Coexistence of nontrivial two-dimensional surface state and trivial surface layer in Kondo insulator SmB$_6$

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A long-standing puzzle in well-known Kondo insulator (KI) SmB$_6$ is the ill-defined “in-gap” state. Very recently, diverse evidences show that such “in-gap” state could be ascribed to novel topological surface state (TSS). Surface nature instead of bulk nature became a central issue in this field. However, how to identify the genuine surface nature in this material is quite challenging. Here, we used AMR method combined with surface modification to further investigate the surface nature and revealed a coexistence of nontrivial two-dimensional surface state and trivial surface layer in SmB$_6$. Sample and experimental details are in supplementary materials (S1–S3).

Evidence for 2D surface state In Fig. 1(c), a very striking evolution of AMR pattern from four-fold to two-fold symmetry was observed below 5K in SmB$_6$ flat sample. Considering the bulk cubic symmetry as shown in Fig. 1(a), the four-fold symmetry in AMR pattern above 5K was compatible with bulk symmetry and could be attributed to bulk contribution. But the two-fold symmetry in AMR pattern below 5K was completely incompatible with bulk symmetry. Two possibilities could contribute to such two-fold symmetry. One is the bulk symmetry broken and the other is 2D surface state. The ability to manifest 2D nature of surface state is a remarkable advantage in AMR measurement (See supplementary materials S5). In order to distinguish above two possibilities, we further measured the AMR pattern on flat samples with (100) and (110) surface respectively and the current was applied along the same (100) direction (See supplementary materials S5). If the two-fold symmetry originated from bulk symmetry broken, the AMR pattern should be independent on surface orientation. Otherwise, 2D surface state should be dominated.

As shown in Fig. 1(d) and (e), a $\pi/4$ phase difference was observed between AMR patterns with different surface orientations at 2K and it disappeared at 5K with the appearance of four-fold symmetry. The $\pi/4$ is exactly angle between (100) and (110) planes. It indicated that the observed two-fold symmetry in AMR pattern relies on surface orientation. Therefore, the possibility of bulk symmetry broken could be excluded and our AMR results proved a 2D surface state contribution in conductivity below 5K. This is one main finding in this study. In previous transport measurement, sample-size effect measurement in SmB$_6$ has already shown evidence on the existence of surface state. However, it is incapable

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to identify the 2D nature of the surface state. In this case, trivial surface layer (e.g., trivial accumulation layer in pure Te[25]) with three-dimensional (3D) nature could also explain sample-size effect. Here, our results gave unambiguous transport evidence on the existence of 2D surface state in SmB$_6$. The two-fold AMR pattern will be used as a benchmark of 2D surface state in the following study.

Robustness of 2D surface state In idea TIs, the transport properties are dominated by thermal activation of insulating bulk at high temperature. As decreasing temperature, the thermal activation behavior will be eventually short-circuited by surface conduction. This is the primary TKI scenario to explain the “in-gap” state
FIG. 4: Temperature-dependent resistivities in logR vs 1/T plot. The slop in this plot stands for activation energy of insulating phase. The blue and red dash lines stand for HTI phase and LTI phase, respectively. The saturation in resistivity was not shown in this plot for visibility. (b)-(e) Temperature-dependent resistivities of Sm$_{1-x}$La$_x$B$_6$ single crystals with smooth and rough surfaces. The current was applied along [001] direction. The red open circles and blue open circles stand for the result with smooth surface and rough surface respectively. (f)-(h) The AMR patterns at 2K with smooth surface of Sm$_{1-x}$La$_x$B$_6$ single crystals. (i)-(k) The AMR patterns at 2K with rough surface corresponding to Sm$_{1-x}$La$_x$B$_6$ single crystals in (f)-(h). The experimental configuration is the same as that in Fig.2(a). The solid lines in (f)-(j) are simulation results with $\Delta R_{\text{AMR}}(2\theta) / R(0)$ or $\Delta R_{\text{AMR}}(00) / R(0)$. The AMR pattern for x = 0.2 sample shown in (h) is divided into two parallel connected components with two-fold symmetry and four-fold symmetry respectively and the two-fold component is the contribution from surface state. The solid line shows above two-components simulation result.

As we know, surface absorption is an essential factor for trivial surface state. We could change the surface absorption by high-temperature annealing under vacuum to examine the surface nature. Our annealing process was conducted under different temperatures as shown in Fig. 2(a) and (b). Below 1000°C, the sample’s surface was very stable and there was no any surface damage to complicate the situation. However, in order to reduce surface absorption efficiently, we needed to raise annealing temperature. Above 1000°C, the sample’s surface started to damage and we had to use boron protection to avoid surface damage (See supplementary materials S4). In both cases, no any change in both temperature-dependent resistivity and two-fold AMR pattern was observed as shown in Fig. 2. It suggested that surface absorption is not crucial for 2D surface state, supporting a nontrivial surface state. In fact, high-temperature annealing process could change the surface properties in some cases as shown in Fig. 3(b) and Fig. S3(b). However, they are ascribed to other factors rather than surface absorption effect (See supplementary materials S4).

