Science Bulletin xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Science Bulletin

journal homepage: www.elsevier.com/locate/scib

Article

# Quantum phase transition and destruction of Kondo effect in pressurized $SmB_6$

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ARTICLE INFO

*Article history:*
Received 9 October 2017
Received in revised form 16 October 2017
Accepted 16 October 2017
Available online xxxx

*Keywords:*
Kondo insulator
Surface state
$SmB_6$
High pressure

ABSTRACT

$SmB_6$ has been a well-known Kondo insulator for decades, but recently attracts extensive new attention as a candidate topological system. Studying $SmB_6$ under pressure provides an opportunity to acquire the much-needed understanding about the effect of electron correlations on both the metallic surface state and bulk insulating state. Here we do so by studying the evolution of two transport gaps (low temperature gap $E_l$ and high temperature gap $E_h$) associated with the Kondo effect by measuring the electrical resistivity under high pressure and low temperature (0.3 K) conditions. We associate the gaps with the bulk Kondo hybridization, and from their evolution with pressure we demonstrate an insulator-to-metal transition at ~4 GPa. At the transition pressure, a large change in the Hall number and a divergence tendency of the electron-electron scattering coefficient provide evidence for a destruction of the Kondo entanglement in the ground state. Our results raise the new prospect for studying topological electronic states in quantum critical materials settings.



## 1. Introduction

Samarium hexaboride ($SmB_6$) is a paradigm Kondo insulator (KI) with a simple cubic structure, comprising a $B_6$ octahedral framework and Sm ions [1]. It has in common with other KIs that an insulating gap opens upon cooling due to the hybridization between the localized *f*-electrons and conduction electrons [2–4], but distinguishes from the other KIs by the presence of a low temperature resistance plateau which has been a puzzle for decades [5,6]. Recent theoretical studies suggest that the resistance plateau may be associated with the existence of an exotic metallic surface state, and that $SmB_6$ could be a new class of topological insulator with strong electron correlations, namely a topological Kondo insulator (TKI) [7,8]. A metallic surface state indeed has been confirmed by a variety of measurements [9–17], but whether it is protected by a non-trivial topology remains to be established [18]. Surprisingly, various energy gaps ranging from 5 to 15 meV have been observed from ambient-pressure resistivity, angle resolved photoemission spectroscopy (ARPES), and scanning tunneling spectroscopy (STS) experiments [6,19–23], but it is still unclear how these gaps relate with the bulk Kondo hybridization between 4*f* electrons and conduction electrons. In addition, the newly discovered quantum oscillation phenomena, which suggest an unusual Fermi surface in $SmB_6$ at low temperature, yield an additional puzzle [24,25]. In order to make progress on these issues, it is crucially important to understand the nature of the multiple electronic gaps.

One of the important means to understand Kondo hybridization gap is the tuning of the Kondo insulators through a quantum phase transition [4], in particular through the phase diagram proposed for such systems at zero temperature [26,27]. For studying

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https://doi.org/10.1016/j.scib.2017.10.008
2095-9273/

Please cite this article in press as: Zhou Y et al. Quantum phase transition and destruction of Kondo effect in pressurized $SmB_6$. Sci Bull (2017), https://doi.org/10.1016/j.scib.2017.10.008

quantum phase transition, it is known that high pressure is a "clean" way of continuously tuning the crystal and electronic structures without introducing additional chemical complexity [28,29]. Before the proposal of topological conducting surface state [5,6,21,30–33], high-pressure phenomena of $SmB_6$ had been extensively investigated down to 1.5 K, however a systematic understanding on the instability of the metallic surface state and Kondo hybridization bulk state is lacking. In this study we use our advanced high-pressure facilities which allow us to perform resistance measurements down to 0.3 K, and apply the *complementary* measurements – adopting the same high pressure cell, the same pressure transmitting medium and the same pressure gauge on the same batch of single crystal – to carry out such investigations on $SmB_6$.

## 2. Methods

High quality single crystals of $SmB_6$ were grown by the Al flux method, as described in Ref. [11]. Pressure was generated by a diamond anvil cell with two opposing anvils sitting on the Be-Cu supporting plates. Diamond anvils with 300 μm flat and non-magnetic rhenium gaskets with 100 μm diameter hole were employed for different runs of the high-pressure studies. The standard four-probe method was applied on the (0 0 1) facet of single crystal $SmB_6$ for all high pressure transport measurements. To keep the sample in a quasi-hydrostatic pressure environment, NaCl powder was employed as a pressure transmitting medium for the high-pressure resistance and Hall coefficient measurements.

High pressure X-ray diffraction (XRD) and X-ray absorption spectroscopy (XAS) experiments were performed at room temperature at beamline 4W2 in the Beijing Synchrotron Radiation Facility and at the beamline 14W1 in the Shanghai Synchrotron Radiation Facility, respectively. Diamonds with low birefringence were selected for the experiments. A monochromatic X-ray beam with a wavelength of 0.6199 Å was adopted for all XRD measurements. To maintain the sample in a hydrostatic pressure environment, silicon oil was used as pressure transmitting medium in the high-pressure XRD and XAS measurements. Pressure was determined by the ruby fluorescence method [34].

