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14. ABSTRACT The ARO-supported research focused on uncovering novel materials and phenomena associated with topological phases. A new area is the Dirac semimetals, Cd ₃ As ₂ and Na ₃ Bi, in which the electrons occupy 3D bulk Dirac states much like graphene, with protected nodes. In a magnetic field, the nodes separate into Weyl nodes. The most important achievement is the observation in Na ₃ Bi of the chiral anomaly which appears as a field-induced enhanced conductivity when the magnetic field is strictly aligned with the electric field (observed as a negative, longitudinal magnetoresistance LMR). The observation, which confirms a prediction from 1992, provides a					
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Report Title

Final Report: Transport Experiments on Topological Insulators

ABSTRACT

The ARO-supported research focused on uncovering novel materials and phenomena associated with topological phases. A new area is the Dirac semimetals, Cd_3As_2 and Na_3Bi , in which the electrons occupy 3D bulk Dirac states much like graphene, with protected nodes. In a magnetic field, the nodes separate into Weyl nodes. The most important achievement is the observation in Na_3Bi of the chiral anomaly which appears as a field-induced enhanced conductivity when the magnetic field is strictly aligned with the electric field (observed as a negative, longitudinal magnetoresistance LMR). The observation, which confirms a prediction from 1983, provides a powerful probe of the chiral current and transport properties of Weyl states in general. A second material (a half-Heusler) has also been found to display the chiral anomaly. Application of a magnetic field leads to the appearance of 2 or 4 Weyl nodes. Again, a large negative LMR is observed. In addition, a large suppression of the thermoelectric coefficient is observed when the chiral anomaly is present. These findings broaden significantly the range of compounds that may exhibit Weyl physics and the chiral anomaly. In a different direction, Murakami has proposed that Weyl nodes always appear when the bulk energy gap in a semiconductor lacking inversion symmetry is forced to close. High pressure has been used to close the gap in PbSnTe . Careful tuning of the pressure allows the quantum oscillations in the MR to be closely monitored as the Fermi sphere expands out of the charge vacuum. An anomalous Hall effect was observed when the lowest Landau level is occupied. This system is an especially attractive platform for exploring Weyl physics in the gap-closing scenario. Finally, the Berry curvature which underpins transport features in Weyl systems, has been shown to produce a Hall effect in neutral spin excitations. In two insulating magnets, a Kagome magnet and a spin liquid pyrochlore, a sizeable thermal Hall effect was observed. In the Kagome magnet, the sign changes are consistent with a theory based on the Berry curvature.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

<u>Received</u>	<u>Paper</u>
08/28/2012 1.00	Jun Xiong, Yongkang Luo, YueHaw Khoo, Shuang Jia, R. Cava, N. Ong. High-field Shubnikov–de Haas oscillations in the topological insulator $\text{Bi}_{1-x}\text{Te}_x$, Physical Review B, (07 2012): 45314. doi: 10.1103/PhysRevB.86.045314
10/01/2013 2.00	Jun Xiong, Yuehaw Khoo, Shuang Jia, R. J. Cava, N. P. Ong. Tuning the quantum oscillations of surface Dirac electrons in the topological insulator $\text{Bi}_{1-x}\text{Te}_x$ by liquid gating, Physical Review B, (07 2013): 1. doi: 10.1103/PhysRevB.88.035128
10/12/2015 13.00	Q. D. Gibson, H. Wu, T. Liang, M. N. Ali, N. P. Ong, Q. Huang, R. J. Cava. Magnetic and electronic properties of CaMn_2Bi_2 : A possible hybridization gap semiconductor, Physical Review B, (02 2015): 85128. doi: 10.1103/PhysRevB.91.085128
10/12/2015 18.00	J. Xiong, S. K. Kushwaha, T. Liang, J. W. Krizan, M. Hirschberger, W. Wang, R. J. Cava, N. P. Ong. Evidence for the chiral anomaly in the Dirac semimetal Na_3Bi , Science, (09 2015): 0. doi: 10.1126/science.aac6089
10/12/2015 17.00	Max Hirschberger, Robin Chisnell, Young S. Lee, N. P. Ong. Thermal Hall Effect of Spin Excitations in a Kagome Magnet, Physical Review Letters, (09 2015): 106603. doi: 10.1103/PhysRevLett.115.106603
10/12/2015 16.00	M. Hirschberger, J. W. Krizan, R. J. Cava, N. P. Ong. Large thermal Hall conductivity of neutral spin excitations in a frustrated quantum magnet, Science, (04 2015): 106. doi: 10.1126/science.1257340
10/12/2015 15.00	Satya K. Kushwaha, Jason W. Krizan, Benjamin E. Feldman, András Gyenis, Mallika T. Randeria, Jun Xiong, Su-Yang Xu, Nasser Alidoust, Ilya Belopolski, Tian Liang, M. Zahid Hasan, N. P. Ong, A. Yazdani, R. J. Cava. Bulk crystal growth and electronic characterization of the 3D Dirac semimetal Na_3Bi , APL Materials, (04 2015): 41504. doi: 10.1063/1.4908158
10/12/2015 14.00	Tian Liang, Quinn Gibson, Mazhar N. Ali, Minhao Liu, R. J. Cava, N. P. Ong. Ultrahigh mobility and giant magnetoresistance in the Dirac semimetal Cd_3As_2 , Nature Materials, (11 2014): 280. doi: 10.1038/nmat4143
10/20/2014 4.00	S. K. Kushwaha, Q. D. Gibson, J. Xiong, I. Pletikoscic, A. P. Weber, A. V. Fedorov, N. P. Ong, T. Valla, R. J. Cava. Comparison of Sn-doped and nonstoichiometric vertical-Bridgman-grown crystals of the topological insulator $\text{Bi}_2\text{Te}_2\text{Se}$, Journal of Applied Physics, (04 2014): 0. doi: 10.1063/1.4871280
10/20/2014 5.00	S K Kushwaha, J W Krizan, J Xiong, T Klimczuk, Q D Gibson, T Liang, N P Ong, R J Cava. Superconducting properties and electronic structure of NaBi , Journal of Physics: Condensed Matter, (05 2014): 0. doi: 10.1088/0953-8984/26/21/212201
10/20/2014 6.00	Mazhar N. Ali, , Jun Xiong, , Steven Flynn, , Jing Tao, , Quinn D. Gibson, , Leslie M. Schoop, , Tian Liang, , Neel Haldolaarachchige,, Max Hirschberger, , N. P. Ong, , R. J. Cava. Large, non-saturating magnetoresistance in WTe_2 , Nature, (10 2014): 205. doi:
10/20/2014 10.00	Tian Liang, Quinn Gibson, Jun Xiong, Max Hirschberger, Sunanda P. Koduvayur, R.J. Cava, N.P. Ong. Evidence for massive bulk Dirac fermions in $\text{Pb}_{1-x}\text{Sn}_x\text{Se}$ from Nernst and thermopower experiments, Nature Communications, (11 2013): 0. doi: 10.1038/ncomms3696

