bare side of the sample.



Figure 11: The $\text{Pb}_{0.9}\text{Sn}_{0.1}\text{Se}$ and In Contacts, with one good contact of Pt wire and Ag paste on the Au-plated side and eight SPCs of 100 μm In pieces and Pt wire on the bare side.

III. Results

My attempt to graph the differential conductance curve of the known superconductor indium at 2 K eventually showed signatures of Andreev reflection, indicating that the contact allows the best chance of observing tunneling into or out of LLs. The initial trial using gold wire on indium samples yielded a differential conductance curve that shows only noise, as did a second sample when prepared and measured using the same method (Figure 12). The test using 5mm strips of indium and annealed platinum wire resulted in a differential conductance showing a distinct spike at 0 mV, but the differential conductance curve lacks any other features (Figure 13). Results from various contacts on the same sample showed similar behavior. The following test, however, using the strip of indium with only one soft-point contact and one good contact, shows a differential conductance with symmetrical spikes that were observed at ± 0.4 mV, tapering off at approximately ± 3 mV (Figure 14). The fritting of this same set of contacts further clarified the features of the differential conductance curve, as claimed by D. Daghero and R. S. Gonnelli² (Figure 15).

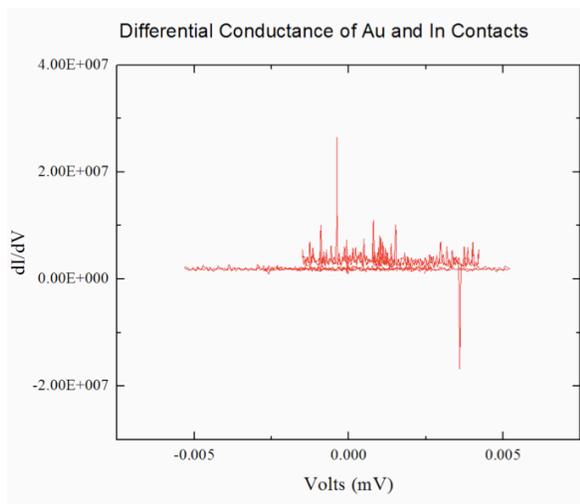


Figure 12: The graph of the differential conductance of Au and In contacts over a voltage range of ± 5 mV.

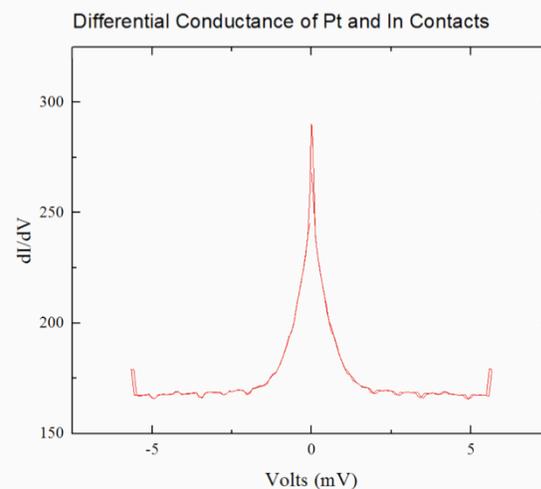


Figure 13: The graph of the differential conductance of Pt and In contacts over a voltage range of ± 5 mV.

The first differential conductance measurement of the $\text{Pb}_{0.9}\text{Sn}_{0.1}\text{Se}$ sample, however, shows only noise, as do the first measurements across the BiTeI sample (Figure 16), necessitating the need for the noise-reducing op-amp circuit.

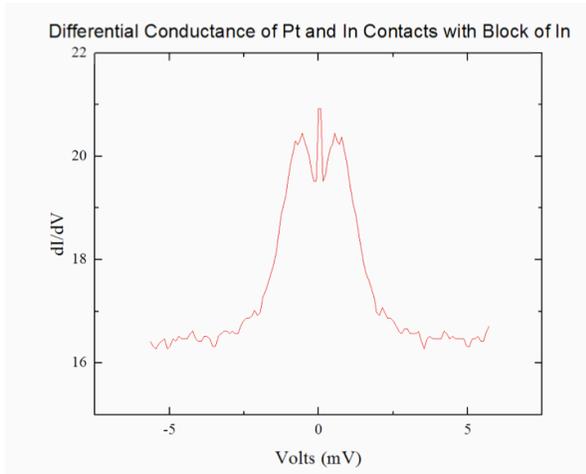


Figure 14: The graph of the differential conductance of Pt and In contacts with only one SPC per sample, as ensured by pressing a large piece of block of In over the contact on one side of each sample.

