Sum-Rule Constraints on the Surface State Conductance of Topological Insulators

K. W. Post, B. C. Chapler, M. K. Liu, J. S. Wu, H. T. Stinson, M. D. Goldflam, A. R. Richardella, J. S. Lee, A. A. Reijnders, K. S. Burch, M. M. Fogler, N. Samarth, and D. N. Basov

Phys. Rev. Lett. **115**, 116804 — Published 11 September 2015

DOI: 10.1103/PhysRevLett.115.116804

Sum rule constraints on the surface state conductance of topological insulators

K. W. Post,[1,*] B. C. Chapler,[1] M. K. Liu,[1] J.S. Wu,[1] H. T. Stinson,[1] M. D. Goldflam,[1] A. R. Richardella,[2] J. S. Lee,[2], A. A. Reijnders,[3] K. S. Burch,[4] M.M. Fogler,[1] N. Samarth,[2] and D. N. Basov[1]

[1]*Physics Department, University of California-San Diego, La Jolla, California 92093, USA*

[2]*Department of Physics, The Pennsylvania State University, University Park, Pennsylvania 16802, USA*

[3]*Department of Physics & Institute for Optical Sciences, University of Toronto, Toronto, Ontario M5S 1A7, Canada*

[4]*Department of Physics, Boston College, Chestnut Hill, Massachusetts 02467, USA*

(Dated: August 31, 2015)

We report the Drude oscillator strength (D) and the magnitude of the bulk band gap (E_g) of the epitaxially grown, topological insulator $(Bi,Sb)_2Te_3$. The magnitude of E_g, in conjunction with the model independent f-sum rule, allowed us to establish an upper bound for the magnitude of D expected in a typical Dirac like system composed of linear bands. The experimentally observed D is found to be at or below this theoretical upper bound, demonstrating the effectiveness of alloying in eliminating bulk charge carriers. Moreover, direct comparison of the measured D to magneto-resistance measurements of the same sample support assignment of the observed low-energy conduction to topological surface states.

PACS numbers:

The prediction and discovery of Dirac-like gapless surface states (SSs) (see Fig.1) at the interface between a topological insulator (TI) and trivial insulator have vaulted TIs to the vanguard of condensed matter physics [1]. Surface probes such as angle resolved photoemission spectroscopy (ARPES) and scanning tunneling spectroscopy (STS) have been extremely successful in verifying and discovering novel phenomena associated with the SSs (*e.g.* Refs. [2–5]). However, progress in the field has been plagued by native defects, resulting in significant concentrations of bulk charge carriers. These bulk dopants inhibit isolation and utilization of SS phenomena [8]. In order to realize many of the novel scientific and technological advances that could blossom from the unique electronic, spin, and optical properties of SS in TIs, it is paramount to eliminate the bulk dopants.

Two archetypal strong TIs (topological invariant ν_0=1 [1]) that are known to suffer from materials issues related to bulk dopants are Bi_2Te_3 and Sb_2Te_3. However, epitaxial films of Bi_2Te_3 can be n-type bulk conductors [7, 8], whereas Sb_2Te_3 films are p-type bulk conductors [9]. Based on this observation, TI materials have been grown where the ratio of Bi:Sb in $(Bi,Sb)_2Te_3$ (BST) is tuned to produce a compensated material with bulk insulating properties[10–12]. Here we prove, using optical spectroscopy, the acute effectiveness of alloying in reducing or eliminating bulk charge carriers. The advantage of our optical experiments is that they give direct access to the frequency dependent electrodynamic response of free carriers in a metal via the Drude peak. In our $(Bi,Sb)_2Te_3$ film we find the Drude oscillator strength D sufficiently low as to lie at or below the upper bound that theoretically anticipated for an isolated Dirac SS Drude response (*i.e.* a Drude response with no bulk contribution) (see Table I). These infrared data are complemented by charge carrier density n and mobility μ from Hall effect measurements, as well as magnetoresistance, which are all consistent with low-energy conduction arising from the topological SS.

We begin by describing the optical response of Dirac electrons in a strong TI, neglecting potential interband bulk→SS transitions (or *vice versa*) [19]. The simplest Dirac electron system is composed of linear bands (LB), without spin or valley degeneracy, where the dispersion is given by $E(k) = kv_F$, as illustrated schematically in Fig. 1a, and 1b. In this system, the total conductance ($G^{\rm LB}_{\rm tot}$) is the sum of an intraband ($G^{\rm LB}_{\rm intra}(\omega)$) and an interband ($G^{\rm LB}_{\rm inter}(\omega)$) component. When the Fermi energy (E_F) is at the Dirac point, theory predicts SS interband transitions give rise to a frequency independent conductance of $G^{\rm LB}_{\rm inter} = \frac{1}{8}\frac{\pi e^2}{h}$ [20]. However, the area of the Fermi surface is zero yielding $G^{\rm LB}_{\rm intra} = 0$. When E_F is shifted away from the Dirac point, empty states suppress interband transitions at energies below $2|E_F|$, as illustrated in Fig. 1b, resulting in $G^{\rm LB}_{\rm inter}(\omega) = 0$ for $\omega < 2|E_F|$. Importantly, the f-sum rule [23] demands that the total spectral weight of the Dirac electrons ($\int_0^\infty G^{\rm LB}_{\rm tot} d\omega$) is conserved [24, 25]. Therefore, the loss of spectral weight in $G^{\rm LB}_{\rm inter}(\omega)$ must be compensated by a gain in $G^{\rm LB}_{\rm intra}(\omega)$,