TOTAL: 12

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

<u>Received</u>	<u>Paper</u>
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TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

2011

- 1) American Physical Society March Meeting, symposium, Dallas, Mar 21-25, 2011 “Transport Experiments on Topological Insulators” (invited).
- 2) Workshop and School on Topological Aspects of Condensed Matter Physics, ICTP, Trieste, Jun 27-Jul 8, 2011, “Introduction to Topological Insulators” (invited lecturer).
- 3) 26th International Low-Temperature Physics Conference (LT-26), Beijing, Aug. 10-17th, 2011, “Quantum Oscillations in Topological Insulators – Evidence for the Dirac $\frac{1}{2}$ shift” (invited).
- 4) Satellite workshop on Topological Insulators and Superconductors, Tsinghua University, Beijing, Aug. 18-21, 2011 “Transport experiments on 3D Topological Insulators” (invited).
- 5) Seminar at Kavli Institute for Theoretical Physics China (KITPC), Beijing, Aug. 22nd, 2011 “3D Topological Insulators”.
- 6) Topological Aspects of Quantum-Coherent States in New Materials, Univ. Chicago, Chicago, Oct. 14-15, 2011, “Transport Experiments on 3D Topological Insulators” (invited).
- 7) FIRST-QS2C Workshop on "Emergent Phenomena of Correlated Materials", Okinawa, Dec 12-15, 2011, “Quantum Oscillations in Topological Insulators – Evidence for the Dirac $\frac{1}{2}$ shift” (invited).

2012

- 1) MRS meeting April 9 - 13, 2012, Moscone West Convention Center, San Francisco, “Tuning towards the Dirac Point in Topological Insulators” (symposium speaker).
- 2) Attended DARPA meeting, 2012 MesoDynamic Architectures Review Meeting, San Diego, Aug. 21-23, 2012.
- 3) M2S 2012, Materials and Mechanisms of Superconductivity, Washington DC, July 29 – August 3, 2012, “Fluctuations in cuprate superconductors” (invited speaker)
- 4) TOPNES Annual Board Review Meeting, Univ. St. Andrews, July 9-11, 2012, “Topological Insulators” (invited).
- 5) Gordon Research Conference, Correlated Electron Systems, June 23-24, 2012, Mount Holyoke College, “Liquid Gating to the Dirac Point in Bi₂Te₂Se” (invited)
- 6) Workshop on Spin Liquids, Kavli Inst. Theor. Phys., UCSB, Oct. 28—Nov. 1, 2012, “Phase Diagram of The Transverse Ising Magnet CoNb₂O₆” (invited).
- 7) Gave Physics seminars/colloquia at UCLA (Nov. 2), CSUN (Nov. 1), Penn St (Oct. 25), Johns Hopkins (Nov. 14), U Maryland (Nov. 15).
- 8) International Symposium on Frontier Sciences, Univ. Tokyo, Dec. 8-9, 2012, “Review of Topological Insulators” (invited).

2013

- 1) International Workshop “Topology and Nonequilibrium in Low-Dimensional Electronic Systems”, 16 - 20 September 2013, Max Planck Inst., Dresden, “The physics of Weyl and Dirac Metals” (invited)
- 2) FIRST-QS2C WS on "Emergent Phenomena of Correlated Materials" November 13-16, 2013, “Beyond Z₂ Topological Insulators: Weyl and Dirac Metals” (invited)
- 3) Seminars at Stanford and at UC Berkeley, Oct. 24-25, 2013, “Ultrahigh mobility and giant MR in Dirac Metal Cd₃As₂” (invited by Stanford)

2014

- 1) Theory Winter School, National High Magnetic Field Laboratory, Tallahassee, Jan 6-10, 2014, “Transport Experiments on Topological Insulators” (2 talks). (invited)
- 2) Seminar at Yale Univ., Sep 24, 2014, “Thermal Hall effect of neutral spin excitations in spin liquid”.
- 3) Seminar at Caltech, Nov. 17, 2014, “Ultrahigh mobility in Dirac Semimetals”.

2015

- Endowed lecturer at University of Groningen, Jan 8th, 2015, “Experiments on Topological Insulators”
- 2) Invited symposium speaker, American Physical Society March meeting, San Antonio, Mar 2-6, 2015 “Detection of the chiral anomaly in a Dirac semimetal.”
 - 3) Seminar at Rutgers University, April 21, 2015, “Thermal Hall effect in insulating quantum magnets.”
 - 4) Invited speaker at workshop “Symmetries and Interactions in Topological Matter,” Fine Theoretical Physics Inst., University of Minnesota, May 1-3, 2015, “Topological Dirac semimetals”.
 - 5) Invited speaker at workshop “Strongly correlated topological insulators: SmB₆ and beyond,” University of Michigan, Ann Arbor, Jun 2-4, 2015, “The chiral anomaly in a Dirac semimetal.”
 - 6) Invited lecturer, 11th Annual Princeton Summer School on Condensed Matter Physics,” July 20-24, 2015, in partnership with Prospects in Theoretical Physics 2015 (PiTP). Gave two lectures on Topological matter.
 - 7) Invited talk, First EPiQS Investigator Symposium, Moore Foundation, Sausalito, Aug. 4-7, 2015, “Fun with Berry Curvature.”
 - 8) Invited speaker in workshop “Big Ideas in Quantum Materials,” La Jolla, Dec. 15-16, 2015, “Observation of the chiral anomaly in the Dirac Metal Na₃Bi”

