Hall-coefficient diagnostics of the surface state in pressurized SmB_6

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(Received 8 October 2019; accepted 28 February 2020; published 19 March 2020)

In this study, we report results of the high-pressure Hall coefficient (R_H) measurements in the putative topological Kondo insulator SmB_6 up to 37 GPa. Below 10 GPa, our data reveal that $R_H(T)$ exhibits a prominent peak upon cooling below 20 K. Remarkably, the temperature at which surface conduction dominates coincides with the temperature of the peak in $R_H(T)$. The temperature dependent resistance and Hall coefficient can be well fitted by a two-channel model with contributions from the metallic surface and the thermally activated bulk states. When the bulk of SmB_6 becomes metallic and magnetic at ~10 GPa, both the $R_H(T)$ peak and the resistance plateau disappear simultaneously. Our results indicate that the $R_H(T)$ peak is a fingerprint to diagnose the presence of a metallic surface state in SmB_6. The high-pressure magnetic state of SmB_6 is robust to 180 GPa, and no evidence of superconductivity is observed in the metallic phase.

DOI: 10.1103/PhysRevB.101.125116

I. INTRODUCTION

Samarium hexaboride (SmB_6) is a unique compound in strongly correlated electron systems, because it is not only a prototypical Kondo insulator but also has an exotic metallic surface state. At high temperatures, SmB_6 behaves as a correlated bad metal but undergoes a metal-to-insulator crossover upon cooling due to the hybridization between the localized f electrons and the conduction electrons [1–10]. Below ~4 K, the electrical resistivity of SmB_6 displays a plateau, which has been a puzzling issue for decades [1,2]. This has been revealed recently to be attributed to an exotic metallic surface state that coexists with a bulk insulating state [4,6,7,10–15]. Thus, SmB_6 could be an example of a class of topological insulators with strong electronic correlations [16–18]. Considerable experimental efforts have been made to confirm the topological nature of the surface states in SmB_6, and the correlation between the resistance plateau and a metallic surface state was established [4,6,7,10–15]. Unlike the nontrivially topological insulators whose metallic surface state is solely protected by its insulating bulk state [19], the exotic surface state of the strongly correlated electron Kondo insulator SmB_6 stems from not only the Kondo physics, i.e., the hybridization between the localized f electrons and the conduction electrons [14], but also the other strongly correlated interactions including the degrees of freedom of lattice and valence state [18,20–23]. Therefore, the surface state of SmB_6 is protected by the insulating bulk in a more complicated way than that of topological insulators. It is expected that SmB_6 should have peculiar response to electric field and magnetic field at low temperature. It is known that it exhibits a resistance plateau and oscillation at low temperature [12,24], however the connection of the $R_H(T)$ peak with the exotic metallic surface state and all the key factors of this strongly correlated system is still lacking, although it has been extensively investigated under pressure before the discovery of its metallic surface state [25–33].

Pressure is a clean and effective way of tuning interactions in solids with multiple degrees of freedom without introducing chemical complexity [23,34–43]. For such a strongly correlated electron system, the change of Hall coefficient with pressure and temperature can reflect the evolutions on the interactions among the lattice instability [44], the valence change of Sm ions [23], and the hybridization between $4f$ electron and conducting electrons [14]. In this work, we performed the combined Hall and resistance measurements on the high quality SmB_6 single crystals under pressure, with an attempt to identify how the metallic surface and insulating bulk states evolve with $R_H(T)$. We propose that the $R_H(T)$ peak can be taken as a fingerprint to diagnose the presence of an exotic surface state in SmB_6.

II. RESULTS AND DISCUSSIONS

In Fig. 1(a) we show the temperature dependence of electrical resistance measured in a SmB_6 sample upon cooling under pressures up to 36.8 GPa. The resistance of the sample