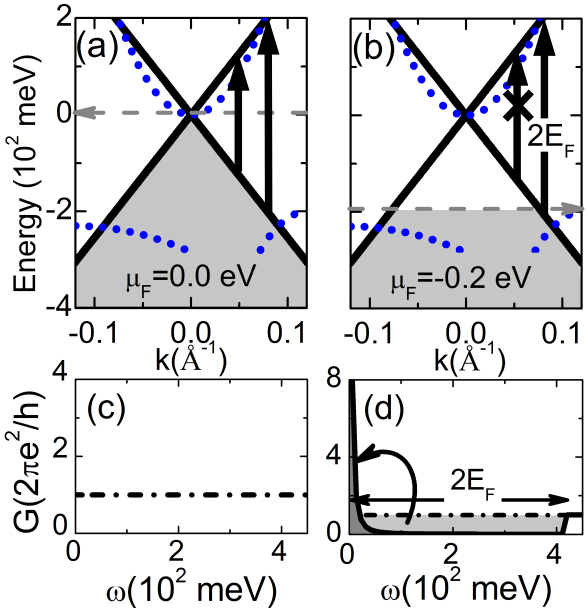

FIG. 1: Panel (a) and (b) show the band structure of a model TI, where the SS is composed of simple linear bands. The bulk band structure is also indicated by the dotted blue lines, consisting of two hole bands and an electron band. In (a) E_F is at the Dirac point, while in (b), E_F is 0.20 eV below the Dirac point, where the arrows in both panels schematically show the allowed transitions. In (a), interband transitions are allowed at all energies, giving rise to the conductance spectra shown in (c). When E_F is shifted below the Dirac point, transitions cannot occur from the depopulated region. Thus, transitions are suppressed below $2|E_F|$, giving rise to the conductance spectra shown in (e).

leading to:

$$G^{\rm LB}_{\rm inter} \cdot 2|E_F| = \int_0^\infty G^{\rm LB}_{\rm intra} \hbar d\omega. \quad (1)$$

It is customary to express the relationship between intraband spectral weight and Drude oscillator strength (D^s_{LB}) as:

$$\int_0^\infty G_{intra,LB}(\omega)\hbar d\omega = \frac{\pi}{30\Omega} D^s_{LB}, \quad (2)$$

which implies the simple relationship between E_F and D^s_{LB}:

$$D^s_{LB} = \frac{60\Omega}{\pi} G^{\rm LB}_{\rm inter}|E_F|. \quad (3)$$

The conductivity of the TI is dominated by the surface states, provide E_F is located outside of the bulk bands. It is customary to measure E_F relative to the Dirac point. Then, for p-type conductivity, E_F, defined relative to the Dirac point, cannot lie within the bulk bands. Therefore, for p-type (n-type) SS conduction, the maximum value of D^s_{LB}, occurs when the Dirac point is at the conduction band minimum (valence band maximum), and E_F is at the valence band maximum (conduction band minimum). Then, the energy that corresponds to the maximum value of E_F is simply E_g, with the maximum value of $D^s_{\rm LB}$ ($D^s_{\rm LB,max}$) given by $D^s_{\rm LB,max} = (N_S)\frac{60\Omega}{\pi} G_{\rm TI} E_g$, where N_S is the number of probed interfaces between systems with different topological indices. Here, $N_S = 2$ since optical transmission probes two topological surfaces, *i.e.* the interface between the film and the vacuum, and the film and the substrate.

TABLE I: Bulk band gap E_g, experimental Drude oscillator strength D from transmission based experiments, modeled Drude oscillator strength $D^s_{\rm 3layer}$ where conductance is assumed to be from 2 topological surfaces with an insulating bulk, and theoretical upper bound of the free Dirac SS Drude oscillator strength $D^s_{\rm LB,max}$ of prototypical strong TIs obtained using the bulk energy gap, and assuming bands with linear dispersion. Values of D are all taken from low-temperature data (6–20 K). All units are meV. †The results of Ref.[11] are from reflection based experiments, and thus only one topological surface is considered.

