Thermoelectric signals of state transition in polycrystalline SmB$_6$

Z J Yue$^{1,3}$, Q J Chen$^2$ and X L Wang$^1$

$^1$ Institute for Superconducting and Electronic Materials, Faculty of Engineering, University of Wollongong, North Wollongong, NSW 2500, Australia
$^2$ School of Physics and Electronics, Hunan University, Changsha 410082, People’s Republic of China

E-mail: zy709@uowmail.edu.au

Received 14 April 2016
Accepted for publication 20 June 2016
Published 1 July 2016

Abstract

Topological Kondo insulator SmB$_6$ has attracted quite a lot of attention from the condensed matter physics community. A number of unique electronic properties, including low-temperature resistivity anomaly, 1D electronic transport and 2D Fermi surfaces have been observed in SmB$_6$. Here, we report on thermoelectric transport properties of polycrystalline SmB$_6$ over a broad temperature from 300 to 2 K. An anomalous transition in the temperature-dependent Seebeck coefficient $S$ from $S(T) \propto T^{-1}$ to $S(T) \propto T$ was observed around 12 K. Such a transition demonstrates a transition of conductivity from 3D metallic bulk states to 2D metallic surface states with insulating bulk states. Our results suggest that the thermoelectric measurements could be used for the characterization of state transition in topological insulators.

Keywords: topological Kondo insulators, thermoelectricity, surface state

(Some figures may appear in colour only in the online journal)
of bars with typical dimensions of about $2 \times 2 \times 10 \text{mm}^3$. The Seebeck coefficient, electrical resistivity, and thermal conductivity were measured using a Quantum Design 14 T physical properties measurement system (PPMS). The four-probe mode was employed in thermoelectric transport measurements between 2 K and 300 K. The applied AC bias current was 2 mA.

Figure 1(a) shows the temperature dependent resistivity of polycrystalline SmB$_6$ in the range from 2 K to 300 K. Above 40 K, the bulk Kondo gap is closed due to thermal energy excitations, and the SmB$_6$ behaves like a metal. As the temperature decreases, the resistivity increases by several orders of magnitude due to the opening of a Kondo gap. Below 3.5 K, the resistivity approaches a constant value of $\sim 0.017 \Omega \cdot \text{m}$, which originates from the surface states of SmB$_6$ [6]. In figure 1(b), the Hall resistance is plotted as a function of magnetic fields at different temperatures. The negative sign in the Hall effect demonstrates that the carriers are electrons at low temperatures. The calculated carrier density in polycrystalline SmB$_6$ is about $3.25 \times 10^{18} \text{cm}^{-3}$ and $1.5 \text{cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$, respectively. Compared to SmB$_6$ single crystals, the mobility is quite low [10]. This is attributable to very high density of boundaries and defects in the polycrystalline SmB$_6$. Figures 1(c) and (d) present the magnetization as a function of temperature and magnetic fields, respectively. The polycrystalline SmB$_6$ demonstrates paramagnetic behaviour at high temperature. The low temperature Curie tail maybe related to the recent observed surface ferromagnetic domain walls [7].

Figure 2(a) shows the total thermal conductivity, $\kappa$, decreases with the temperature decreasing. A subtle transition of the $\kappa$ happens at $\sim 40$ K which corresponds to an opening of the Kondo gap. As the Kondo gap (hybridization gap) opens, the contribution to the $\kappa$ from electrons decreases. The thermal conductivity is the sum of two contributions of electrons and phonons, which can be expressed as $\kappa = \kappa_e + \kappa_p$. At low temperatures, the reduced dimensions from 3D bulk states to 2D surface states reduces the electron contribution. At high temperature, large $\kappa$ stems from high density of bulk electrons with strong correlated interactions.

Figure 2(b) shows the Seebeck coefficient, $S$, as a function of the temperature. A dramatically low-temperature peak of $S$ was seen at $\sim 12$ K. The low temperature value of the $S$ peak is $-324 \ \mu \text{VK}^{-1}$. Below 3.5 K, $S$ is linearly dependent on temperature. We find that the $S$ as a function of the temperature in SmB$_6$ represents a 3D bulk value at higher temperature with the relation $S(T) \propto T^{-1}$. With the temperature decreasing, the relation changes to $S(T) \propto T$ below a critical temperature. In this temperature range, the dominant carriers are electrons from 2D surface states.

Above 12 K, the Seebeck coefficient $S$ arises from the electronic contribution in the 3D SmB$_6$ bulk. Based on the Boltzmann equation and the Mott relation, the Seebeck coefficient is described as [11],

$$S = -\frac{1}{eT} \int L(\varepsilon)(\varepsilon - u)\left(-\frac{\partial f}{\partial \varepsilon}\right)\mathrm{d}\varepsilon.$$  

Here, $L(\varepsilon) = \mu(\varepsilon)v(\varepsilon)^2\tau(\varepsilon)$, where $\mu(\varepsilon)$ is density of states, $v(\varepsilon)$ is the electron velocity, $\tau(\varepsilon)$ is the electron relaxation time, and $u$ is the chemical potential.