Number of Presentations: 29.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

TOTAL:

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

TOTAL:

(d) Manuscripts

<u>Received</u>	<u>Paper</u>
08/16/2016 12.00	Tian Liang, Quinn Gibson, Mazhar N. Ali , Minhao Liu, R. J. Cava, N. P. Ong. Ultrahigh mobility and giant magnetoresistance in Cd ₃ As ₂ , Nature Materials (05 2014)
10/12/2015 19.00	Jun Xiong , Satya K. Kushwaha , Tian Liang, Jason W. Krizan, Wudi Wang, R. J. Cava, N. P. Ong. Signature of the chiral anomaly in a Dirac semimetal -- a current plume steered by a magnetic field ^L , Cond-mat arXiv (03 2015)
10/20/2014 7.00	Tian Liang , Quinn Gibson , Mazhar N. Ali , Minhao Liu, R. J. Cava, N. P. Ong. Ultrahigh mobility and giant magnetoresistance in Cd ₃ As ₂ : protection from backscattering in a Dirac semimetal, Nature Materials (07 2014)
10/20/2014 8.00	Max Hirschberger, Jason W. Krizan , R. J. Cava, N. P. Ong. Large thermal Hall conductivity of neutral spin excitations in a frustrated quantum magnet, Science (05 2014)
10/20/2014 9.00	Jun Xiong, Satya Kushwaha, Jason Krizan, Tian Liang, R. J. Cava, N. P. Ong. Anomalous conductivity tensor in the Dirac semimetal Na ₃ Bi, Physical Review Letters (08 2014)
TOTAL:	5

Number of Manuscripts:

Books

<u>Received</u>	<u>Book</u>
TOTAL:	

Received

Book Chapter

TOTAL:

Patents Submitted

US Patent:

~~Electronic interconnects and devices with topological surface states and methods for fabricating same~~

Ali Yazdani, N Phuan Ong, Robert J Cava

Application number
14/919,008

Patents Awarded

US Patent:

~~Electronic interconnects and devices with topological surface states and methods for fabricating same~~

Ali Yazdani, N Phuan Ong, Robert J Cava

Application number
14/919,008

Awards

Elected to the Board of Review Editor, Science. Served from Jan. 2012 to Feb. 2014

~~Inducted to U.S. National Academy of Sciences, 2012~~

Selected as Moore investigator (EPiQS award, Gordon and Betty Moore Foundation), 2014

Selected for the list of Highly Cited Researchers (in Physics), compiled by Web of Science, Thomas Reuters, 2014 and 2015

Graduate Students

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	Discipline
Jun Xiong	0.50	
Max Hirschberger	0.30	
Tian Liang	0.30	
FTE Equivalent:	1.10	
Total Number:	3	

Names of Post Doctorates

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	National Academy Member
N. Phuan Ong	0.20	Yes
FTE Equivalent:	0.20	
Total Number:	1	

Names of Under Graduate students supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	Discipline
Ben Osherson	0.20	Physics
Carina A. Belvin	0.10	Physics
FTE Equivalent:	0.30	
Total Number:	2	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 2.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:..... 2.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:..... 2.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):..... 2.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields: 2.00

Names of Personnel receiving masters degrees

<u>NAME</u>
Total Number:

Names of personnel receiving PHDs

<u>NAME</u>
Jun Xiong (PhD 2016)
Tian Liang (PhD 2016)
Total Number:

Names of other research staff

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
FTE Equivalent:	
Total Number:	

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

In the past 3 years, Ong and his students have achieved the following milestones in topological phases of matter under ARO support. Most of the experiments are in collaboration with the group of Bob Cava.

1) Ultrahigh mobility in Cd₃As₂

Dirac semimetals are the 3D analogs of 2D graphene. The bulk states exhibit a Dirac dispersion with nodes that are protected by crystalline symmetry (against gap formation via hybridization with nearby states). The possibility of exploring Weyl physics and the detection of chiral currents in strong magnetic field B has sparked intense interest in Dirac semimetals. Ong has studied in detail transport behavior in the two known Dirac semimetals, Cd₃As₂ and Na₃Bi.

In the report last year, the initial research on both the topological Dirac semimetals Cd₃As₂ and Na₃Bi was described. In the case of Cd₃As₂, the main findings was that some of the crystals (called Type A) which grow as needles display a very steep decrease (by up to a factor of 1,000) in the scattering rate of the carriers by phonons as the temperature T decreases below 50 K to 2 K. This results in an anomalously low bulk residual resistivity (in zero magnetic field B) and a concomitant giant mobility (as high as 10 million cm²/Vs, close to that in ultrapure 2D GaAs heterostructures grown by molecular beam epitaxy). In Cd₃As₂, the application of a modest magnetic field suppresses the high mobility leading to a very large B-linear magnetoresistance (MR). The results were published in Nature Materials [pub. 8]

2) The chiral anomaly in Na₃Bi

One of the holy grails in the field of topological Dirac and Weyl semimetals is the chiral anomaly. [For background, we explain what the chiral anomaly is and its importance in quantum field theory. Shortly after the Dirac equation was proposed, Weyl showed that if the mass m is set to zero, the 4-spinor Dirac wave function separates into two Weyl 2-spinors which describe massless fermions with opposite handedness called chirality $\chi = \pm 1$. The two chiral populations are independent and never transform into each other (chiral symmetry is rigorously respected). These Weyl states were obvious candidates for describing left- and right-handed neutrinos (only the former are seen in our universe). In 1968, Adler, Bell and Jackiw showed that turning on the interaction with electromagnetic fields (photons) immediately ruins the chiral symmetry. The coupling, which engenders an "axial current" that mixes the two populations, is called the chiral or ABJ anomaly. The ABJ anomaly successfully explained the biggest mystery in pion physics at that time (why neutral pions decay 300 million times faster than charged pions). In the ensuing 4 decades, the chiral anomaly has come to assume a central, if still deeply enigmatic, role in the quest to unify the four fundamental forces. For e.g., it acts as an unforgiving litmus test (all gauge theories must be anomaly-free to satisfy renormalizability). The chiral anomaly is also related to the Atiyah-Singer index theorem.]