TI material	E_g	D	$D^s_{\rm 3layer}$	$D^s_{LB,\rm max}$	Refs.
Bi_2Se_3	300	0.5–0.70	–	0.17	[2, 17]
Bi_2Te_3	142	1.26	–	0.08	[7]
$Bi_{1.5}Sb_{0.5}Te_{1.8}Se_{1.2}$	340	1.42	–	0.20	[27]
Bi_2Te_2Se†	290	–	0.83	0.08	[11]
$Bi_{0.92}Sb_{1.08}Te_3$	207	0.13	0.14	0.12	This work
Bi_2Se_3	300	0.09	–	0.17	[56]

The straightforward relationship between D_{LB} and E_F in Eqn. 3 is useful for estimating the Drude spectral weight that could arise from SS, and for intuitively understanding the spectral weight transfer in TI systems. Importantly, the resilience of the f-sum rule to external parameters has been experimentally verified numerous times in a prototypical surface state conductor: graphene. Even with gating, where additional charge carriers are introduced, the f-sum rule is found to be correct within the experimental error bars [37]. The reason for this is that the dominant effect of gating is a redistribution of spectral weight, while the increase in carriers is a relatively small perturbation. Importantly, TI materials are often not well modeled by linear bands. Instead, significant hexagonal warping effects are observed in Bi_2Te_3 and $(Bi,Sb)_2Te_3$ as well as asymmetry between the upper and lower portions of the Dirac cone [4, 10]. These combined effects modify the conductance from the constant value shown in Fig. 1c, and therefore change the value of D^s for a given E_F [20]. To more accurately estimate $D^s_{\rm max}$, we have considered the optical response of a more realistic SS dispersion of a similarly doped sample [10, 20]. This additional analysis suggests that $D^s_{\rm max}$ could be enhanced by approximately 25%, depending on the sample in question [21].

In light of the constraints on D imposed by the f-sum rules, we experimentally determined D and E_g of

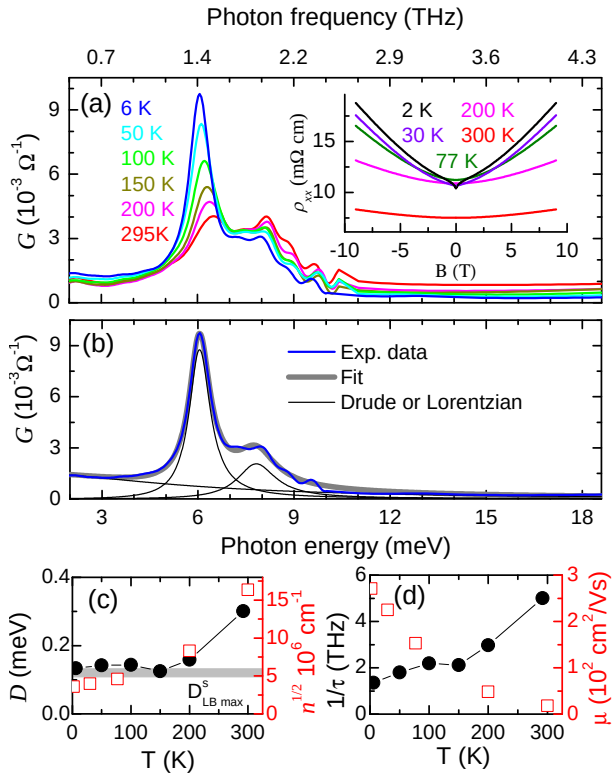

FIG. 2: a) Temperature dependent spectrum of the conductance of $(Bi,Sb)_2Te_3$ obtained from THz-TDS and FTIR. The inset shows (symmetrized) DC magneto-resistivity data for this film. The "cusp" at low-B field in the 2 K data is indicative of weak antilocalization. b) Drude-Lorentz model fit (Eq. 4) (gray) of the 6 K experimental conductance (blue). Individual Drude or Lorentzian oscillators are displayed as thin black lines. c) Temperature dependence of the Drude oscillator strength D (black points) corresponding the the left axis, and square root of the charge carrier density n (open red squares) extracted from Hall effect measurements, corresponding the the right axis. The upper bounds for the Drude response of the linear SS ($D^s_{\mathrm{LB,max}}\pm10\%$), and more realistic estimate of the SS contribution including hexagonal warping ($D^s_{\mathrm{BST,max}}$) in $(Bi,Sb)_2Te_3$, are indicated by the light gray and dark gray bars, respectively. d) Temperature dependence of the free carrier scattering rate $1/\tau$ from optics (black points, left axis) and charge mobility μ from Hall effect measurements (open red squares, right axis).