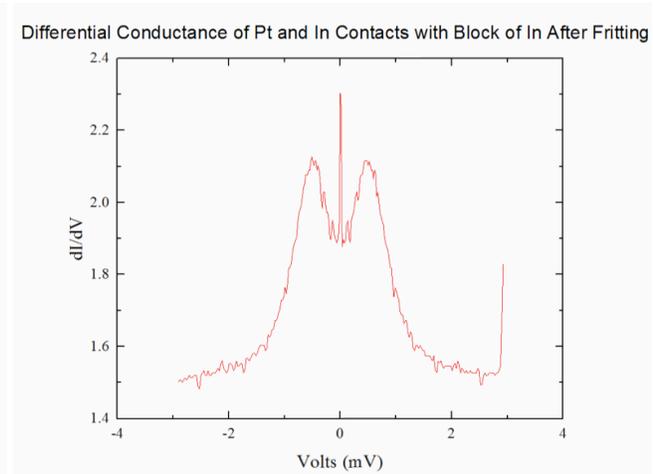


Figure 15: The graph of the differential conductance of Pt and In contacts with only one SPC per sample after fritting the sample with a 1mA pulse from a Keithley 6221 current source.

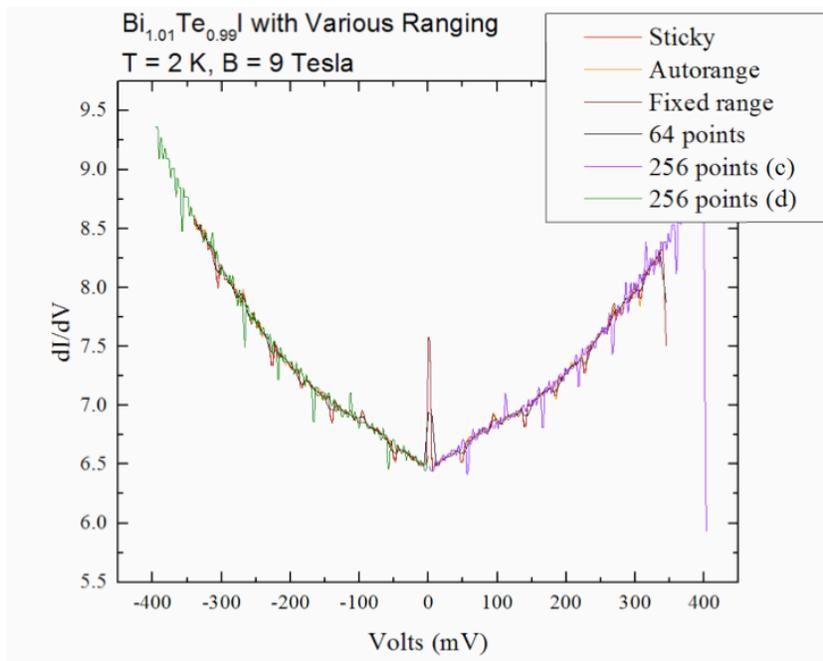


Figure 16: The differential conductance BiTeI and In contacts measured with various settings in the PPMS, each of which provided consistent results.

After the adjustments to the summation circuit (Figure 17), the dI/dV curve from the op-amp circuit matched that of the PPMS for the same indium and platinum wire sample at the same conditions, 2 K and 0 T (Figure 18). The op-amp circuit curve, however, shows far less noise in the indium's

differential conductance graph than the PPMS curve, as hoped. For both the op-amp circuit and the PPMS, the two local maxima of the Indium sample's differential conductance are located at approximately ± 0.4 mV.

