A Complete Analysis of the Intrinsic Properties of the Topological States in Bi_2Se_3 by Angle Resolved Photoemission Spectroscopy

E. J. Reichwein[1,*] and G.H. Gweon[1]

[1]*Department of Physics, University of California, Santa Cruz, CA 95064*

(Dated: July 31, 2014)

We present the first complete analysis of Angle Resolved Photoemission Spectroscopy (ARPES) line shapes using both energy distributed curves (EDC) and momentum distributed curves (MDC). The topologically protected surface states of Bi_2Se_3 have been studied previously but exclusively by MDC analysis. By using Shastry's Fermi liquid model, we were able to extract a coherent picture of the surface states in topological insulators. From there we determined the electron-phonon coupling constant to be the smallest ever observed in any material which definitively shows surface states are topologically protected. We have also drawn conclusions about the intrinsic nature of the high T_c superconductor background through the comparison of the topological states in Bi_2Se3.

A new class of exotic materials called topological insulators (TI) have recently been discovered. They are insulating in the bulk and have metallic surface states because of the non-trivial topology of their Hamiltonian. An important characteristic of TI's is that the spin-orbit coupling energy is of the same order as the bulk band gap. The surface states are very robust against perturbations such as defects and nonmagnetic impurities because they are topologically protected by time reversal symmetry. Arguably, the most common TI is Bi_2Se_3 which has been studied extensively using angle resolved photoemission spectroscopy (ARPES) but not completely. We present in this letter a complete line shape analysis of Bi_2Se_3 with momentum distributed curves (MDC) and energy distributed curves (EDC) treated on equal footing .

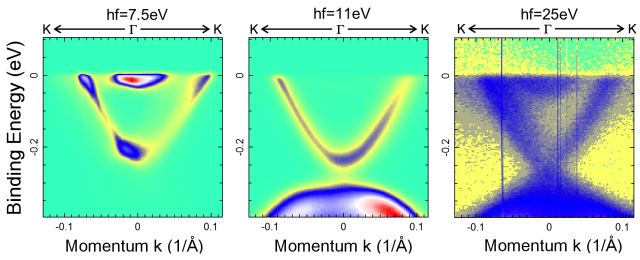

FIG. 1: Shows the effect of the ARPES spectra with various photon energies. Note that the bulk conduction band extends approximately 40meV under the Fermi edge.

The Bi_2Se_3 samples were grown at Brookhaven National Laboratory by standard self-flux techniques by Dr. Genda Gu. The ARPES data was taken exclusively on beamline 5-4 at Stanford's Synchrotron Radiation Lightsource. The Bi_2Se_3 samples were cleaved *in situ* in a ultra high vacuum chamber of pressure $\sim 5 \times 10^{-11}$ Torr. We explored many photon energies between 7.5 eV to 25 eV, but have found that 11 eV produced the cleanest data. We produce the photon energies of 7.5eV, 11eV, and 25eV below in Figure 1.

The energy resolution is 15meV and the angular resolution is 0.3°. We took data from 300K down to 25K, in steps of 25K. The aim of this project is to combine the EDC and MDC analysis of the samples into one coherent picture by extracting the . We will use Lorentzian based Fermi Liquid Theory (FLS) for the EDC's and purely Lorentzian analysis for the MDC's. The reason for the discrepancy in fit models for EDC and MDC is because many-body interactions, such as electron-electron and phonon coupling, effect the distribution in energy more than the distribution in momentum.

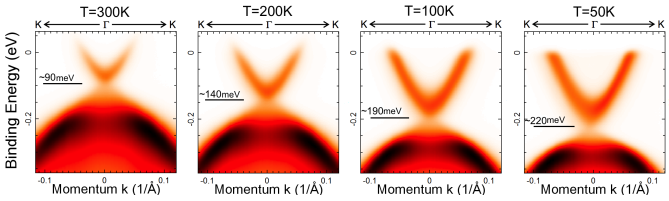

FIG. 2: Shows the increase in binding energy of the Dirac point with respect to temperature. Notice how for each $\Delta T =$ 100K we see an approximate increase of binding energy of 50meV.

In Figure 2 we see the effect of changing the temperature on the ARPES spectra, by increasing the energy of the Dirac point with decreasing temperature. We also see an absence in temperature broadening in the spectra, indicating a lack of strong electron-electron and electron-phonon coupling. Comparing our spectra to Pan *et. al.* and Park *et. al.*'s reported spectra, we see that our sample has the lowest energy Dirac point. This indicates are samples are in fact of better quality than theirs. The ideal sample would put the Fermi edge at the point of the Dirac cone.