$(Bi,Sb)_2Te_3$ through a combination of terahertz time-domain spectroscopy (THz-TDS) and Fourier transform infrared spectroscopy (FTIR). Details of the sample growth and experimental techniques are included in [21]. In Fig. 2a, we plot the frequency dependent conductance spectra $G(\omega)$ (real part of $\tilde{\sigma}(\omega)\times$ thickness) of our $(Bi,Sb)_2Te_3$ film from 2–18 meV, the relevant energy scale for the Drude response. The most prominent feature at all temperatures is the narrow resonance centered near 6 meV, which is attributed to a phonon. There is another, weaker phonon resonance, slightly higher in energy, at roughly 8 meV. The assignment of these phonon modes is discussed in [21] At energies below these features, we observe a relatively flat but finite $G(\omega)$ related to the Drude response.

To quantify the Drude oscillator strength D, we construct a Drude-Lorentz model to describe the conductance of the BST system:

$$G(\omega)=\frac{1}{15\Omega}\frac{D\tau}{1+(\omega\tau)^2}+\sum_{j=1}^{n}\frac{A_j\Gamma_j\omega^2}{(\omega^2-\omega_{0j}^2)^2+(\Gamma_j\omega)^2}, \quad (4)$$

where the first term describes the Drude response and the second term corresponds to Lorentzian resonances with amplitude A, width Γ, and center frequency ω_0. A representative Drude-Lorentz model fit is shown in Fig. 2b for the 6 K spectrum. The Drude-Lorentz models and the corresponding data at higher temperatures are shown in [21]. To independently confirm the values of D obtained from the model, we also considered a multilayer model where the BST system consisted of two conducting surface layers and an insulating bulk [21]. The Drude oscillator strength attributed to these conducting surface layers was consistent with the value of D extracted from the measured $G(\omega)$ spectra.

Above 150 K, both D and $1/\tau$ for $(Bi,Sb)_2Te_3$ (black points Fig. 2c and 2d) decrease as temperature is reduced. Since $D\propto\sqrt{n}$, for 2D Dirac states, the reduction in D indicates a reduction in charge carriers, consistent with thermal activation at higher temperatures.The charge carrier density (n) extracted from Hall effect measurements on this film indicate p-type conduction, as is typical of $(Bi_{1-x}Sb_x)_2Te_3$ systems [10, 12], with thermal activation at T>150K (open red squares Fig. 2c). Moreover, the magneto-resistance data at 200K and 300K, shown in the inset to Fig. 2a, show a nonlinear trend with magnetic field B, suggesting multi-band transport, also consistent with the notion of thermally activated bulk states coexisting with SS. Furthermore, since thermally activated charge carriers are not topologically protected, it is likely that their presence would yield a lower mobility (larger $1/\tau$). Therefore, the simultaneous decrease in D and $1/\tau$ as the temperature is lowered, supports the notion that thermally activated charge carriers are frozen out, leaving only high mobility carriers. The Hall parameters n and μ for these high temperature data in Fig. 2 were fit only in the linear regime (-1 T<B<1 T).

Below 150 K, D and $1/\tau$ remain relatively constant near 0.13 meV and 2 THz, respectively. Importantly, our low temperature D value lies right at the threshold of $D^s_{\mathrm{LB,max}}\pm10\%$ for $(Bi,Sb)_2Te_3$, indicated by the light gray bar in Fig. 2c. This latter fact indicates that the measured Drude spectral weight is consistent with a SS dominated response. Moreover, the low temperature value of D is roughly 5 to 10 times lower than most of those previously reported for other representative TIs (see Table I), with the exception of a recent

report in Cu-doped Bi_2Se_3 [56]. The next lowest experimentally measured D come from Bi_2Se_3 [17], in which the D values suggest E_F is in the bulk conduction band. However in this latter case, corrections to the Bi_2Se_3 SS band structure mentioned earlier [17] and the persistence of 2D topological SSs to well above the conduction band minimum [16], do imply D is consistent with a significant SS transport component in Bi_2Se_3.

To verify that the measured parameters $D^{\rm exp}$=0.13 meV and $n_{2D}^{\rm exp}$=1.3×10^{13}cm^{-2} are consistent with surface dominated transport, we can compare our results to that expected from experimentally obtained SS dispersion of TI materials with a similar $(Bi,Sb)_2Te_3$ composition [10, 12]. For this comparison we will utilize Eqn. 3, and the relationship between E_F and and the carrier density of a single surface (n_{SS}) [37]:

$$E_F = \hbar v_0 \sqrt{4\pi n_{SS}}, \tag{5}$$

to directly relate E_F, n_{SS} and D via:

$$D_{LB}^{s} = (N_s)\frac{60\Omega}{\pi} G_{TI} \hbar v_0 \sqrt{4\pi n_{SS}}. \tag{6}$$