Besides surface absorption, we also modified surface roughness. As shown in Fig.3(a), the low-temperature resistivity was found two orders of magnitude smaller after surface polishing to raise the surface roughness. Meanwhile, the AMR pattern at 2K was changed from two-fold symmetry to four-fold symmetry as shown in Fig. 3(c) and (d). This is a very surprising result. If the remarkable reduction in resistivity was ascribed to the improvement of 2D surface state, the two-fold symmetry in AMR pattern should remain. The appearance of four-fold symmetry indicated that the resistivity change comes from bulk contribution. How does surface polishing change the bulk resistivity? The only solution is to consider a trivial surface layer different from inner bulk. Such trivial surface layer kept 3D bulk symmetry considering the four-fold symmetry in AMR pattern. In this case, we could not distinguish trivial surface layer from inner bulk contribution only by AMR measurement. Here, surface-roughness dependent resistivity played key role to figure out the existence of trivial surface layer. The appearance of two-fold symmetry or four-fold symmetry in AMR pattern is strongly dependent on the competition between contributions from 2D surface state and trivial surface layer. After surface polishing, such trivial surface layer was dominated in resistivity and 2D surface state contribution was hidden. In order to reduce the contribution from trivial surface layer on rough surface, we annealed the rough-surface sample at 1000°C for 12h. After annealing, the total resistivity was enhanced and the two-fold symmetry recovered in AMR pattern as shown in Fig. 3(b) and (e). Although additional contribution from trivial surface layer jumped in,
above results still suggest that the 2D surface state could also survive on rough surface. Very recently, STM experiment on cleaved SmB$_6$ surface showed robust evidence on “in-gap” state with different surface terminations[21]. Their results suggested that the “in-gap” state is quite robust against modification of surface terminations which is consistent with our present results.

**Doping effect on surface state** We have also examined the above surface properties in La-doped SmB$_6$ samples. As shown in Fig. 4(b)-(e), the whole insulating behavior in temperature-dependent resistivity was monotonously suppressed with increasing La$^{3+}$ ions doping level. However, there is a distinct insulating behavior appears below 10K with La doping as shown in Fig. 4(a). Such distinct insulating behavior could also be observed with increasing pressure[22]. In fact, such different low-temperature insulating behavior was already observed in parent SmB$_6$ sample as shown in Fig. 4(a). We could define these two different insulating behaviors as high-temperature-insulating(HTI) phase and low-temperature-insulating(LTI) phase respectively. The HTI phase was suppressed continuously with increasing La$^{3+}$ ions doping level but the LTI phase only disappeared abruptly over 20% doping level. Furthermore, we have measured the AMR pattern and rough-surface resistivity on the La-doped samples. As shown in Fig. 4(b)-(k), the similar two-fold symmetry pattern and surface-roughness dependent resistivity were also observed in La-doped samples up to 20% doping level. Moreover, we found that the surface-roughness effect only works on LTI phase in all samples. It suggests that the LTI phase originates from above-mentioned trivial surface layer. As the LTI phase disappeared abruptly over 20% doping level, the two-fold symmetry pattern also became unmeasurable. This correlation suggests that the previous revealed 2D surface state should develop from the trivial surface layer instead of inner bulk phase. This is another main finding in our study.

In previous surface analysis, the concentration ratio of Sm$^{3+}$ ions to Sm$^{2+}$ ions in the vicinity of the SmB$_6$ (001) surface was found to be much higher than that in bulk[22]. This suggests a quite different electrical properties of surface region from those of the bulk in SmB$_6$. Recently, an anomalous capacitance effect was found in SmB$_6$ and it was ascribed to a structure of inner bulk enwrapped with distinct surface layer[5]. We also observed the similar effect in our samples.(See supplementary materials S10). All these results support that a different surface layer from 2D surface state should be considered in SmB$_6$ which is responsible for the LTI phase below 10K. Such surface layer was not recognized in previous surface-dominated transport measurement[7, 8]. How to understand such surface layer needs further investigation. Although the origin of such surface layer remains elusive, it’s strongly dependent on surface situation, suggesting its trivial nature. As we know, the accumulation layer in semiconductor could play the role as such trivial surface layer. But its possibility needs further theoretical study.

In present study, we gave unambiguous evidence on a robust 2D surface state and identified a trivial surface layer in SmB$_6$. This indicated that there are two conducting channels near the surface, consisting of robust 2D surface state channel and another trivial surface layer channel. A two-channel model near the surface was needed to account for the low-temperature transport in SmB$_6$ and it would play a key role to understand the genuine surface nature in SmB$_6$. Our results would promote the final identification of SmB$_6$ as the first correlated topological insulator.

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### Author contributions

F.C. and C.S. performed sample growth and all experiments with assistance from T.W., A.F.W. and X.G.L.. F.C. and T.W. analyzed the data. F.C., T.W. and X.H.C. wrote the paper. T.W. and X.H.C. conceived and coordinated the project. X.H.C are responsible for the infrastructure and project direction. All authors discussed the results and commented on the manuscript.

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