In 1983, Nielsen and Ninomiya proposed that the chiral anomaly can appear in a crystal that exhibits bulk Dirac states that are strictly massless. In applied parallel electric and magnetic fields (E||B), the axial current flowing between right and left-moving chiral states should be observable as a negative longitudinal magnetoresistance (LMR).

With the discovery of Cd₃As₂ and Na₃Bi (which have protected Dirac states), many experimental groups worldwide undertook the task with urgency. In the Ong-Cava collaboration, the initial interest was on Cd₃As₂. However, the Fermi energy E_F was too high above the node (because of unintended doping by vacancies). Turning to Na₃Bi, Ong and Cava found that the initial crystals also had a high E_F . After 2 years of research, Cava's group succeeded in lowering E_F by a factor of 10 (400 meV to 30 meV). This involved segregating the false phase NaBi from the growth and determining the optimal temperature to hold the melt. In these crystals, Ong's group immediately observed the chiral anomaly as a large enhancement of the longitudinal conductivity when E was applied parallel to B. To further test the finding, Ong's group showed that the conductivity enhancement was confined to a narrow "axial" plume parallel to B. If B is rotated in the xy plane, the plume follows B. This anomalous steering property is incompatible with conventional transport theory, especially when B is modest. By comparing the B² increase in the conductivity with theory, Ong inferred that the axial current is backscattered at a rate 40-60 times slower than the conventional Drude current. It appears that the impurities responsible for backscattering are "blind" to the axial current. This is a significant accomplishment. The results were published in Science 2015 [pub. 12] (DOI: 10.1126/science.aac6089).

2) Chiral anomaly and thermoelectric current in a half Heusler

With the expertise gained, Ong and Cava have identified another semimetal that exhibits the chiral anomaly. The half-Heusler family XB₂M, with X a rare earth and M a transition metal, grows with a zinc blende lattice structure. Its band structure is topologically trivial in zero B. Because the spin-orbit interaction (SOI) is large, the starting atomic orbitals are labelled by total angular momentum quantum numbers |J, M_J). As in the III-V semiconductors (e.g. GaAs), the heavy hole and light-hole states |3/2, +1/2) and |3/2, -1/2) are degenerate at the Gamma point $\Gamma = (0,0,0)$. The material does not have bulk Dirac states with protected nodes. However, when B is applied, the Zeeman energy causes two of these states to cross, leading to protected Weyl nodes. Ong and Cava have investigated the transport properties of GdBiPt and observed strong similarities between its MR and that of Na₃Bi. In longitudinal field, the LMR shows a strong decrease associated with the chiral anomaly. More significantly, because this material is not air-sensitive (unlike Na₃Bi), Ong's student Max Hirschberger has succeeded in observing how the chiral anomaly affects the transport of energy (measured by the thermoelectric conductivity). A notable observation is that the thermoelectric coefficient a, which relates the charge current J to an applied temperature gradient $-\partial_x T$ via the relation $J = a(-\partial_x T)$, is strongly suppressed when the chiral anomaly is present. The appearance of this dramatic

suppression is a challenge to theory. Recently, 2 theoretical papers (Fiete et al., Spivak et al.) addressing thermopower in Weyl metals have appeared.

The present findings hold the promise that the chiral anomaly may be observed in a large family of semimetals with strong SOI (e.g. compounds with zinc blende lattice such as HgCdTe). A further advantage is that the number of Weyl nodes is very small (2 or 4 depending on the direction of B, compared with 24 in Weyl metals e.g. TaAs). This allows a far simpler comparison of transport experiments with theory. The report was published online in Nature Materials (pub. 14).

3) Gap-closing route to Weyl nodes achieved by tuning pressure

In last year's report, pressure experiments on PbSnTe were described. Ong's group used hydrostatic pressure P (up to 27 kbars) to "tune" the bulk gap in PbSnTe doped with In. In samples in which the Sn content $x(\text{Sn}) = 0.25$, they uncovered highly unusual electronic properties. In the metallic phase (10 to 25 kbars), they observe a dramatic lowering of the resistivity in zero field suggestive of a highly conducting state at low temperatures. The Hall conductivity displays a step change across $B = 0$. However, when B is finite, the high-conductance state is destroyed and the resistivity increases rapidly. The gap inversion occurs within the pressure window $10 < P < 25$ kbars. Interestingly, outside this window ($P > 25$ kbars), the system shows the opposite behavior. In zero field, it is a gapped insulator, but when the field exceeds 3-5 T, the sample reverts back to the high-conductance state, displaying a giant, negative MR.

During the past 12 months, the pressure experiments have been extended to the composition $x(\text{Sn}) = 0.5$. A major motivation for these experiments is a recent theory by Murakami et al. who predict that all semiconductors that break inversion symmetry should display protected Weyl nodes when their energy gaps are closed by tuning some parameter (P, doping or alloying). At ambient pressure, the composition $x(\text{Sn}) = 0.5$ is a good insulator with a small energy gap in the bulk. At a critical pressure $P_c \sim 10$ kbar, a metal insulator transition signals the closing of the gap and the appearance of a small Fermi sphere (FS). Because this material lacks inversion symmetry, the Berry curvature Ω is finite. Hence when the FS appears, Ω exerts a strong influence on the transport properties. By taking incremental steps slowly through the critical value P_c , Ong's group tracked the nucleation of the small FS out of the charge vacuum (as it were). The quantum oscillations in the MR, which reflect successive emptying of Landau levels as B increases, allows the diameter of the FS to be measured accurately. A major surprise is that the Hall resistivity ρ_{yx} saturates to a plateau when the quantum limit is attained (last Landau level occupied). The relation of this surprising result to Weyl states created by pressure is being explored. In light of this theoretical insight, the present system provides an especially attractive system for producing Weyl states. However, the abrupt saturation of ρ_{yx} related to Ω , is not predicted by Murakami's theory. The manuscript reporting the pressure results is under preparation.