For direct comparison between transport and optics, we use $n_{SS} = n_{2D}^{exp}/2$, assuming the carriers were divided between two surfaces and v_0=3.8×10^{5}m/s, consistent with both photoemission and transport measurements of p-type SS carriers [12, 13]. These values yield D_{LB}^{s}=0.13 meV, exactly reproducing our measured value. Moreover, this analysis corresponds to E_F=0.22 eV, in close agreement with the measured E_G of 0.206 eV, suggesting the optical response is close to the constraints imposed by the sum rule analysis. If the additional contribution to D from the bulk bands is taken into account, in the regime where $E_F \approx E_G$, we recover our experimentally measured value of D when $E_F = 0.211$eV. This analysis, which is detailed in [21], suggests E_F penetrates only up to 5 meV into the bulk bands, and bulk carriers constitute only 2% of the total n_{2D}. Hexagonal warping, and other modifications to the SS dispersion, discussed more in [21], suggest that even smaller values of E_F, where $E_F < E_G$, may reproduce the measured D and $n_{2D}^{\rm exp}$ values. Alternatively, we considered the case that the optical response arises from the bulk valence bands. Prior work has shown that the bulk valence band structure in the $(Bi,Sb)_2Te_3$ system consist of of a light hole band (LHB) and heavy hole band (HHB) [3, 14, 15]. The effective masses of LHB and the HHB are 0.11m_e and 1.0 m_e respectively, and the top of the HHB is approximately 30 meV below the LHB [14, 15]. To recover the correct value of D for this sample, E_F would be 30 meV below the top of the LHB, However, this would yield an n_{2D} of 5 × 10^{12}cm^{-2}, over a factor of two smaller than what was measured.

Prior experiments have also pointed out that conventional SS may coexist with the topologically protected SS of a TI [16]. Typically, the conventional SS appear as a result of band bending, and are close in energy to the bulk bands, and contribute to conductivity when $E_F \approx E_G$. Therefore, the large magnitude E_F required to access the conventional SS, combined with their additional metallic response, would likely push D well above the limit imposed by f-sum rule analysis. It is therefore unlikely that our measured response has a significant component from the conventional SS.

Further evidence of a dominant topological SS contribution to the response of the $(Bi,Sb)_2Te_3$ system is identified in magneto-resistance measurements, plotted in the inset to Fig. 2. At low temperature, the magneto-resistance data show linear behavior with B, indicative of single band transport. Moreover, the figure reveals weak antilocalization in (symmetrized) magnetoresistance measurements (*i.e.* the cusp at low-B field in the 2K data), which has been thought to be a hallmark signature topological SS transport [35]. However, theoretical work has shown that antilocalization may occur even in systems with bulk dominated transport, and therefore, anti localization by itself, cannot be considered conclusive evidence for surface states [36]. Despite this stipulation, the magneto-resistance data, in conjuction with the low value of D revealed from optics, are consistent with low temperature transport that is dominated by topologically protected SS.

The picture that emerges from the comparison of transport, optics and previously measured photoemission is that the Dirac point is near the bottom of the conduction band, with E_F near the top of the valence band, as is illustrated in Fig. 1b. Likewise, we illustrate the bulk band structure consisting of two hole bands offset from the Γ point, with an electron band at the Γ point, as measured previously [10, 12, 15]. The relative positioning of E_F, the Dirac point, and bulk bands that we propose has been observed in similar, albeit more highly doped, $(Bi_{1-x}Sb_x)_2Te_3$ thin films [12]. Indeed, the distance from the Dirac point to the top of the Valence Band, as well as the v_0 in this latter work is consistent with our measurements. It is important to note that photoemission measurements of similarly doped samples often show that the Dirac point is below E_F [10], in contrast to our measurement of p-type carriers. This discrepancy can be understood in light of the aging effects, often observed TI systems, which generally shift the Dirac point downward relative to E_F. It is then likely that the capping layer of Se prevents the band bending, and the resulting accumulation layers, observed in other samples [13, 16] yielding a response dominated by p-type SS carriers. Unfortunately, this same capping layer prevents direct verification of this hypothesis in our particular sample via photoemission.

The totality of our data clearly show that $(Bi,Sb)_2Te_3$ alloys are a promising vehicle for advancing the field of TIs. In particular, the Drude oscillator strength lies at

the threshold of the SS response upper bound, demonstrating the effectiveness of the alloy in reducing or eliminating bulk charge carriers. Furthermore, comparison of Drude parameters from optics to transport parameters from the Hall effect, coupled with weak antilocalization observed in low-temperature magneto-transport, shows consistency with surface transport with $v_0 \sim 4 \times 10^5$ m/s, in accord with ARPES results [12]. Therefore, our data imply $(Bi,Sb)_2Te_3$ is an optimal candidate for isolating SS properties in further studies of keen interest, such as magneto-optical measurements [38, 40, 41] and electrostatic modification of SS charge carrier density in electric field effect devices [22, 28, 42].

Sample growth and characterization at Penn State was supported by DARPA (N66001-11-1-4110), ONR (N00014-12-1-0117), ARO-MURI (W911NF-12-1-0461) and (ECS-0335765). Work at UCSD is supported by DOE-BES.

[1] M. Hasan and C. Kane, Reviews of Modern Physics **82**, 3045 (2010), ISSN 0034-6861, URL http://link.aps.org/doi/10.1103/RevModPhys.82.3045.