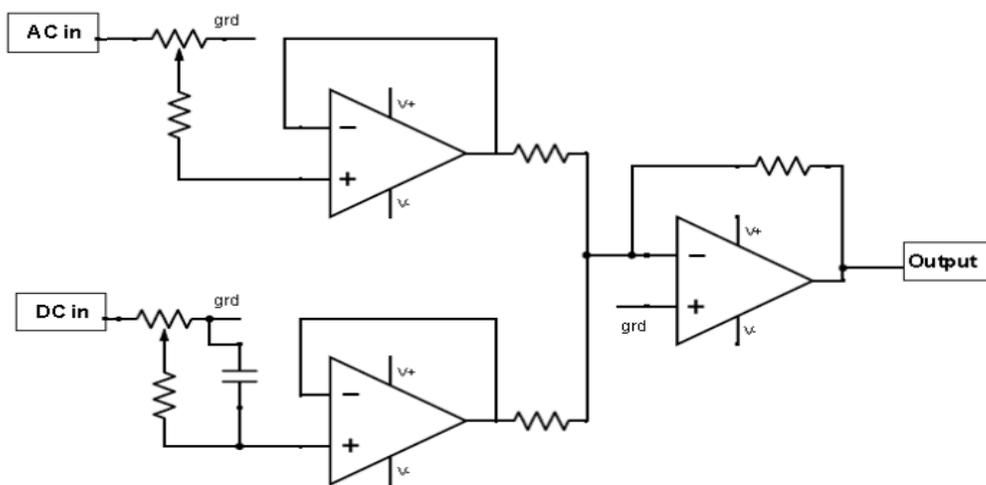


Figure 17: Modified op-amp circuit, with both the AC and DC buffers removed.

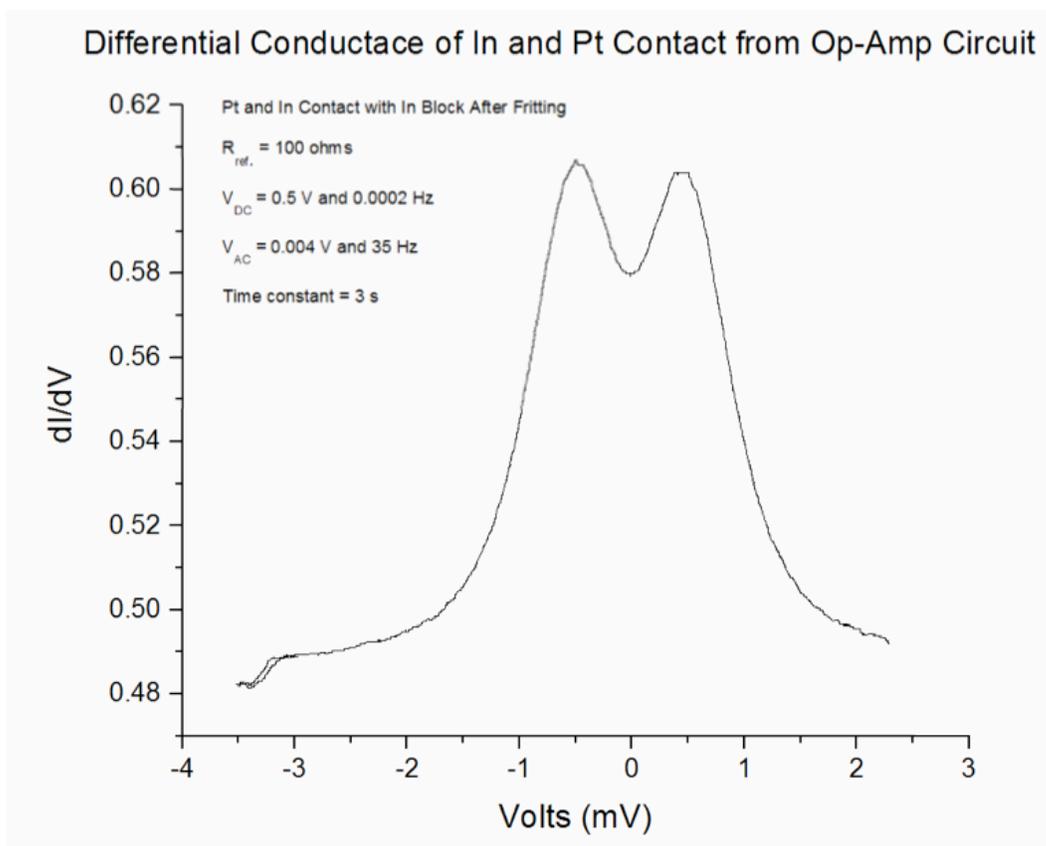


Figure 18: The differential conductance curve found by the op-amp circuit across the Pt and In sample with the In block, after fritting with the 1 mA pulse. This graph can be nicely compared with fig. 11.