4) Giant MR and proximity effect in WTe₂

Last year's report described the discovery of a very large, non-saturating magnetoresistance MR in the semimetal WTe₂. At 4 K, the resistivity in an applied transverse magnetic field B increases rapidly as B² to attain values 200,000 times the zero-B value at 65 Tesla, without signs of deviation from the B² law. The report, which has appeared in Nature (Ali et al., Pub. 8), has triggered strong interest in this semimetal with ongoing debate about whether it is a topological material with unusually large MR.

Recently, Ong's group succeeded in inducing superconductivity in WTe₂ by the proximity effect with Nb as the superconductor. Ong's group found that a supercurrent can be induced below 1 K, even when the superconducting electrodes are separated by ~ 700 nm (in conventional metals the proximity length is typically 100 nm or less). Below 300 mK, the supercurrent rises steeply to attain a maximum value of ~ 300 nA at 10 mK. In a weak B, the critical current displays the periodic modulation described by the Fraunhofer pattern. The preliminary results suggest that the very long mean free path of the carriers enables the supercurrent to penetrate deep into the semimetal. The results are very promising for employing superconducting pairing as a probe of possible Weyl states in WTe₂ as well as in related semimetals. The proximity experiments are ongoing.

5) Thermal Hall effect of neutral currents

A key quantity that relates topological states to transport phenomena is the Berry curvature Ω , which may be regarded as a "magnetic field" that lives in k space. For example, when Dirac nodes separate into 2 Weyl nodes with opposite chirality, the Weyl nodes become sources and sinks of the Berry curvature (also called Chern flux). To date, experiments have focused exclusively on the effect of Ω on electrical transport. In two experiments, Ong's group recently showed that Ω leads to prominent effects on neutral currents as well. In insulating magnets, the heat current below 2 K is largely carried by spin waves or magnons (phonons die out rapidly below 2 K). In a certain class of magnets with a Kagome lattice, theory predicts that Ω produces a thermal Hall conductivity K_{xy} in applied B. This is counter to semiclassical theory because the Lorentz force is undefined for neutral particles. Ong's group has confirmed this surprising prediction in the layered Kagome magnet Cu(1,3-bdc) [pub 7]. The magnitude of K_{xy} observed is quite sizeable. As a function of both B and T, K_{xy} is observed to change signs in accord with a calculation based on Ω . The close correlation between K_{xy} and the longitudinal thermal conductivity K_{xx} rules out a competing theory based on the phonon Hall effect. In a second experiment, Ong's group showed that K_{xy} is large in the pyrochlore Tb₂Ti₂O₇ [pub. 6], which does not order magnetically down to 50 mK despite a large exchange coupling between local moments on Tb (the ground state is a quantum spin ice or a spin liquid). The observed K_{xy} again arises from Ω acting on the neutral spin excitations. As Tb₂Ti₂O₇ does not order, the spin excitations are not magnons (theorists call them "spinons"). The 2 experiments confirm that Ω can exert a large Hall-like force on excitations even when they are charge neutral. The results were published in Science and in PRL (Pubs. 10 and 11).

6) Optimizing the bulk resistivity of Bi₂Te₂Se

One of the earliest examples of 3D topological insulators (TIs) studied by ARPES was Bi₂Se₃, which is the leading example of the so-called Z₂ time-reversal invariant (TRI) topological insulators. Unfortunately, the Fermi energy E_F in as-grown crystals lies in the bulk conduction band because exchange defects and Se vacancies are impossible to suppress. While several groups have succeeded in lowering E_F into the bulk gap by chemical doping or gating, this results in greatly reduced surface mobility in the topological states. A much more promising candidate Z₂ TI is Bi₂Te₂Se (BTS) which grows with much fewer vacancies and defects so that E_F lies inside the bulk gap. Residual lattice disorder leads to a high density of states in the impurity band within the gap. In the past 3 years, they expended considerable effort to optimize the crystal quality, and succeeded in increasing the bulk resistivity by a factor of 200 to 20 Ωcm at 4 K. Carriers in the surface states display the largest Shubnikov de Haas (SdH) oscillation amplitudes (among all 3D TIs). The surface mobilities vary from 3,000 to 4,000 cm^2/Vs . In crystals cleaved to thickness of 20 μm , the surface conductance can comprise up to 50% of the total conductance. Monitoring the SdH oscillations in an intense magnetic field (45 Tesla), Ong's group succeeded in lowering E_F to the $n = 1$ Landau level (LL). This allowed us to establish with much higher certainty than before that the quantum oscillations have a phase shift of π (in the limit $1/H \rightarrow 0$), characteristic of Dirac states. In addition, they used ionic liquid gating to tune E_F at the surface. They showed that E_F can be tuned to below the $n=1$ LL at 14 T. The liquid gating allows the two conductances to be separately analyzed and the surface and bulk mobilities to be determined. The latter was found to be 20 cm^2/Vs (roughly 100 times smaller than the former). Details of the crystal growth were also reported.

The results were published in pub. 1, 2 and 4

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Technology Transfer

Interim report on work accomplished to date under ARO Award No. W911NF-11-1-0379

N. Phuan Ong, Dept. of Physics, Princeton University

August 15th, 2016

Abstract

The ARO-supported research focused on uncovering novel materials and phenomena associated with topological phases. A new area is the Dirac semimetals, Cd_3As_2 and Na_3Bi , in which the electrons occupy 3D bulk Dirac states much like graphene, with protected nodes. In a magnetic field, the nodes separate into Weyl nodes. The most important achievement is the observation in Na_3Bi of the chiral anomaly which appears as a field-induced enhanced conductivity when the magnetic field is strictly aligned with the electric field (observed as a negative, longitudinal magnetoresistance LMR). The observation, which confirms a prediction from 1983, provides a powerful probe of the chiral current and transport properties of Weyl states in general. A second material (a half-Heusler) has also been found to display the chiral anomaly. Application of a magnetic field leads to the appearance of 2 or 4 Weyl nodes. Again, a large negative LMR is observed. In addition, a large suppression of the thermoelectric coefficient is observed when the chiral anomaly is present. These findings broaden significantly the range of compounds that may exhibit Weyl physics and the chiral anomaly. In a different direction, Murakami has proposed that Weyl nodes always appear when the bulk energy gap in a semiconductor lacking inversion symmetry is forced to close. High pressure has been used to close the gap in PbSnTe . Careful tuning of the pressure allows the quantum oscillations in the MR to be closely monitored as the Fermi sphere expands out of the charge vacuum. An anomalous Hall effect was observed when the lowest Landau level is occupied. This system is an especially attractive platform for exploring Weyl physics in the gap-closing scenario. Finally, the Berry curvature which underpins transport features in Weyl systems, has been shown to produce a Hall effect in neutral spin excitations. In two insulating magnets, a Kagome magnet and a spin liquid pyrochlore, a sizeable thermal Hall effect was observed. In the Kagome magnet, the sign changes are consistent with a theory based on the Berry curvature.