[2] Y. Xia, D. Qian, D. Hsieh, L. Wray, a. Pal, H. Lin, a. Bansil, D. Grauer, Y. S. Hor, R. J. Cava, et al., Nature Physics **5**, 398 (2009), ISSN 1745-2473, URL http://www.nature.com/doifinder/10.1038/nphys1274.

[3] Y. L. Chen, J. G. Analytis, J.-H. Chu, Z. K. Liu, S.-K. Mo, X. L. Qi, H. J. Zhang, D. H. Lu, X. Dai, Z. Fang, et al., Science (New York, N.Y.) **325**, 178 (2009), ISSN 1095-9203, URL http://www.ncbi.nlm.nih.gov/pubmed/19520912.

[4] D. Hsieh, Y. Xia, D. Qian, L. Wray, F. Meier, J. Dil, J. Osterwalder, L. Patthey, a. Fedorov, H. Lin, et al., Physical Review Letters **103**, 146401 (2009), ISSN 0031-9007, URL http://link.aps.org/doi/10.1103/PhysRevLett.103.146401.

[5] S.-Y. Xu, M. Neupane, C. Liu, D. Zhang, A. Richardella, L. A. Wray, N. Alidoust, M. Leandersson, T. Balasubramanian, J. Sánchez-barriga, et al., Nature Physics **8**, 616 (2012), ISSN 1745-2473, URL http://dx.doi.org/10.1038/nphys2351.

[6] P. Di Pietro, F. M. Vitucci, D. Nicoletti, L. Baldassarre, P. Calvani, R. Cava, Y. S. Hor, U. Schade, and S. Lupi, Physical Review B **86**, 045439 (2012), ISSN 1098-0121, URL http://link.aps.org/doi/10.1103/PhysRevB.86.045439.

[7] B. C. Chapler, K. W. Post, A. R. Richardella, J. S. Lee, J. Tao, N. Samarth, and D. N. Basov, Physical Review B **89**, 235308 (2014), ISSN 1550235X, 1405.4916.

[8] A. Hashibon and C. Elsässer, Physical Review B **84**, 144117 (2011), ISSN 1098-0121, URL http://link.aps.org/doi/10.1103/PhysRevB.84.144117.

[9] Y. Jiang, Y. Y. Sun, M. Chen, Y. Wang, Z. Li, C. Song, K. He, L. Wang, X. Chen, Q.-K. Xue, et al., Physical Review Letters **108**, 066809 (2012), ISSN 0031-9007, URL http://link.aps.org/doi/10.1103/PhysRevLett.108.066809.

[10] D. Kong, Y. Chen, J. J. Cha, Q. Zhang, J. G. Analytis, K. Lai, Z. Liu, S. S. Hong, K. J. Koski, S.-K. Mo, et al., Nature nanotechnology **6**, 705 (2011), ISSN 1748-3395, URL http://www.ncbi.nlm.nih.gov/pubmed/21963714.

[11] A. A. Reijnders, Y. Tian, L. J. Sandilands, G. Pohl, I. D. Kivlichan, S. Y. F. Zhao, S. Jia, M. E. Charles, R. J. Cava, N. Alidoust, et al., Physical Review B **89**, 075138 (2014), ISSN 1098-0121, URL http://link.aps.org/doi/10.1103/PhysRevB.89.075138.

[12] J. Zhang, C.-Z. Chang, Z. Zhang, J. Wen, X. Feng, K. Li, M. Liu, K. He, L. Wang, X. Chen, et al., Nature communications **2**, 574 (2011), ISSN 2041-1723, URL http://www.ncbi.nlm.nih.gov/pubmed/22146393.

[13] A. A. Taskin, Z. Ren, S. Sasaki, K. Segawa, and Y. Ando, Physical Review Letters **107**, 016801 (2011), ISSN 0031-9007, URL http://link.aps.org/doi/10.1103/PhysRevLett.107.016801.

[14] V. A. Kulbachinskii, Z. M. Dashevskii, M. Inoue, M. Sasaki, H. Negishi, W. X. Gao, P. Lostak, J. Horak, and A. de Visser, Phys. Rev. B **52**, 10915 (1995).

[15] V. Kulbachinskii, V. Kytin, P. Tarasov, and N. Yuzeeva, Physics of the Solid State **52**, 1830 (2010).

[16] M. S. Bahramy, P. D. C. King, A. de la Torre, J. Chang, M. Shi, L. Patthey, G. Balakrishnan, P. Hofmann, R. Arita, N. Nagaosa, et al., Nature communications **3**, 1159 (2012), ISSN 2041-1723, URL http://www.ncbi.nlm.nih.gov/pubmed/23093196.

