



Quasiparticle scattering and the protected nature of the topological states in a parent topological insulator Bi_2Se_3

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We report on angle-resolved photoemission spectroscopic studies on a parent topological insulator, Bi_2Se_3 . The linewidth of the spectral function (inverse of the quasiparticle lifetime) of the topological metallic states shows an anomalous behavior. This behavior can be reasonably accounted for by assuming the decay of the quasiparticles predominantly into bulk electronic states through electron-electron interaction and defect scattering. Studies on aged surfaces reveal that topological metallic states are very much unaffected by the potentials created by adsorbed atoms or molecules on the surface, indicating that topological states could be indeed protected against weak perturbations.

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Topological insulators (TIs) are materials with bulk gaps due to spin-orbit coupling. TIs are classified into “weak TI” and “strong TI” according to Z_2 topological invariants.^{1,2} Strong TIs have odd number of topological metallic (TM) Dirac cones on the surface which is robust (protected) against disorder or impurities.

TM states realized on the surface of a strong TI are important and could be useful.^{3,4} The properties of the TM are also set by the topological nature of the TI. The essential properties of TMs can be summarized as follows. First, electron spins in TM are locked into the momenta, forming spin chiral states.⁵ Such spin chiral states are also well known in the field of surface science in terms of Rashba effects in surface states [for example, Sb (111),⁶ Bi(111),⁷ and Au (111) (Ref. 8) surfaces states]. Second, backscattering is suppressed due to the spin chirality,⁹ meaning a relatively long quasiparticle lifetime. Third, metallic bands are protected against perturbations to the first order due to the topological nature. This point has yet to be experimentally observed.

Experimental verification of the novel properties of TM is not only important in the fundamental aspect but also necessary for the use of these materials for future applications. Due to the surface nature of the TM states, most of the experimental data thus far came from angle-resolved photoemission (ARPES) (Refs. 5 and 10–12) and to a less degree from scanning tunneling microscopy (STM).⁹ By using ARPES, it was shown that there exist odd number of bands crossing the Fermi level in this class of materials.^{10,11} Moreover, spin-resolved photoemission results show that the electron spins are indeed locked into the momentum and form spin chiral states.⁵ As for STM studies, a recent study shows the suppression of backscattering, consistent with spin chiral states.⁹

Studies mentioned above are about the existence and spin chiral states of TM, and the experimental verification of whether topological states are in general protected or not has not been discussed. Protected topological states should manifest themselves with long quasiparticle lifetime. In that regard, ARPES is an important tool because long quasiparticle lifetime should result in a sharp ARPES line shape. In spite

of the intensive recent efforts on TM electronic structure studies, photoemission line-shape issue has not been addressed. This point, however, is important because recent discussions on the use of TIs for spintronic applications require very low scattering rates to properly convey the spin information.

To address the quasiparticle lifetime issue, high-resolution ARPES data on various TIs have to be obtained. We begin the effort with studies on a parent TI Bi_2Se_3 . The goal of this work is to analyze the high-quality ARPES spectral function in terms of various scattering channels to see the mechanisms behind the quasiparticle scattering. In that analysis, the bulk states play an important role in quasiparticle dynamics in the TM band: electrons in TM are strongly scattered to the bulk electronic states by coupling to phonons, impurity or defect created potentials, and other electrons. In addition, we find the TM states protected against adsorbate-induced disorder potentials.

Single crystals were grown by a self-flux technique, following the previously reported recipe.¹³ ARPES measurements were performed at the beam line 7U of UVSOR-II.¹⁴ Various photon energies between 8 and 21 eV were used. The total energy resolution was set to be 10 meV at 8 eV, and the angular resolution was 0.1° . Samples were cleaved *in situ* and the chamber pressure was about 2×10^{-10} torr. The measurement temperature was kept at 15 K.