Interim report on work accomplished to date under ARO Award No. W911NF-11-1-0379

N. Phuan Ong, Dept. of Physics, Princeton University

August 15th, 2016

In the past 3 years, Ong and his students have achieved the following milestones in topological phases of matter under ARO support. Most of the experiments are in collaboration with the group of Bob Cava.

1) Ultrahigh mobility in Cd_3As_2

Dirac semimetals are the 3D analogs of 2D graphene. The bulk states exhibit a Dirac dispersion with nodes that are protected by crystalline symmetry (against gap formation via hybridization with nearby states). The possibility of exploring Weyl physics and the detection of chiral currents in strong magnetic field $\mathbf{B}$ has sparked intense interest in Dirac semimetals. Ong has studied in detail transport behavior in the two known Dirac semimetals, Cd_3As_2 and Na_3Bi .

In the report last year, the initial research on both the topological Dirac semimetals Cd_3As_2 and Na_3Bi was described. In the case of Cd_3As_2 , the main findings was that some of the crystals (called Type A) which grow as needles display a very steep decrease (by up to a factor of 1,000) in the scattering rate of the carriers by phonons as the temperature T decreases below 50 K to 2 K. This results in an anomalously low bulk residual resistivity (in zero magnetic field B) and a concomitant giant mobility (as high as 10 million cm^2/Vs , close to that in ultrapure 2D GaAs heterostructures grown by molecular beam epitaxy). In Cd_3As_2 , the application of a modest magnetic field suppresses the high mobility leading to a very large B -linear magnetoresistance (MR). The results were published in *Nature Materials* [pub. 8]

2) The chiral anomaly in Na_3Bi

One of the holy grails in the field of topological Dirac and Weyl semimetals is the chiral anomaly. [For background, we explain what the chiral anomaly is and its importance in quantum field theory. Shortly after the Dirac equation was proposed, Weyl showed that if the mass m is set to zero, the 4-spinor Dirac wave function separates into two Weyl 2-spinors which describe massless fermions with opposite handedness called chirality $\chi = \pm 1$. The two chiral populations are independent and never transform into each other (chiral symmetry is rigorously respected). These Weyl states were obvious candidates for describing left- and right-handed neutrinos (only the former are seen in our universe). In 1968, Adler, Bell and Jackiw showed that turning on the interaction with electromagnetic fields (photons) immediately ruins the chiral symmetry. The coupling, which engenders an “axial current” that mixes the two populations, is called the chiral or ABJ anomaly. The ABJ anomaly successfully explained the biggest mystery in pion physics at that time (why neutral pions decay 300 million times faster than charged pions). In the ensuing 4 decades, the chiral anomaly has come to assume a central, if still deeply enigmatic, role in the quest to unify the four fundamental forces. For e.g., it acts as an unforgiving litmus test (all gauge theories must be anomaly-free to satisfy renormalizability). The chiral anomaly is also related to the Atiyah-Singer index theorem.]

In 1983, Nielsen and Ninomiya proposed that the chiral anomaly can appear in a crystal that exhibits bulk Dirac states that are strictly massless. In applied parallel electric and magnetic fields ($\mathbf{E} \parallel \mathbf{B}$), the axial current flowing between right and left-moving chiral states should be observable as a negative longitudinal magnetoresistance (LMR).

With the discovery of Cd_3As_2 and Na_3Bi (which have protected Dirac states), many experimental groups worldwide undertook the task with urgency. In the Ong-Cava collaboration, the initial interest was on Cd_3As_2 . However, the Fermi energy E_F was too high above the node (because of unintended doping by vacancies). Turning to Na_3Bi , Ong and Cava found that the initial crystals also had a high E_F . After 2 years of research, Cava’s group succeeded in lowering E_F by a factor of 10 (400 meV to 30 meV). This involved segregating the false phase NaBi from the growth and determining the optimal temperature to hold the melt. In these crystals, Ong’s group immediately observed the chiral anomaly as a large enhancement of the longitudinal conductivity when $\mathbf{E}$ was applied parallel to $\mathbf{B}$. To further test the finding, Ong’s group showed that the conductivity enhancement was confined to a narrow “axial” plume

parallel to $\mathbf{B}$. If $\mathbf{B}$ is rotated in the xy plane, the plume follows $\mathbf{B}$. This anomalous steering property is incompatible with conventional transport theory, especially when B is modest. By comparing the B^2 increase in the conductivity with theory, Ong inferred that the axial current is backscattered at a rate 40-60 times slower than the conventional Drude current. It appears that the impurities responsible for backscattering are “blind” to the axial current. This is a significant accomplishment. The results were published in *Science* 2015 [pub. 12] (DOI: 10.1126/science.aac6089).