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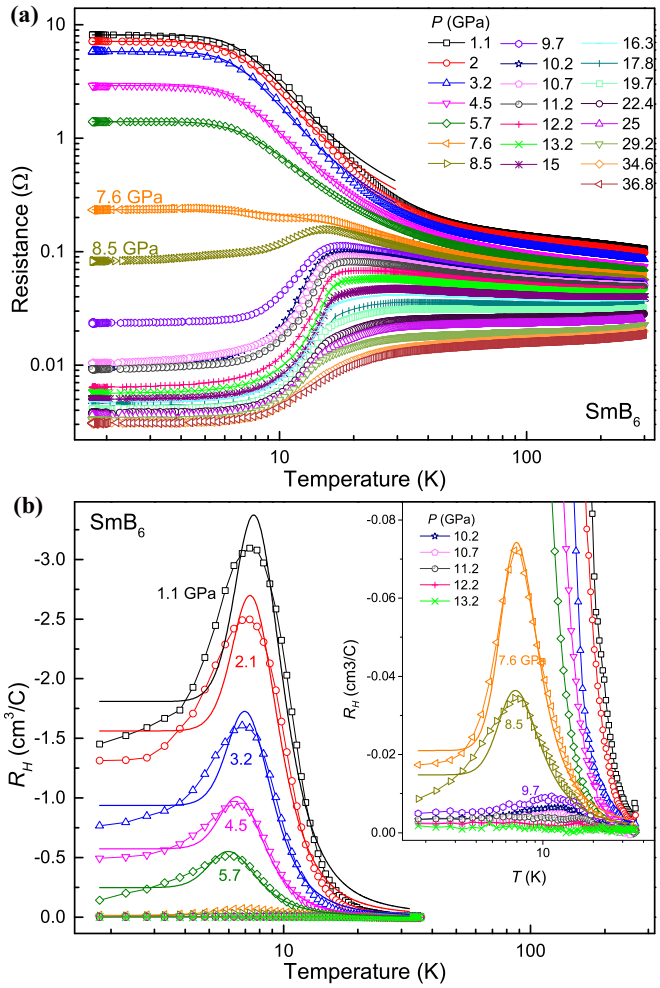

FIG. 1. Transport properties in pressurized SmB_6. (a) Temperature dependence of resistance from 1.7 to 300 K at different pressures in log-log scale. The solid line is the fitting by a two-channel model. (b) Hall coefficient (R_H) as a function of temperature measured at different pressures. The inset is the large view of $R_H(T)$ obtained at higher pressure. Solid lines are the fitting result.

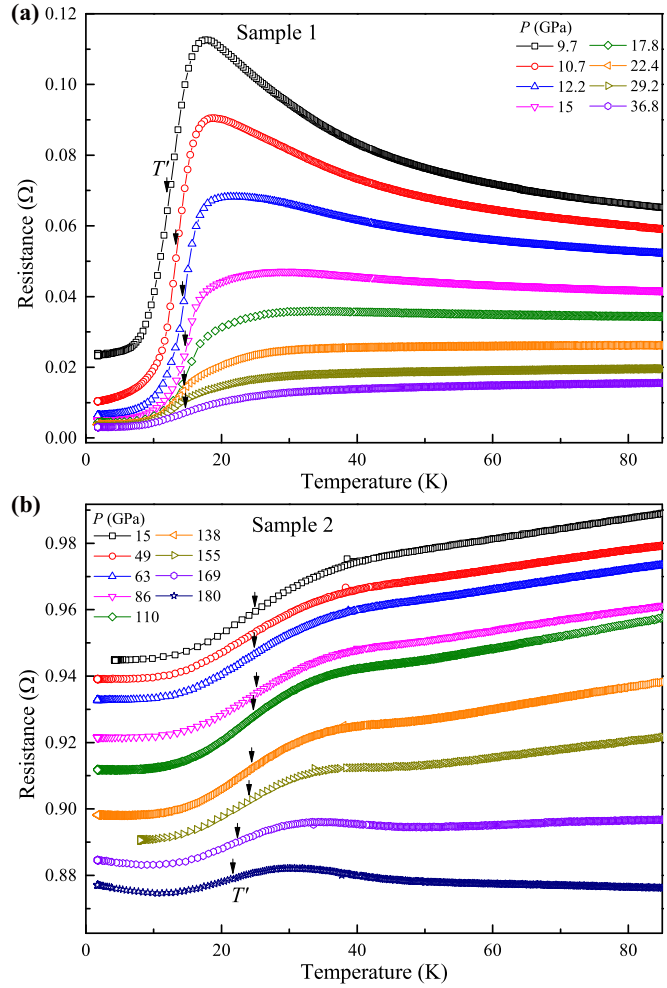

FIG. 2. High pressure behavior of metallic SmB_6. (a) Resistance versus temperature in the pressure range of 9.7 ∼ 36.8 GPa for the sample 1. (b) Temperature dependence of resistance under pressure up to 180 GPa for ample 2. T' represents maximum of dR/dT curves indicating the resistance drop in the metallic phase. Samples 1 and 2 were cut from the same batch, which were loaded into the different two high pressure cells for high pressure (∼37 GPa) and very high pressure measurements (∼180 GPa).