The EDCs give us information on how the energy of electrons is distributed at certain k values. The reason there is a spread in energy values is because of the many

*Electronic address: ereichwe@ucsc.edu; URL: `http://www.ericreichwein.com`

body effects and interactions and non-zero temperature, and as mentioned previously alter the distribution of energies more than it does to the spread in momenta. Due to these consequences, we must use the more sophisticated FLS theory for the EDC's. There are three main (different than those established in section 1.2) parameters that are introduced by using the FLS theory which are: the ARPES kink ω_0, the quasi-particle residue Z, and the lifetime relation η. The results of the EDC fits are shown in Figure 3 for T =50K, 150K and 300K.

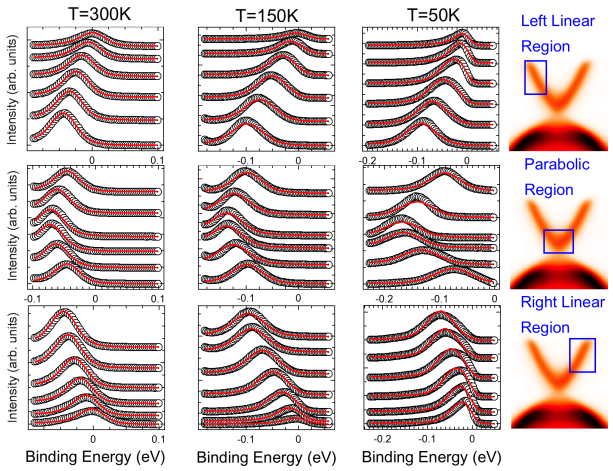

FIG. 3: FLS EDC results. We present the fits for T = 50K, 150K and 300K. The columns represent the EDC fits for the three temperatures and the rows represent the three different dispersion, $\epsilon(k)$, regions: left linear, parabolic and right linear. We present (on the far right of each column) generic ARPES spectra with the corresponding $\epsilon(k)$ region boxed.

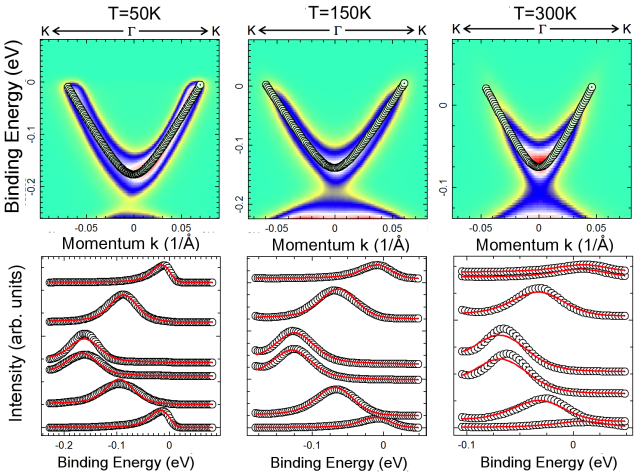

FIG. 4: FLS EDC results. We present the fits for T = 50K, 150K and 300K. The columns represent the EDC fits for the three temperatures. The first row is the fitted dispersion $\epsilon(k)$ of **??** over the ARPES spectra. The second row shows the EDC line shape fits for the dispersion $\epsilon(k)$.

In Figure 4 we see that the dispersion fit function for T=50K, 150K and 300K is

$$\epsilon(k) = v_f(\sqrt{|k-\delta k|^2 + \Delta_v^2} - \Delta E) \qquad (1)$$

Where v_f is the Fermi velocity, k is the momentum, δk is Dirac point momentum shift from Γ, $\Delta_v \equiv \frac{\Delta}{v_f}$, Δ is the gap formed where the Dirac cone should be and ΔE is to fix the Fermi crossing point. Since Δ_v is small, we see that in the limiting case of $k \to 0$, **??** becomes parabolic and for k large **??** becomes approximately linear. We determined this by first fitting the dispersion with free parameters then determine the parameters of the fit function for the $\epsilon(k) = a(\sqrt{|k-b^2+c^2} - d)$. As seen in Figure 4, the determined dispersion is actually a very good fit. We kept the T=50K FLS parameters [1] fixed for the rest of the analysis. The Fermi velocities for the fit function (**??**) of T=50K, 150K and 300K were ≈ 4.0, 3.85 and 3.5 $eV/$Å, respectively. These values are almost a perfect match for the EDC and MDC self-energy comparisons. The T=150K EDC determined v_f is slightly higher than the T=150 MDC v_f, which we believe is due the robustness of the FLS theory. To obtain more detail from the EDC line shape analysis we re-fit the dispersion in three different regions with the same FLS parameters in Figure 3.