2) Chiral anomaly and thermoelectric current in a half Heusler

With the expertise gained, Ong and Cava have identified another semimetal that exhibits the chiral anomaly. The half-Heusler family $X\text{Bi}M$, with X a rare earth and M a transition metal, grows with a zinc blende lattice structure. Its band structure is topologically trivial in zero B . Because the spin-orbit interaction (SOI) is large, the starting atomic orbitals are labelled by total angular momentum quantum numbers $|J, M_J\rangle$. As in the III-V semiconductors (e.g. GaAs), the heavy hole and light-hole states $|3/2, \pm 3/2\rangle$ and $|3/2, \pm 1/2\rangle$ are degenerate at the Gamma point $\Gamma = (0,0,0)$. The material does not have bulk Dirac states with protected nodes. However, when $\mathbf{B}$ is applied, the Zeeman energy causes two of these states to cross, leading to protected Weyl nodes. Ong and Cava have investigated the transport properties of GdBiPt and observed strong similarities between its MR and that of Na_3Bi . In longitudinal field, the LMR shows a strong decrease associated with the chiral anomaly. More significantly, because this material is not air-sensitive (unlike Na_3Bi), Ong’s student Max Hirschberger has succeeded in observing how the chiral anomaly affects the transport of energy (measured by the thermoelectric conductivity). A notable observation is that the thermoelectric coefficient α , which relates the charge current $\mathbf{J}$ to an applied temperature gradient $-\vec{\nabla}T$ via the relation $\mathbf{J} = \alpha(-\vec{\nabla}T)$, is strongly suppressed when the chiral anomaly is present. The appearance of this dramatic suppression is a challenge to theory. Recently, 2 theoretical papers (Fiete *et al.*, Spivak *et al.*) addressing thermopower in Weyl metals have appeared.

The present findings hold the promise that the chiral anomaly may be observed in a large family of semimetals with strong SOI (e.g. compounds with zinc blende lattice such as HgCdTe). A further advantage is that the number of Weyl nodes is very small (2 or 4 depending on the direction of $\mathbf{B}$, compared with 24 in Weyl metals e.g. TaAs). This allows a far simpler comparison of transport experiments with theory. The report was published online in *Nature Materials* (pub. 14).

3) Gap-closing route to Weyl nodes achieved by tuning pressure

In last year’s report, pressure experiments on PbSnTe were described. Ong’s group used hydrostatic pressure P (up to 27 kbars) to “tune” the bulk gap in PbSnTe doped with In. In samples in which the Sn content $x(\text{Sn}) = 0.25$, they uncovered highly unusual electronic properties. In the metallic phase (10 to 25 kbars), they observe a dramatic lowering of the resistivity in zero field suggestive of a highly conducting state at low temperatures. The Hall conductivity displays a step change across $B = 0$. However, when B is finite, the high-conductance state is destroyed and the resistivity increases rapidly. The gap inversion occurs within the pressure window $10 < P < 25$ kbars. Interestingly, outside this window ($P > 25$ kbars), the system shows the opposite behavior. In zero field, it is a gapped insulator, but when the field exceeds 3-5 T, the sample reverts back to the high-conductance state, displaying a giant, negative MR.

During the past 12 months, the pressure experiments have been extended to the composition $x(\text{Sn}) = 0.5$. A major motivation for these experiments is a recent theory by Murakami *et al.* who predict that all semiconductors that break inversion symmetry should display protected Weyl nodes when their energy gaps are closed by tuning some parameter (P , doping or alloying). At ambient pressure, the composition $x(\text{Sn}) = 0.5$ is a good insulator with a small energy gap in the bulk. At a critical pressure $P_c \sim 10$ kbar, a metal insulator transition signals the closing of the gap and the appearance of a small Fermi sphere (FS). Because this material lacks inversion symmetry, the Berry curvature $\mathbf{\Omega}$ is finite. Hence when the FS appears, $\mathbf{\Omega}$ exerts a strong influence on the transport properties. By taking incremental steps slowly through the critical value P_c , Ong’s group tracked the nucleation of the small FS out of the charge vacuum

(as it were). The quantum oscillations in the MR, which reflect successive emptying of Landau levels as B increases, allows the diameter of the FS to be measured accurately. A major surprise is that the Hall resistivity ρ_{yx} saturates to a plateau when the quantum limit is attained (last Landau level occupied). The relation of this surprising result to Weyl states created by pressure is being explored. In light of this theoretical insight, the present system provides an especially attractive system for producing Weyl states. However, the abrupt saturation of ρ_{yx} related to Ω , is not predicted by Murakami's theory. The manuscript reporting the pressure results is under preparation.

4) Giant MR and proximity effect in WTe₂

Last year's report described the discovery of a very large, non-saturating magnetoresistance MR in the semimetal WTe₂. At 4 K, the resistivity in an applied transverse magnetic field B increases rapidly as B^2 to attain values 200,000 times the zero- B value at 65 Tesla, without signs of deviation from the B^2 law. The report, which has appeared in *Nature* (Ali *et al.*, Pub. 8), has triggered strong interest in this semimetal with ongoing debate about whether it is a topological material with unusually large MR.

Recently, Ong's group succeeded in inducing superconductivity in WTe₂ by the proximity effect with Nb as the superconductor. Ong's group found that a supercurrent can be induced below 1 K, even when the superconducting electrodes are separated by ~ 700 nm (in conventional metals the proximity length is typically 100 nm or less). Below 300 mK, the supercurrent rises steeply to attain a maximum value of ~ 300 nA at 10 mK. In a weak B , the critical current displays the periodic modulation described by the Fraunhofer pattern. The preliminary results suggest that the very long mean free path of the carriers enables the supercurrent to penetrate deep into the semimetal. The results are very promising for employing superconducting pairing as a probe of possible Weyl states in WTe₂ as well as in related semimetals. The proximity experiments are ongoing.

5) Thermal Hall effect of neutral currents

A key quantity that relates topological states to transport phenomena is the Berry curvature Ω , which may be regarded as a "magnetic field" that lives in $\mathbf{k}$ space. For example, when Dirac nodes separate into 2 Weyl nodes with opposite chirality, the Weyl nodes become sources and sinks of the Berry curvature (also called Chern flux). To date, experiments have focused exclusively on the effect of Ω on electrical transport. In two experiments, Ong's group recently showed that Ω leads to prominent effects on neutral currents as well. In insulating magnets, the heat current below 2 K is largely carried by spin waves or magnons (phonons die out rapidly below 2 K). In a certain class of magnets with a Kagome lattice, theory predicts that Ω produces a thermal Hall conductivity κ_{xy} in applied $\mathbf{B}$. This is counter to semiclassical theory because the Lorentz force is undefined for neutral particles. Ong's group has confirmed this surprising prediction in the layered Kagome magnet Cu(1,3-*bdc*) [pub 7]. The magnitude of κ_{xy} observed is quite sizeable. As a function of both B and T , κ_{xy} is observed to change signs in accord with a calculation based on Ω . The close correlation between κ_{xy} and the longitudinal thermal conductivity κ_{xx} rules out a competing theory based on the phonon Hall effect. In a second experiment, Ong's group showed that κ_{xy} is large in the pyrochlore Tb₂Ti₂O₇ [pub. 6], which does not order magnetically down to 50 mK despite a large exchange coupling between local moments on Tb (the ground state is a quantum spin ice or a spin liquid). The observed κ_{xy} again arises from Ω acting on the neutral spin excitations. As Tb₂Ti₂O₇ does not order, the spin excitations are not magnons (theorists call them "spinons"). The 2 experiments confirm that Ω can exert a large Hall-like force on excitations even when they are charge neutral. The results were published in *Science* and in PRL (Pubs. 10 and 11).