## 3. Experimental results

We first performed *in situ* high pressure resistance measurements down to low temperatures (0.3 K) on single crystal $SmB_6$. As demonstrated in Fig. 1a and b, the resistivity for sample A and B shows the same behavior at pressures below 4 GPa, *i.e.* the resistivity increases continuously upon decreasing temperature and then displays a plateau below ~4 K, which is consistent with the results reported previously [5,6,21]. Upon further increasing pressure above 4 GPa, the resistivity shows a remarkable drop at low temperature, which manifests a pressure-induced insulator-metal transition (MIT). It is noteworthy that the different pressure transmitting medium can give rise to different pressure environments for the sample. This pressure medium effect on sample is particularly serious in $SmB_6$, as reported by Derr et al. in Ref [35]. The MIT of the $SmB_6$ in a hydrostatic pressure environment (using an argon gas as pressure medium) is pushed up to 10 GPa, while in a quasi-hydrostatic pressure environment (using steatite [35–37] or NaCl in this study as pressure medium) the MIT occurs at ~4 GPa.

Interestingly, the onset temperature ($T^*$) of the resistance plateau exhibits a downward trend with increasing pressure, and disappears at ~4 GPa (Fig. 1c). Above 4 GPa, while the resistivity vs. temperature becomes metallic-like at low temperatures ($T < \sim 20$ K), it still exhibits insulating-like behavior at higher temperatures ($T > \sim 20$ K). This is so for pressures below ~10 GPa; only at even higher pressures (>10 GPa) does the system changes to metallic-like behavior over the entire measured temperature range (Fig. S1, Supplementary Information). In the pressure range investigated, no crystal structural phase transition is observed (Fig. S2, Supplementary Information), in agreement with the results reported [32,33,38]. This indicates that the pressure-induced resistance evolution stems from purely electronic physics.

To determine the nature of the ground state at 4 GPa and above, we fitted the resistivity-temperature (*R-T*) data from 0.3 to 4 K with the power law, $\rho(T) = \rho_0 + AT^n$ (where $\rho_0$ is the residual resistivity, $A$ the electron-electron scattering coefficient and $n$ the exponent), we find that below 0.7 K the *R-T* curve at 4 GPa clearly follows a Fermi liquid (FL) behavior with $n = 2$ (Fig. 2a). A linear *R-T* behavior at 4 GPa presents only in the temperature range of 1.5–4 K (Fig. 2a), in agreement with previous measurements at higher temperatures (>1.5 K) [21]. Within the temperature range of 0.7–1.5 K, $n$ lies between 1 and 2. Our results reveal that the real ground state of $SmB_6$ at 4 GPa is a FL state, instead of a non-FL state. Our investigation of high-pressure resistance measurements at 4.5 GPa down to 0.03 K further confirms the FL ground state (Fig. S3, Supplementary Information). Fits to the *R-T* data measured down to 0.3 K for the pressures ranging from 5 to 19.6 GPa demonstrates that the $T_{FL}$, the temperature into the Fermi liquid regime, appears to increase linearly with pressure (Fig. 2b, c, Figs. S3, S4, Supplementary Information). Interestingly, the $A$ coefficient in $\rho(T) = \rho_0 + AT^2$ as a function of pressure (as shown in Fig. 2d) shows that the $A$ coefficient (3.3 μΩ cm/K$^2$) at 4 GPa is about 720 times larger than its value (0.046 μΩ cm/K$^2$) at 19.6 GPa. Following the Kadowaki-Woods relation [39], this corresponds to an increase of the effective carrier mass ($m^*$) by a factor of ~27 as the pressure is reduced towards the transition pressure 4 GPa. We also fit the *R-T* data measured down to 0.03 K, and found that the value of the $A$ coefficient is on the data line, suggesting that the $A$ coefficient extracted from the *R-T* data measured down to 0.3 K should be very close to the one obtained from the *R-T* curve measured down to 0.03 K. By analogy with the behavior of heavy fermion metals [4,28], the striking divergence tendency in the effective mass with pressure on approach of 4 GPa signifies the destruction of Kondo entanglement (Kondo singlet) at the insulator-metal quantum phase transition.

## 4. Discussions and summary

The pressure-temperature phase diagram established from our transport data is shown in Fig. 3a. Three low temperature regions appear in this phase diagram. The region below 4 GPa represents the putative topological Kondo insulating (P-TKI) state characterized by the resistance plateau. In the intermediate-pressure region between 4 and 10 GPa, with decreasing temperature the sample shows a non-metallic behavior down to the $T_P$ (the temperature of the resistance peak indicated by the arrow in Fig. S1, where the resistivity of the sample undergoes a change from non-metallic behavior to a metallic behavior) and then enters into a FL ground state below the $T_{FL}$. In the region above 10 GPa, the sample displays metallic behavior over the entire measured temperature range, and also shows a FL behavior at low temperature, where a long-range magnetic order is observed [30,31]. To further address the nature of the quantum phase transition at ~4 GPa, we performed Hall measurements at a fixed temperature of 1.7 K at different pressures up to ~20 GPa (Fig. 3b and Figs. S5–S7). The inverse Hall coefficient undergoes a large change across ~4 GPa. Comparing this low-temperature isothermal behavior with its counterparts at higher temperatures, we find that the crossover feature of the inverse Hall coefficient sharpens as the temperature is lowered (Fig. 3b, inset). Even though the Hall data is expected to contain contributions from both surface and bulk carriers, our results are compatible with a rapid change of the carrier number

Please cite this article in press as: Zhou Y et al. Quantum phase transition and destruction of Kondo effect in pressurized $SmB_6$. Sci Bull (2017), https://doi.org/10.1016/j.scib.2017.10.008