As can be seen in he MDC fits are very good, except for some asymmetry in the intensities, as seen in Figure 5. The asymmetric intensities should be expected because positive momentum corresponds to a different crystallographic direction than the negative momentum. The effect is slight and can be noticed in others data, but is neglected due to minimal effect on the actual analysis.

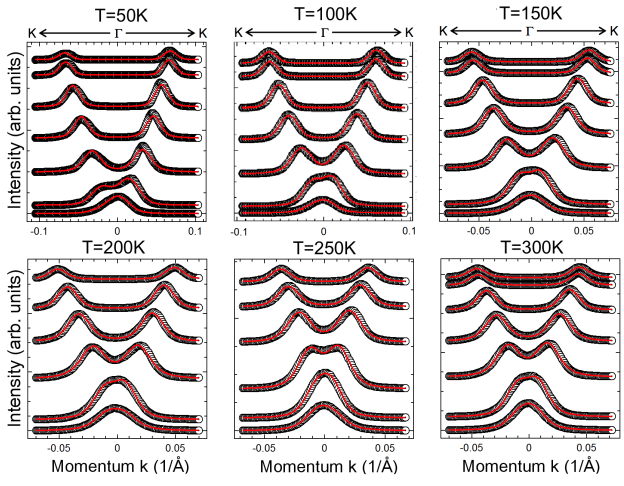

FIG. 5: Double Lorentzian MDC results. We present the fits for T = 50K-300K in increments of 50K.

[1] η, Z, ω_0, Gaussian convolution, etc.

As expected the MDC analysis, shown in Figure 5, can be fit to very good accuracy with just simple Lorentzian's. Although, we don't take into account the fact that TI's are strongly correlated materials we believe this is adequate model because of the excellent fits, as compared to the EDC's without FLS theory applied (even with $\epsilon(k)$ as a free parameter). From our MDC's we can determine the FWHM (constrained to be equal for both peaks), which has units of momentum. Multiplying the FWHM by $v_g/2$ will give us the true imaginary part of the complex self-energy. The dispersion's $\epsilon(k)$ fits the ARPES spectra quite nicely as well.

All the interaction information is stored in the imaginary part of the complex self-energy. To determine the types of interactions in Bi_2Se_3, we have to compare the self-energies of the EDC and MDC analysis. In Figure 6, we present the imaginary part of the complex self-energy which was extracted from the MDC data and are comparing it to the FLS theoretical self-energy with the parameters from our EDC analysis.

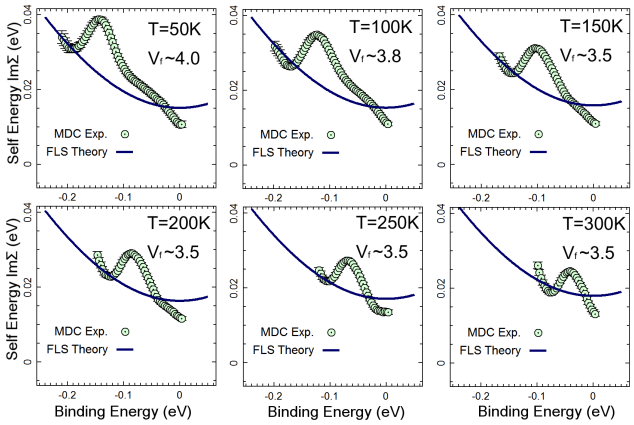

FIG. 6: Comparison of EDC FLS theoretical and MDC actual self-energies for various temperatures.