at 1.1 GPa displays a continuous increase upon cooling and then exhibits a plateau below 5 K, identical to the behavior at ambient pressure [1,9,14,25,45–48]. Further, the resistance plateau is clearly visible at pressures below 7.6 GPa, but becomes indistinguishable at 8.5 GPa. Further compression to pressures above 9.7 GPa leads to the disappearance of the resistance plateau and to a substantial drop of the magnitude of the electrical resistance at low temperature, a signature of an insulator-to-metal transition. It is worth noting that the critical pressure of the insulator-to-metal transition found in this study is ∼10 GPa (see the experimental details in the Supplemental Material [49]), in excellent agreement with the critical pressure applied to the sample using argon as pressure medium [28].

High-pressure Hall coefficient measurements were performed upon warming after the resistance measurements. As shown in Fig. 1(b), in the temperature range of 1.8–30 K, $R_H(T)$ is negative at all pressures, indicating that the electron carriers are dominant. Remarkably, the plot of R_H versus temperature displays a domelike behavior in the temperature range below 10.7 GPa [Fig. 1(b) and inset]. The domelike $R_H(T)$ observed in the SmB_6 sample is attributed to the combined contribution from surface and the bulk states. For temperatures ranging from 300 to 10 K, the temperature dependence of resistance exhibits an insulating behavior, implying that the bulk state is dominant [Fig. 1(a)]. In this case, the charge carriers decrease upon cooling and corresponding R_H increases. At the lower temperature where the resistance plateau presents, R_H shows a decease upon cooling due to the dominance of the metallic surface state [21]. The temperature dependence of both resistance and Hall coefficient obtained under pressure can be fitted by a two-channel model consisting of a thermally activated bulk in parallel with a temperature-independent surface state [21,22]. The resistance can be described as

$$R = (1/R_S + 1/R_B)^{-1}, \tag{1}$$

where R_S and R_B represent the resistance from surface and bulk channels, respectively. Here, $R_B = R_{B_0} e^{E_g/k_B T}$, in which

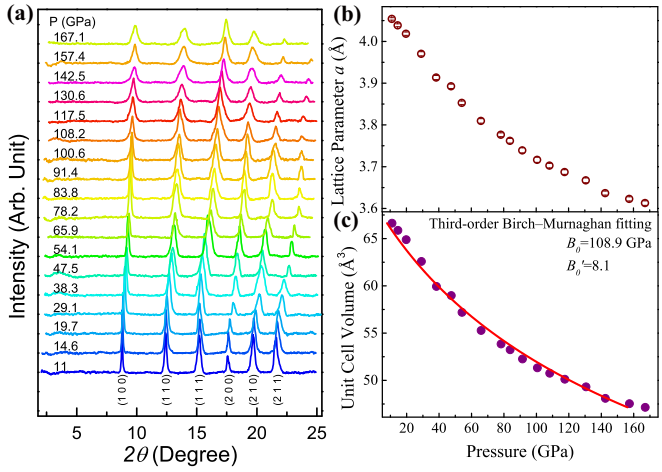

FIG. 3. High pressure structure information for SmB_6. (a) X-ray diffraction patterns of SmB_6 collected at different pressures. (b) and (c) Pressure dependences of lattice parameter and volume. Error bars in (b) are in the brown circles. The volume versus pressure data are fit to the third-order Birch-Murnaghan equation of state [9], as displayed by the solid line in (c). The B_0 is the isothermal bulk modulus at ambient pressure, B_0' is the pressure derivative of B_0 evaluated at zero pressure.