[17] L. Wu, M. Brahlek, R. Valdés Aguilar, A. V. Stier, C. M. Morris, Y. Lubashevsky, L. S. Bilbro, N. Bansal, S. Oh, and N. P. Armitage, Nature Physics **9**, 410 (2013), ISSN 1745-2473, URL http://www.nature.com/doifinder/10.1038/nphys2647.

[56] L. Wu, W.-K. Tse, M. Brahlek, C. Morris, R. Valdès Aguilar, N. Koirala, S. Oh, and N. Armitage (2015), 1502.04577v1.

[19] It is unclear optical transitions between SSs and bulk states are theoretically possilbe due to symmetry and spatial separation, but moreover, such transitions cannot change the key result (Eq. 3) relating the SS Drude oscillator strength to the Fermi level.

[20] Z. Li and J. P. Carbotte, Physical Review B **87**, 155416 (2013), ISSN 1098-0121, URL http://link.aps.org/doi/10.1103/PhysRevB.87.155416.

[21] See Supplemental Material, which includes Refs. [43–56]

[22] Z. Q. Li, E. A. Henriksen, Z. Jiang, Z. Hao, M. C. Martin, P. Kim, H. L. Stormer, and D. N. Basov, Nature Physics **4**, 532 (2008), ISSN 1745-2473, URL http://www.nature.com/doifinder/10.1038/nphys989.

[23] D. N. Basov, R. Averitt, D. van der Marel, M. Dressel, and K. Haule, Rev. Mod. Phys. **83**, 471 (2011), ISSN 0034-6861, URL http://link.aps.org/doi/10.1103/RevModPhys.83.471.

[24] V. P. Gusynin, S. G. Sharapov, and J. P. Carbotte, Physical Review B **75**, 165407 (2007), ISSN 1098-0121, URL http://link.aps.org/doi/10.1103/PhysRevB.75.165407.

[25] J. Sabio, J. Nilsson, and A. H. Castro Neto, Physical Review B **78**, 075410 (2008), ISSN 1098-0121, URL http://link.aps.org/doi/10.1103/PhysRevB.78.075410.

[26] K. W. Post, B. C. Chapler, L. He, X. Kou, K. L. Wang, and D. N. Basov, Phys. Rev. B **88**, 075121 (2013).

[27] C. S. Tang, B. Xia, X. Zou, S. Chen, H.-W. Ou, L. Wang, a. Rusydi, J.-X. Zhu, and E. E. M. Chia, Scientific reports **3**, 3513 (2013), ISSN 2045-2322, URL http://www.pubmedcentral.nih.gov/articlerender.

fcgi?artid=3865466\&tool=pmcentrez\&rendertype=abstract.

[28] J. Horng, C.-F. Chen, B. Geng, C. Girit, Y. Zhang, Z. Hao, H. a. Bechtel, M. Martin, A. Zettl, M. F. Crommie, et al., Physical Review B **83**, 165113 (2011), ISSN 1098-0121, URL http://link.aps.org/doi/10.1103/PhysRevB.83.165113.

[29] E_F may be slighlty into the bulk band and still have an isolated SS Drude response for the special case of a disorder-induced localization of the bulk states.

[30] L. Fu, Physical Review Letters **103**, 266801 (2009), ISSN 0031-9007, URL http://link.aps.org/doi/10.1103/PhysRevLett.103.266801.

[31] K. Kechedzhi and S. Das Sarma, Physical Review B **88**, 085403 (2013), ISSN 1098-0121, URL http://link.aps.org/doi/10.1103/PhysRevB.88.085403.

[32] M. Yu, Peter Y., Cardona, *Fundamentals of semiconductors* (Springer-Verlag, Berlin, 2010), 4th ed., ISBN 9783642007095.

[33] L. A. Wray, S. Xu, Y. Xia, D. Qian, A. V. Fedorov, H. Lin, A. Bansil, L. Fu, Y. S. Hor, R. J. Cava, et al., Phys. Rev. B **83**, 224516 (2011), URL http://link.aps.org/doi/10.1103/PhysRevB.83.224516.

[34] W. Richter, H. Kohler, and C. Becker, Phys. Stat. Sol. (b) **84**, 619 (1977).

[35] L. Fu and C. L. Kane, Physical Review B **76**, 045302 (2007), ISSN 1098-0121, URL http://link.aps.org/doi/10.1103/PhysRevB.76.045302.

[36] I. Garate and L. Glazman, Phys. Rev. B **86**, 035422 (2012), URL http://link.aps.org/doi/10.1103/PhysRevB.86.035422.

[37] D. N. Basov, M. M. Fogler, a. Lanzara, F. Wang, and Y. Zhang, Reviews of Modern Physics **86**, 959 (2014), ISSN 15390756, 1407.6721.

[38] A. A. Schafgans, K. W. Post, A. A. Taskin, Y. Ando, X.-L. Qi, B. C. Chapler, and D. N. Basov, Physical Review B **85**, 195440 (2012), ISSN 1098-0121, URL http://link.aps.org/doi/10.1103/PhysRevB.85.195440.