Notice in Figure 6 the bulk ends around the 50meV point [2]. Impurity and electron-phonon interactions should show a spike in the Σ_{Im} near the Fermi edge, indicating scattering from surface states to bulk and electron-electron interactions dominating at high energies. However, we do not see this exactly. Our results do agree with the electron-electron interactions dominating at high energies by matching an overall increase in Σ_{Im} for increasing binding energy. However, there is no spike in Σ_{Im} for low energy electrons (near Fermi edge), rather there is one for high binding energies. This spike is characteristic of electron-phonon scattering as seen in **??**, but it is not in the same region as the bulk. Since we know the relative change in energy of the initial and final states of these interactions is relatively small, but has to have a large change in momentum, we can conclude that the interacting phonon's have relatively small energies. However, seeing that the spike is quite large, we predict that there is a large number of these small amplitude phonon's that produce coupling between surface states. These intra-surface interactions are concentrated well below the bulk conduction band and have sharp kinks leading into the spike of self-energy.

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

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

At higher temperatures ($k_BT < \Omega_0/3$) we can approximate the electron-phonon coupling constant λ as,

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

just as Pan *et. al.* showed[2]. This tells us how much the surface states are coupling to phonons in the bulk. According to ref. [2], the phonon coupling constants were reported as $\lambda = 0.088 \pm 0.009$ for the first sample and $\lambda = 0.076 \pm 0.007$ for the second sample. They claim these are the lowest values, even lower than the theoretically lower limit value presented by S. Giraud and R. Egger [1]. This however, could be underestimated due to the finite resolution of detector. They also noted the absence of the typical ARPES "kink", which is in agreement with Park *et. al.* [3] and, to our knowledge, all other studies as

[2] exact position of the bottom of the bulk conduction band depends on the temperature

well. This again shows the low energy surface states are well protected from scattering/coupling due to phonons.

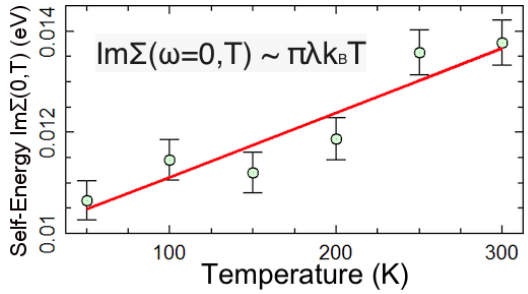

FIG. 7: A plot of $\Sigma_{\mathrm{Im}}(\omega = 0, T)$. The slope tells us the approximate electron-phonon coupling constant λ.

From the slope of the data in Figure 7, we have determined the electron-phonon coupling to be an astonishing low value of $\lambda = 0.049 \pm 0.007$. This value is nearly half as much as the previously reported phonon coupling constant value, and within the same margin of error. This agrees with lack of the "kink" in the phonon energy range we mentioned earlier and shown in **??** and the minimal temperature broadening of the ARPES spectra[2]. However, to determine the temperature broadening accurately would require significantly higher energy resolution than we have now. The small value of the electron-phonon coupling constant and evidence of strongly protected topological states is extremely important for developing new technologies that require precise control of spin properties in materials, as well as new insight into high T_c superconductors.

From Figure 7, we have determined the electron-phonon coupling to be $\lambda = 0.049 \pm 0.007$. This value is nearly half as much as the previously reported phonon coupling constant value, and within the same margin of error. This agrees with lack of the "kink" in the phonon energy range we mentioned earlier and shown in **??** and the minimal temperature broadening of the ARPES spectra[2]. However, to determine the temperature broadening accurately would require significantly higher energy resolution than we have now. The deficiency of electron-phonon coupling and evidence of strongly protected topological states is extremely important for developing new technologies that require precise control of spin properties in materials, as well as new insight into high T_c superconductors.

We have conducted ARPES experiments on Bi_2Se_3 at various photon energies and temperatures. We first noticed hints of inter surface quasi-particle interactions in some dirty data. After obtaining very clean and superior data we performed line shape analysis of both MDC and EDC. By investigating how the electrons momentum and energies are spread out we can determine insightful and valuable information about the strongly correlated electronic system of topological insulators. Specifically, in this study, we were able to observe intra-surface interactions in the topological insulator Bi_2Se_3 by comparing the self-energies extracted from both MDC and EDC analysis. From the imaginary parts of the complex self-energies at Fermi edge we were able to determine the electron-phonon coupling constant to be $\lambda = 0.049 \pm 0.007$, which is the weakest coupling constant reported to date.

When optimally doped the compound $Bi_2Sr_2CaCu_2O_{8+x}$ (Bi-2212) enters a superconducting state at very high temperatures. Topologically protected states of Bi_2Se_3 have similar properties to that Bi-2212. We can compare these properties through the use of EDC analysis with FLS theory.