6) Optimizing the bulk resistivity of $\text{Bi}_2\text{Te}_2\text{Se}$

One of the earliest examples of 3D topological insulators (TIs) studied by ARPES was Bi_2Se_3 , which is the leading example of the so-called Z_2 time-reversal invariant (TRI) topological insulators. Unfortunately, the Fermi energy E_F in as-grown crystals lies in the bulk conduction band because exchange defects and Se vacancies are impossible to suppress. While several groups have succeeded in lowering E_F into the bulk gap by chemical doping or gating, this results in greatly reduced surface mobility in the topological states. A much more promising candidate Z_2 TI is $\text{Bi}_2\text{Te}_2\text{Se}$ (BTS) which grows with much fewer vacancies and defects so that E_F lies inside the bulk gap. Residual lattice disorder leads to a high density of states in the impurity band within the gap. In the past 3 years, they expended considerable effort to optimize the crystal quality, and succeeded in increasing the bulk resistivity by a factor of 200 to $20\ \Omega\text{cm}$ at 4 K. Carriers in the surface states display the largest Shubnikov de Haas (SdH) oscillation amplitudes (among all 3D TIs). The surface mobilities vary from 3,000 to 4,000 cm^2/Vs . In crystals cleaved to thickness of 20 μm , the surface conductance can comprise up to 50% of the total conductance. Monitoring the SdH oscillations in an intense magnetic field (45 Tesla), Ong's group succeeded in lowering E_F to the $n = 1$ Landau level (LL). This allowed us to establish with much higher certainty than before that the quantum oscillations have a phase shift of π (in the limit $1/H \rightarrow 0$), characteristic of Dirac states. In addition, they used ionic liquid gating to tune E_F at the surface. They showed that E_F can be tuned to below the $n=1$ LL at 14 T. The liquid gating allows the two conductances to be separately analyzed and the surface and bulk mobilities to be determined. The latter was found to be 20 cm^2/Vs (roughly 100 times smaller than the former). Details of the crystal growth were also reported. The results were published in pub. 1, 2 and 4.

Summary of the most important results to date

1) The most important result achieved is the observation of the chiral anomaly in the topological Dirac semimetal. In addition to confirming a famous 1983 prediction, the experiment uncovers a novel feature of electronic transport properties intrinsic to Dirac and Weyl states, with the potential of future applications in electronics (exploiting the chiral magnetic effect). This achievement is the culmination of a systematic research program that started with the Z_2 topological insulators. The long series of experiments on optimizing the surface Dirac states in Bi_2Se_3 and $\text{Bi}_2\text{Te}_2\text{Se}$ (involving close collaboration between Ong's and Cava's groups) were essential for improving the crystalline quality. More importantly, they led the Ong-Cava team to think more broadly about 3D Dirac states with protected nodes. This expanded investigation, also influenced by several theory groups (led by Kane, Bernevig, Murakami, Vishwanath, Dai), led to an early start on researching Cd_3As_2 and Na_3Bi . By the same process of crystal optimization, Ong and Cava eventually solved the problem of bringing the Fermi energy in Na_3Bi to values close to the node. This last step was critical for the chiral anomaly publication in *Science*.

2) A related, if distinct, result is the discovery that the half-Heusler compounds also display the chiral anomaly. The experiment shows that even when the zero- B electronic bands are not topologically interesting, they can be converted to topological metals with protected Weyl nodes in a magnetic field. The advantage is that the Heuslers are not air-sensitive, so the range of possible experiments is greatly expanded. In addition, the finding expands the scope of potential Weyl metals to all the zinc blende semiconductors that have a strong spin-orbit interaction. Working with Bernevig, Ong and Cava are expanding the search to the larger class of full Heuslers.

3) Theories based on the Berry curvature predict that it should produce a large transverse force on neutral excitations such as magnons (or more exotic spin excitations) leading to a thermal Hall effect. Ong's group has published experiments on two insulating magnets that have confirmed these predictions. The

first is a layered Kagome magnet in which magnons dominate the heat transport below 2 K. The second is the pyrochlore $\text{Tb}_2\text{Ti}_2\text{O}_7$ which has a ground state described as quantum spin ice or spin liquid. Both systems are devoid of free electrons, but exhibit a sizeable thermal Hall conductivity in agreement with the Berry curvature prediction that neutral excitations can produce a sizeable Hall current in a magnetic field.

4) Finally, two unexpected discoveries were made in the group of semimetals investigated. One is the very large, non-saturating MR found in WTe_2 , which may reflect an unusual property of topological states. The second is the very unusual changes to both the MR and Hall effect in the topological crystalline material PbSnTe under modest hydrostatic pressure. The findings suggest that PbSnTe provides a very clean system for exploring the gap-closing scenario in a system lacking inversion symmetry (Murakami and others recently identified gap-closing in inversion lacking semiconductors as a very promising source of Weyl metals). The transport anomalies suggest that the Berry curvature has a very pronounced effect when the system “nucleates” a small Fermi surface out of the charge vacuum.

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1. Jun Xiong, Yongkang Luo, YueHaw Khoo, Shuang Jia, R. J. Cava, and N. P. Ong, “High-field Shubnikov–de Haas oscillations in the topological insulator $\text{Bi}_2\text{Te}_2\text{Se}$,” *Phys. Rev. B* 86, 045314 (2012). doi: 10.1103/PhysRevB.86.045314

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