ARPES data from Bi_2Se_3 (111) are plotted in Fig. 1. Figure 1(a) shows data from freshly cleaved surface with an 8 eV photon. Relatively sharp and strongly dispersive electron pocket is observed. This is the TM band having a helical spin structure.⁵ As indicated by white arrow in the panel, the position of the TM band at the Γ point (Dirac point) is around 0.33 eV. One can also see a broad weak feature near E_F at the Γ point which originates from the bulk states.¹⁰

Figure 1(b) shows ARPES data taken with the same photon energy but 4 days after cleaving (aged surface). It is remarkable to note that the TM band feature remains intact and looks almost the same as that from a freshly cleaved surface. One difference is that the Dirac point is now located at around 0.43 eV, about 0.1 eV higher than the original

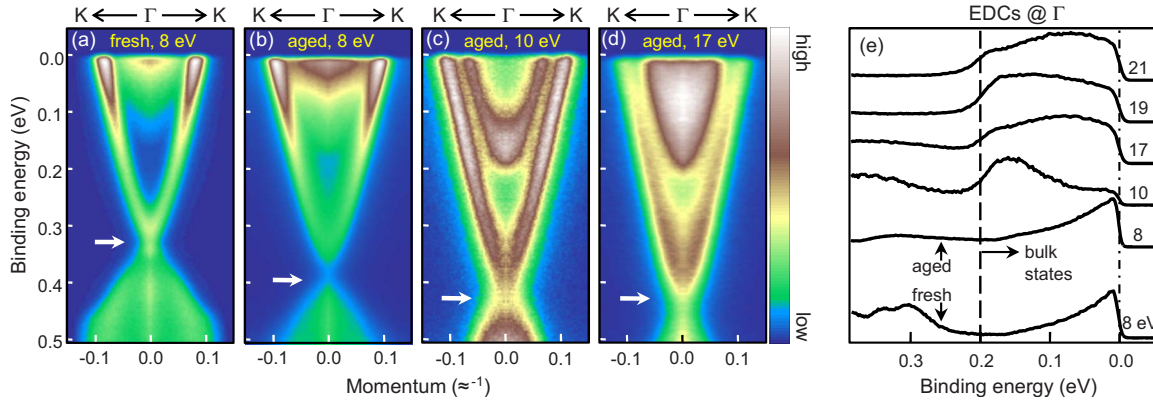


FIG. 1. (Color online) ARPES data taken with (a) 8, (b) 8, (c) 10, and (d) 17 eV photon energies along the Γ to K points. The data in (a) were taken on a freshly cleaved surfaces while data in (b)–(d) were taken on aged (4-day-old) surfaces. (e) EDCs at the Γ point taken with various photon energies

position. This shift of the Dirac point is attributed to charge transfer from physisorbed atoms or molecules. While a similar surface doping effect in Bi_2Te_3 has already been discussed,^{12,15,16} no such an effect has been reported in Bi_2Se_3 (111). The difference in the bulk states other than the uniform shift is due to the k_z -selection rule, which will be discussed below.

Figures 1(c) and 1(d) plot the data from aged surface taken with 10 and 17 eV. Compared to the 8 eV data, they have almost the same TM features while bulk bands look quite different. To see the photon energy dependence more clearly, we plot in Fig. 1(e) energy distribution curves (EDCs) at the Γ point from fresh and aged surfaces taken with various photon energies between 8 and 21 eV. The bulk state feature in the EDC taken with an 8 eV photon is relatively sharp. This feature becomes broader as photon energy increases. This can be explained by the decreasing photoelectron lifetime with increasing photon energy, resulting in the inclusion of a broader range of k_z .¹⁷ This, in combination with the fact that probed k_z varies with the photon energy, results in a clear observation of the bulk conduction-band bottom (BCBB) edge at about 0.2 eV in the EDCs taken with 17, 19, and 21 eV photons as indicated by the dashed line in Fig. 1(e), while the 8 eV EDC does not display a clear BCBB edge. In addition, 10 eV is the right photon energy to observe the BCBB as can be seen in Figs. 1(c) and 1(e). Considering the fact that there is 0.1 eV shift in the band position between fresh and aged surfaces, we can determine that the BCBB of the fresh surface is at about 0.1 eV.

We now wish to analyze the self-energy of the TM states. Before we proceed to analyzing the actual data, however, we need to touch upon various scattering channels. In estimating the self-energy, we note that scattering between TM electrons should be small due to the limited number of states (compared to the bulk states) and helical spin structure.⁹ Therefore, the dominant mechanism for TM electron (or hole) decay is through transition to bulk states. Figure 2(a) shows possible scattering channels between a hole in the TM states and a bulk electron. A hole in the TM state may decay into a bulk electronic state with the total energy and momentum conserved through electron-hole (e-h) pair creation, phonon emission or absorption, and impurity created potential as il-

lustrated in Fig. 2(a). A rough estimate of the imaginary part of the TM electron self-energy (or $1/\tau$) can be made by calculating the available phase-space volume. We assume that phonons between 0 and 0.03 eV equally contribute to scattering and also that the bulk density of states is proportional to $(E - E_{BB})^{1/2}$, where E_{BB} is the BCBB energy. A schematic of the result at 0 K is shown in Fig. 2(b). While $\text{Im} \Sigma$ in the Fermi-liquid theory¹⁸ increases proportional to ω^2 , $\text{Im} \Sigma$ due to e-h pair creation in Bi_2Se_3 increases at low binding energy side and saturates slightly after E_{BB} . On the other hand, phonons and impurities provide major scattering channels for the states with binding energies lower than E_{BB} . These channels however do not contribute to the states with binding energies higher than E_{BB} .