In Figure 8, we see the EDC taken at the Fermi level for both Bi-2212 and Bi_2Se_3 at T=300K. There are three regions for both materials: above the Fermi level (BE>0eV) , the peak(-0.1<BE<0eV), and the background/bulk (BE<-0.1eV). There is excellent agreement in the EDCs for above the Fermi level and going into the peak region. There is a slight deviation due to peak values being off by $\approx$ 7meV. This results in a lower energy side of the peak slopes being somewhat different. For the higher energy side of the peaks we see greater differences. The main difference is caused by the inherit background of the high temperature superconductor as compared to the simple bulk of the topological insulator.

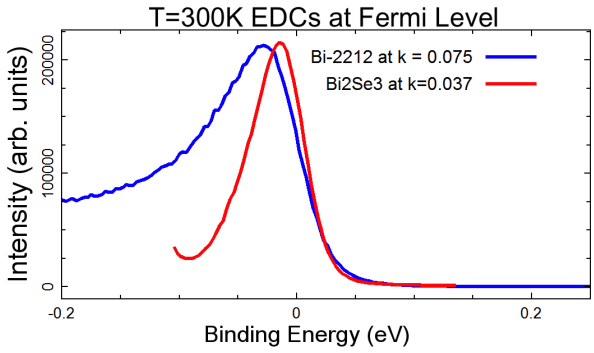

FIG. 8: A comparison of the EDC's taken at the Fermi level for T=300K data. The Bi-2212 and Bi_2Se_3 Fermi momenta values are $k_f = 0.075\text{Å}^{-1}$ and $k_f = 0.037\text{Å}^{-1}$, respectively.

Since the main difference in the EDCs is the background we can assume this is due to some strong scattering effects in Bi-2212. As we have presented in previous sections that the Bi_2Se_3 has highly protected topological states which could, in theory, prevent any background from being observed. Where as Bi-2212, is not topologically protected from certain scattering events because the electronic transport mechanism is different in Bi-2212 (and other high temperature superconductors) than that of TIs. However, an interesting fact is that superconductivity can be induced in TIs by proximity effects and is currently being studied to aid in the development of quantum computers. Now the thing here is we have con-

cise descriptions TIs through very accurate theories, but we cannot describe high temperature superconductors. So by looking at the peaks in the EDCs of the ARPES spectrum we can use TIs to understand the peak, and to help determine the cause of the background, which is the main difference between TIs and high temperature superconductors. In Matsuyama *et. al.*'s paper[**?**] describing the ARPES spectra of Bi-2212 (among others) with both MDC and EDC treated equally, they were able to perfectly fit the ARPES spectra using extremely correlated Fermi-liquid theory (ECFL). Since ECFL theory was able to fit MDC and EDC for multiple materials it indicates that it is possible to establish a phenomenological model capable of describing large classes of high temperature superconductors. This could ultimately lead to breakthroughs in high temperature superconductor theory and experiment which could lead to the ever sought after room temperature superconductor and open up numerous industries such as spintronics and quantum information.

We have conducted ARPES experiments on Bi_2Se_3 at various photon energies and temperatures. We first noticed hints of inter surface quasi-particle interactions in some dirty data. After obtaining very clean and superior data we performed line shape analysis of both MDC and EDC. By investigating how the electrons momentum and energies are spread out we can determine insightful and valuable information about the strongly correlated electronic system of topological insulators. Specifically, in this study, we were able to observe intra-surface interactions in the topological insulator Bi_2Se_3 by comparing the self-energies extracted from both MDC and EDC analysis. From the imaginary parts of the complex self-energies at Fermi edge we were able to determine the electron-phonon coupling constant to be $\lambda = 0.049 \pm 0.007$, which is the weakest coupling constant reported to date.

[1] Giraud, S. and Egger, R., Phys. Rev. B **83**, 245322 (2011).
[2] Pan, Z.-H., Fedorov, A. V., Gardner, D., Lee, Y. S., Chu, S., and Valla, T., Phys. Rev. Lett. **108**, 187001 (2012).
[3] Park, S. R., Jung, W. S., Kim, C., Song, D. J., Kim, C., Kimura, S., Lee, K. D., and Hur, N., Phys. Rev. B **81**, 041405 (2010).