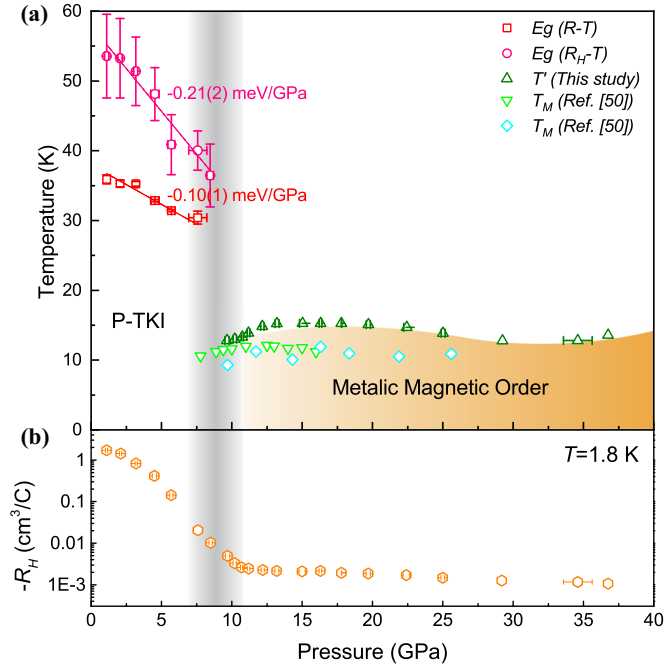

FIG. 4. Pressure-temperature phase diagram and Hall coefficient at 1.8 K. (a) Plots of pressure versus characteristic temperatures. P-TKI stands for putative topological Kondo insulator. Pink circles and red squares are the activated gap Eg obtained from temperature-dependent R-T curves and Hall coefficient respectively and is converted to temperature by Eqs. (1) and (2). The olive triangles are the characteristic temperatures T' obtained from our R-T curves. Green inverted triangles and cyan rhombuses are the magnetic ordering temperature taken from Ref. [45] (b) Plot of Hall coefficient (R_H) versus pressure obtained at 1.8 K. The gray interval indicates the pressure region of insulator-to-metal transition.

E_g is the activated energy gap, k_B is Boltzmann constant, R_{B_0} is the bulk resistance in the high-temperature limit, and T is temperature.

The Hall coefficient can be expressed as

$$R_H = \left(R_{H-S} d\rho_B{}^2 + R_{H-B}(d\rho_S)^2\right) / (d\rho_S + \rho_B)^2, \quad (2)$$

where R_{H-S} and R_{H-B} are the Hall coefficient of the surface and bulk states, respectively. ρ_S and ρ_B are resistivity of the surface and bulk states, and d is the thickness of the sample (the estimated d used in the fit was $\sim 10\,\mu$m).

The solid lines in Figs. 1(a) and 1(b) are the fit results. At low pressures, our results are fairly described by the two-channel model. The surface state dominates the electrical conductivity when the resistance plateau appears, whereas the bulk insulating state dominates above the temperature of the resistance plateau formation. At ~10.7 GPa, $R_H(T)$ is featureless, indicating that the conductance of the bulk state is comparable with that of the surface state due to the pressure-induced metallization. Although our experimental results deviate from the fit at the temperatures below the $R_H(T)$ peak [Fig. 1(b)], this should be an intrinsic feature of the sample, because SmB_6 is a temperature sensitive compound whose lattice and valence show complicated changes upon cooling. Its valence displays a decrease upon cooling and then shows an increase at lower temperature where its resistance saturates [23,44]. We propose that the deviation of the fit to our experimental results at temperatures below the peak of $R_H(T)$ should be attributed to the combined effects of pressure and temperature. These results demonstrate that the domelike $R_H(T)$ can be taken as a fingerprint to distinguish the coexistence of the metallic surface in SmB_6.

Magnetic order has been found previously in pressurized SmB_6 by nuclear forward scattering of synchrotron radiation measurements [50]. The magnetic ordering temperature (T_M) has been confirmed to lie in the 10–12 K range at ~10 GPa [50]. We find that the midpoint temperatures (T') of the resistance drop in the metallic SmB_6 are close to its corresponding T_M measured by nuclear magnetic resonance measurements. If T' is taken as a characteristic temperature of T_M [Fig. 2(a)], it is surprising to find that T_M is robust under pressures as high as 180 GPa [Fig. 2(b)]. Because $R(T)$ curves measured in the pressure range of 10–180 GPa exhibit similar behavior and no structure phase transition is observed under pressures to 167 GPa (Fig. 3), we propose that the magnetically ordered state remains in the pressurized metallic phase throughout this pressure range.