ARPES spectral function $A(k, \omega)$ is proportional to the imaginary part of the Green's function,

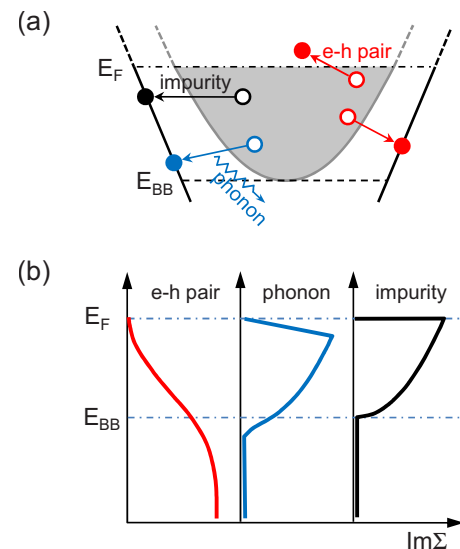


FIG. 2. (Color online) (a) Schematic of various scattering channels for a photohole in the TM band. Only the transition to the bulk states is considered. (b) Corresponding $\text{Im} \Sigma$ for various channels.

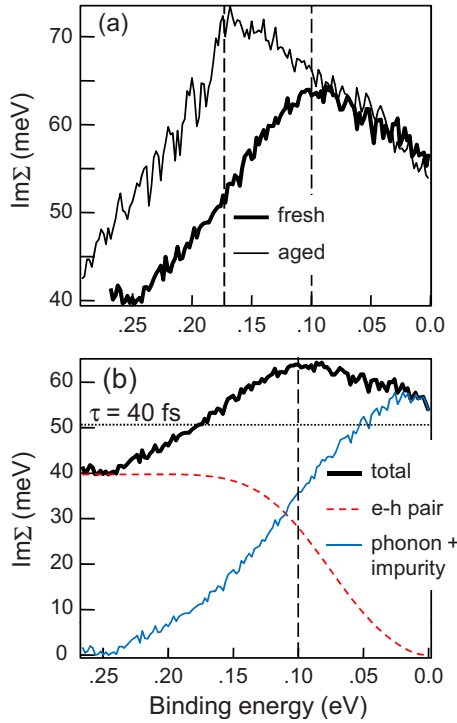


FIG. 3. (Color online) (a) TM band $\text{Im}\Sigma$ from fresh and aged surfaces of Bi₂Se₃ (111) taken with 8 eV photons. (b) Analysis of $\text{Im}\Sigma$ from fresh surface in terms of e-h pair creation, and phonon plus impurity channels.

$$\text{Im} G(k, \omega) = \frac{\text{Im} \Sigma(k, \omega)}{[\omega - \varepsilon_k - \text{Re} \Sigma(k, \omega)]^2 + \text{Im} \Sigma(k, \omega)^2}. \quad (1)$$

If the self-energy $\Sigma(k, \omega)$ is slowly varying with ω , EDCs are Lorentzians and the half width at half maximum (HWHM) of each EDC gives $\text{Im}\Sigma$. The extracted EDC peak width has been used to study the electron dynamics in Mo(111) surface states and cuprates.^{19–21} Fitting an EDC with a Lorentzian function however is often difficult in correlated materials due to, for example, the incoherent spectral weight. Instead, a commonly used method is to obtain $\text{Im}\Sigma$ by multiplying HWHMs of momentum distribution curves by the band velocity,^{22,23} which we also use in analyzing the data. Another note is that it helps to isolate the TM contribution of the spectral function. In the case of Bi₂Se₃ (111), bulk bands are located close to the TM band, making it difficult to extract the TM spectral weight from ARPES data precisely. However, we note that the bulk state spectral function taken with an 8 eV photon is much more suppressed than other data sets as shown in Figs. 1(a)–1(c). This is, as discussed above, k_z -selection rule that suppresses photoemission from the bulk states at this photon energy. This provides us an opportunity to extract the TM spectral function and do self-energy analysis more reliably. We thus analyze and present self-energy analysis on the 8 eV data.