Despite the successful construction of the summation circuit, the subsequent differential conductance measurement of the $\text{Pb}_{0.9}\text{Sn}_{0.1}\text{Se}$ sample prepared with a 20 μm drop of silver paint and taken with the op-amp circuit shows only noise over a range of ± 5 mV. The measurement of the $\text{Pb}_{0.9}\text{Sn}_{0.1}\text{Se}$ sample with one good contact and eight soft point-contacts, though, showed a consistent drop in the conductance at approximately 1-8 mV at 9 T and 2 K. This drop remained present under varied magnetic fields up to 9 T (Figure 19), was shown to be temperature dependent (Figure 20), and was also observed when the measurement was repeated with a new $\text{Pb}_{0.9}\text{Sn}_{0.1}\text{Se}$ sample made annealed platinum wire and indium contacts (Figure 21).

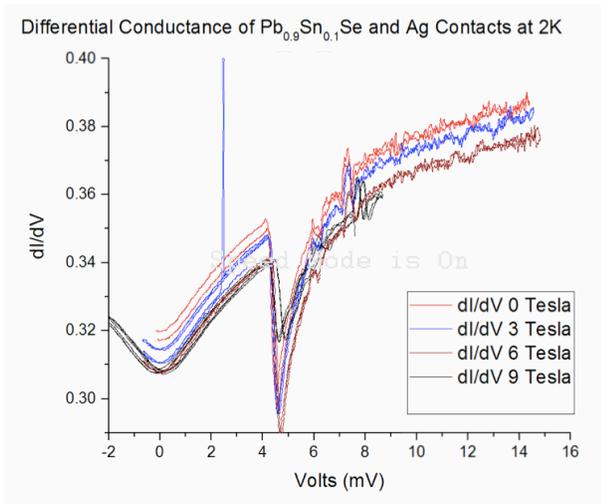


Figure 19: The differential conductance of the $\text{Pb}_{0.9}\text{Sn}_{0.1}\text{Se}$ contacts at the constant temperature of 2 K and varied magnetic fields.

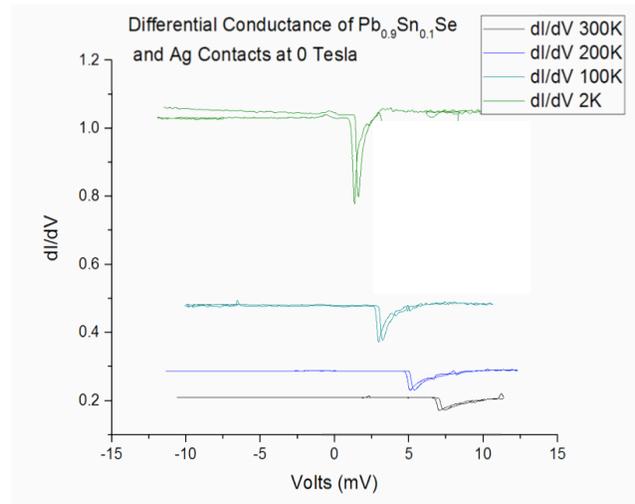


Figure 20: The differential conductance of the $\text{Pb}_{0.9}\text{Sn}_{0.1}\text{Se}$ and Ag contacts is shown at varied temperatures. The reason for the temperature dependence of the differential conductance drop remains unknown.