[39] E. H. Hwang and S. Das Sarma, Phys. Rev. B **75**, 205418 (2007), URL http://link.aps.org/doi/10.1103/PhysRevB.75.205418.

[40] W.-K. Tse and A. H. MacDonald, Physical Review Letters **105**, 057401 (2010), ISSN 0031-9007, URL http://link.aps.org/doi/10.1103/PhysRevLett.105.057401.

[41] J. Maciejko, X.-L. Qi, H. D. Drew, and S.-C. Zhang, Physical Review Letters **105**, 166803 (2010), ISSN 0031-9007, URL http://link.aps.org/doi/10.1103/PhysRevLett.105.166803.

[42] G. S. Jenkins, D. C. Schmadel, A. B. Sushkov, H. D. Drew, M. Bichler, G. Koblmueller, M. Brahlek, N. Bansal, and S. Oh, Physical Review B **87**, 155126 (2013), ISSN 1098-0121, URL http://link.aps.org/doi/10.1103/PhysRevB.87.155126.

[43] M. Dressel and G. Gruener, *Electrodynamics of Solids: Optical Properties of Electrons in Matter*, vol. 70 (Cambridge University Press, New York, New York, USA, 2002), 1st ed., ISBN 0 511 01439 2.

[44] C.-X. Liu, X.-L. Qi, H. J. Zhang, X. Dai, Z. Fang, and S.-C. Zhang, Phys. Rev. B **82**, 045122 (2010), URL http://link.aps.org/doi/10.1103/PhysRevB.82.045122.

[45] B. Wunsch, T. Stauber, F. Sols, and F. Guinea, New Journal of Physics **8**, 318 (2006), ISSN 1367-2630, URL http://stacks.iop.org/1367-2630/8/i=12/a=318?key=crossref.1ac84bfc3f85e7367b9a281336c35735.

[46] C. Kittel, *Introduction to Solid State Physics* (2005), 8th ed.

[47] A. B. Kuzmenko, Review of Scientific Instruments **76**, 083108 (2005), ISSN 00346748, URL http://link.aip.org/link/RSINAK/v76/i8/p083108/s1&Agg=doi.

[48] R. D. Averitt and A. J. Taylor, Journal of Physics: Condensed Matter **14**, R1357 (2002).

[49] R. Yu, W. Zhang, H.-J. Zhang, S.-C. Zhang, X. Dai, and Z. Fang, Science (New York, N.Y.) **329**, 61 (2010), ISSN 1095-9203, URL http://www.ncbi.nlm.nih.gov/pubmed/20522741.

[50] K. W. Post, Y. S. Lee, B. C. Chapler, A. A. Schafgans, M. Novak, A. A. Taskin, K. Segawa, M. D. Goldflam, H. T. Stinson, Y. Ando, et al., Phys. Rev. B **91**, 165202 (2015), URL http://link.aps.org/doi/10.1103/PhysRevB.91.165202.

[51] R. Valdés Aguilar, A. V. Stier, W. Liu, L. S. Bilbro, D. K. George, N. Bansal, L. Wu, J. Cerne, a. G. Markelz, S. Oh, et al., Physical Review Letters **108**, 087403 (2012), ISSN 0031-9007, URL http://link.aps.org/doi/10.1103/PhysRevLett.108.087403.

[52] H. Zhang, C.-X. Liu, X.-L. Qi, X. Dai, Z. Fang, and S.-C. Zhang, Nature Physics **5**, 438 (2009), ISSN 1745-2473, URL http://www.nature.com/doifinder/10.1038/nphys1270.

[53] D. Hsieh, Y. Xia, D. Qian, L. Wray, J. H. Dil, F. Meier, J. Osterwalder, L. Patthey, J. G. Checkelsky, N. P. Ong, et al., Nature **460**, 1101 (2009), ISSN 1476-4687, URL http://www.ncbi.nlm.nih.gov/pubmed/19620959.

[54] M. Neupane, S.-Y. Xu, L. A. Wray, A. Petersen, R. Shankar, N. Alidoust, C. Liu, a. Fedorov, H. Ji, J. M. Allred, et al., Physical Review B **85**, 235406 (2012), ISSN 1098-0121, URL http://link.aps.org/doi/10.1103/PhysRevB.85.235406.

[55] T. Arakane, T. Sato, S. Souma, K. Kosaka, K. Nakayama, M. Komatsu, T. Takahashi, Z. Ren, K. Segawa, and Y. Ando, Nature communications **3**, 636 (2012), ISSN 2041-1723, URL http://www.ncbi.nlm.nih.gov/pubmed/22273674.

[56] L. Wu, W.-K. Tse, M. Brahlek, C. Morris, R. Valdés Aguilar, N. Koirala, S. Oh, and N. Armitage (2015), 1502.04577v1.