We summarize our results in Fig. 4. SmB_6 hosts a metallic surface state below 10 GPa, which is characterized by the resistance plateau and the peak in $R_H(T)$. This peak decreases with increasing pressure and eventually disappears at ~10 GPa as the bulk state of the sample becomes metallic state [Fig. 4(a)]. Significantly, the thermal instability of $R_H(T)$ indicates that the temperature-induced lattice change plays a vital role in tuning the valence state of Sm ions and the population of conducting electrons, as well as their interaction [44], which are the central issues for such a strongly correlated electron system with Kondo physics. Moreover, we noted that the criterion of the $R_H(T)$ peak can be applied to topological insulators such as Bi_2Te_2Se and $Bi_{1.08}Sn_{0.02}Sb_{0.9}Te_2S$

[51,52], whose R_H appears decrease dramatically in the temperature range of the surface state dominance. The behaviors of the $R_H(T)$ peak found in the compounds of SmB_6 or topological insulators with the coexistence of metallic surface state and the bulk insulating state lead us to suggest that the $R_H(T)$ peak may be a common feature for the materials hosted the exotic surface state, no matter that the surface state stems from nontrivially topological physics or electron correlated physics. This interesting issue deserves further experimental and theoretical investigations.

We also find that the midpoint of the resistance drop, $T'(P)$, coincides with the magnetic transition temperature T_M detected by nuclear scattering forward measurements [50], and it is present to 180 GPa. These results suggest that a robust magnetically ordered state is stabilized, which prevents the emergence of superconductivity. The corresponding pressure dependence of the R_H obtained at 1.8 K is shown in Fig. 4(b). Below ∼10 GPa, R_H decreases with increasing pressure and then stays almost constant in the metallic magnetic state up to ∼37 GPa. It is known that SmB_6 is a mixed valence compound at ambient pressure with valence $\nu_{Sm} \sim 2.6$. The application of pressure drives the valence change with the tendency from nonmagnetic Sm^{2+} to magnetic Sm^{2+} ions. Previous high-pressure absorption measurements [20,23,33,53] indicated that its mean valence is very close to 3+ at $P > \sim 10$ GPa. The pressure-induced valence change of $Sm^{2+}(4f^6) \rightarrow Sm^{3+}(4f^5 + 5d)$, together with its stable cubic lattice structure, should be responsible for the robustness of long-ranged magnetic order [20,23,33,53].

III. CONCLUSION

In conclusion, the prominent feature of the temperature dependent Hall coefficient for the coexistence of surface and bulk states in a putative topological insulator SmB_6 has been revealed. The intimate correlation between the low-temperature R_H and the exotic surface state suggests that $R_H(T)$ is one of the most useful diagnostic methods to identify the existence of the exotic surface state in SmB_6 and in some of topological insulators such as Bi_2Te_2Se and $Bi_{1.08}Sn_{0.02}Sb_{0.9}Te_2S$. Furthermore, we find the extraordinary robustness of the crystal structure and metallic state in compressed SmB_6 up to 180 GPa and no superconductivity is observed in the pressure range investigated.

ACKNOWLEDGMENTS

The authors are indebted to P. Coleman, J. Thompson, L. Greene for stimulating discussions. The work in China was supported by the National Key Research and Development Program of China (Grants No. 2016YFA0300300, No. 2017YFA0302900, and No. 2017YFA0303103), the NSF of China (Grants No. 91321207, No. U1532267, No. 11604376, No. 11374361, and No. 11522435), the Strategic Priority Research Program (B) of the Chinese Academy of Sciences (Grant No. XDB07020300). Work at Los Alamos was performed under the auspices of the U.S. Department of Energy, Division of Materials Sciences and Engineering. Work at Rice University was supported by the National Science Foundation Grant No. DMR-1920740 and the Robert A. Welch Foundation Grant No. C-1411.

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