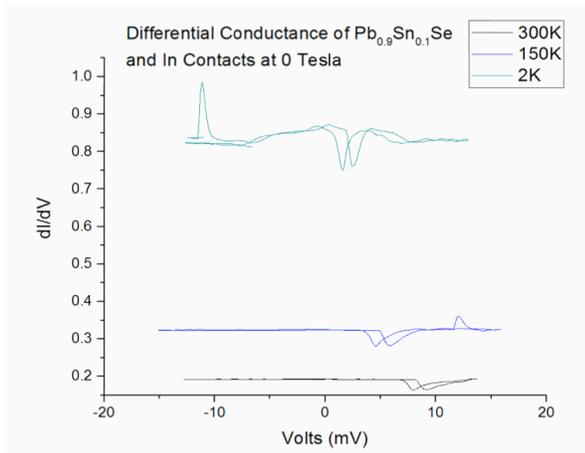


Figure 21: Differential conductance of $\text{Pb}_{0.9}\text{Sn}_{0.1}\text{Se}$ and In Contacts at 0 T and varied temperature.

IV. Discussion/Conclusion

The observed features of the differential conductance curves from the op-amp summation circuit support the use of soft-point contact spectroscopy for the purpose of detecting the superconducting gap, as was demonstrated by the final dI/dV curve of the indium sample with annealed platinum wire. Because the superconducting gap of indium is known to be 1.05 meV at 0 K, and because the equation for the temperature dependent superconducting gap is

$$\Delta(T) = 3.07 k_b T_c (1 - T/T_c)^{1/2}, \quad (3)$$

the dI/dV curve is expected to show features at positive and negative 0.5772 mV. The curve yielded from the op-amp circuit showed a peak at ± 0.4 mV, a value that, when thermal fluctuations of the sample are taken into consideration, is consistent with the predicted gap. This test confirms that the refined SPCS method provides a way to measure the superconducting gap of materials at higher pressures than were previously accessible by STM and PCS.

Additionally, the investigation in $Pb_{0.9}Sn_{0.1}Se$ shows promise in using SPCS for the measurement of the gaps between LLs of potential Dirac materials at various magnetic fields. From the R vs. H graph, clear quantum oscillations in the resistivity are visible, indicating the movement of LLs with changing magnetic field (Figure 22). This

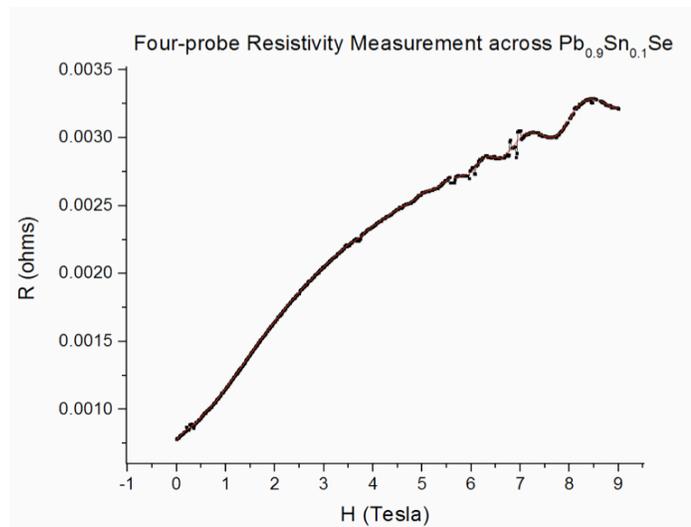


Figure 22: The four-probe resistivity measurement across $Pb_{0.9}Sn_{0.1}Se$ shows quantum oscillations indicating the presence of Landau levels in the material.

measurement verifies the behavior of the LLs, but it fails to reveal their energy spacing. If the drops in the differential conductance stem from tunneling into LLs, the differential conductance

may be compared at various temperatures and fields. Because the drop observed in the differential conductance graph of this multiple-lead sample is independent of magnetic field, existing at both 0 and 9 T, this feature is not indicative of LL tunneling. The drop is possibly an artifact stemming from the electrical setup, which should be tested more extensively in future experiments.

Continued measurements of the dI/dV curves using the noise reducing electronic setup should be performed using $Pb_{0.9}Sn_{0.1}Se$ as well as other likely samples such as $BiTeI$. If meaningful differential conductance curves are found, these measurements will be performed under hydrostatic pressure up to a few GPa in search of signatures of pressure-induced phase transitions from normal materials to Dirac materials, which may be evident with sufficient LL measurements.

Acknowledgements

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