Figure 3(a) plots $\text{Im}\Sigma$ from fresh and aged surfaces. $\text{Im}\Sigma$ initially increases but starts to decrease at around 0.10 and 0.17 eV binding energies for fresh and aged surfaces, respectively. In addition, the fresh surface data hint flattening of the

$\text{Im}\Sigma$ curve from 0.25 eV binding energy. These behaviors contrast with ordinary metallic cases where $\text{Im}\Sigma$ monotonically increases due to electron-electron scattering.^{18,19,21–23} The decrease in $\text{Im}\Sigma$ reminds us of the drop in $\text{Im}\Sigma$ from phonon and impurity channels at E_{BB} . Thus, we can attribute the $\text{Im}\Sigma$ behavior to the fact that TM electrons are dominantly scattered to bulk electronic states which exist only below E_{BB} .

We analyze the experimental $\text{Im}\Sigma$ of the 8 eV data from fresh surfaces based on the interpretation illustrated in Fig. 2. The results are plotted in Fig. 3(b). It is seen from the result that, while $\text{Im}\Sigma$ at the high-energy side mainly comes from the electron-electron interaction (electron-hole pair creation) channel, impurity and phonon channels dominate the low-energy dynamics which controls the transport properties. As contributions from both impurity and phonon channels are approximately proportional to the bulk density of states, it is difficult to distinguish them. However, we believe that the low-energy part of $\text{Im}\Sigma$ mainly comes from the defect scattering because characteristic phonon kink in $\text{Im}\Sigma$ illustrated in Fig. 2(b) was not observed. This is not unreasonable considering the fact that these materials tend to be nonstoichiometric. This will be the main obstacle to overcome if dissipationless spin currents on TIs were to be realized. The lifetime (τ) and the mean free path (l_m) of the electrons near E_F are roughly $\tau = 40$ fs and $l_m = \tau v_g = 0.02$ μm , respectively.

As discussed above, the decay of quasiparticles in TM states appears to mainly come from scattering to the bulk states, not to other TM states. Indeed, there are indications in other ARPES data that the intrinsic lifetime of quasiparticles in TM bands is fairly long. For example, Bi₂Te₃ shows a relatively sharp ARPES line shape.¹² Even though not an insulator, Sb(111) also has TM states with very sharp ARPES peaks.²⁴ For the low-energy electron dynamics, we showed that defect scattering plays the main role. It is therefore important to make stoichiometric Bi₂Se₃ not only to make insulating bulk states but also to reduce the scattering between TM and bulk states.

Finally, we discuss if the TM states are protected. We observe that the Fermi-surface volume of the TM band from aged surfaces increased by about 2.3% compared to that of the fresh surfaces, which means that at least 0.023 electrons per surface unit cell are transferred presumably from adsorbed atoms or molecules on the sample surface. Assuming that each atom or molecule donates one electron and that all the donated electrons are localized at the surface states, there are $6 \times 10^{13}/\text{cm}^2$ adsorbate atoms or molecules which translate into ≈ 13 \AA interadsorbate distance. Considering the fact that bulk states also receive electrons, the interadsorbate distance should be shorter. Since the adsorbates should induce disorder potential near the surface, additional scattering channels between surface electronic states should be induced in ordinary cases. Therefore, we expect an increase in $\text{Im}\Sigma$ near E_F . However, we do not observe such an increase in the data in Fig. 3(a). Note that the mean free path estimated from the lifetime in Fig. 3(b) is about 200 \AA , which is much longer than the estimated interadsorbate distance.

There can be different reasons why the scattering rate did not increase. Interesting case would be from the protected nature of the TM states. The estimated number of adsorbate

atoms or molecules on the surface may be enough to increase the scattering rate substantially. However, the scattering rate may have not increased because the TM states are topologically protected from weak disorder from the potential induced by adsorbates. If it is indeed the case, this would be an experimental evidence from ARPES for the protected nature of TM states, to our best knowledge. However, one has to be careful not to make a foregone conclusion because the induced potential can vary depending on the adsorbates. For example, relatively small amount of H atoms on graphene destroys the metallic states while K atoms have almost no effect except electron doping.²⁵ In order to resolve this issue,

we propose a controlled ARPES experiments in combination with first-principles electronic structure calculations.

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