The formula yields the well known Seebeck coefficient for semiconductors,

$$S(T) \approx -\frac{k_B E_{\text{g},\nu}}{e} \frac{\mu}{k_B T}.$$  

Here $E_{\text{g},\nu}$ is the gap edge position of the conduction or valence band, and $k_B$ is Boltzmann’s constant. Assuming that $\frac{\partial \mu}{\partial T}$ is negligible, $S$ is proportional to $-\frac{1}{k_B}$ at high temperature.

Hence, $S(T) \propto -\frac{1}{T}$

$S$ is sensitive to the characteristics of the electronic structure. According to ARPES, the band structures of SmB$_6$ display a Kondo gap of about 15 meV at the Fermi level and surface states below 15 K [12]. The 2D metallic surface states also emerge below 15 K. Below $T = 12$ K, the Seebeck coefficient $S$ arises from the electronic contribution in the metallic surface states. Takahashi et al have theoretically studied thermoelectric transport in 3D topological insulators by the Boltzmann equation. In a 2D topological insulator, the transport matrix for describing thermoelectric transport is [13],

$$\left( j/\nu \right) = \left( \begin{array}{c} L_0 \\ L_1 \\ L_2 \end{array} \right) \left( \begin{array}{c} d\mu \\ dx \\ T \end{array} \right).$$

Here, $j$ is the electric current induced by electric field and $w$ is the thermal current induced by thermal gradient. $q$ is charge, and $u$ is the chemical potential.

From the matrix elements, the thermoelectric parameters can be expressed as,

$$\sigma = e^2 L_0$$

$$S = -\frac{1}{eT} \frac{L_1}{L_0}$$

$$k_e = \frac{1}{T} \frac{L_0 L_2 - L_1^2}{L_0}$$

$$ZT = \frac{L_1^2}{L_0 L_2 - L_1^2 + k_\ell T L_0}.$$  

Here, the $k_e$ and $k_\ell$ is the thermal conductivity from electrons and phonons, respectively, $\sigma$ is the electrical conductivity, and $ZT$ is the thermoelectric figure of merit. In this theory, $k_\ell$ is a constant. Considering a thin slab of 3D topological insulator with a small thickness $d$, so that the bulk is treated as 2D, from the Boltzmann transport equation, the surface state transport can be expressed as,

$$L_\nu = c(k_B T) \int_0^\infty \frac{(x - u)e^{-x + \frac{u}{\tau}}}{(e^{-x + \frac{u}{\tau}} + 1)^2} \mathrm{d}x.$$
\[ E = \hbar c L v_n V \]  

Here \( v_n \) is the Dirac velocity, \( V \) is the average impurity potential, and \( n_i \) is the density of impurities. Hence,

\[ \hat{u} = \frac{u}{k_B T} \quad \text{and} \quad \Delta = \frac{\Delta}{k_B T}. \]  

Figure 1. (a) Resistivity of polycrystalline SmB\(_6\) as a function of temperature. (b) Hall resistance of polycrystalline SmB\(_6\) as a function of external magnetic fields. (c) Magnetization of polycrystalline SmB\(_6\) as a function of temperature. (d) Magnetization of polycrystalline SmB\(_6\) as a function external magnetic fields.

Figure 2. (a) Thermal conductivity of polycrystalline SmB\(_6\) as functions of temperature. (b) Seebeck coefficient of polycrystalline SmB\(_6\) as functions of temperature.

\[ c = \frac{1}{\pi L_y n_i V^2(0)} \]  

\[ \bar{u} = \frac{u}{k_B T} \quad \text{and} \quad \Delta = \frac{\Delta}{k_B T}. \]  

Here \( v \) is the Dirac velocity, \( V \) is the average impurity potential, and \( n_i \) is the density of impurities. Hence,

\[ S = -\frac{1}{eT L_0} \]  

\[ S(T) = -k_B \int_{-\Delta}^{0} \frac{(x - \bar{u})e^{-x+\bar{u}}}{e^{-\Delta} (e^{-x+\bar{u}} + 1)^2} dx \]  

\[ S(T) \propto \frac{1}{T}. \]
Chao et al have calculated the thermoelectric transport in the surface states of topological insulators in the presence of randomly distributed impurities [14]. They deduced the generalized Mott formula for $S$ when the temperature is close to zero, $T \to 0$,
\[
S_{ij} = -\frac{\pi^2 k_B^2 T}{3e} \sum_m \Omega^{-1}_{mn} \left[ \partial \sigma / \partial u \right]_{mn}, \tag{15}
\]
Here, $\sigma$ is the electrical conductivity.

When the impurity potential $V = 0$ and the chemical potential $u \approx 0$, $S \propto -\frac{k_B T}{u}$.

For a clean surface state with $V \approx 0$, the density of impurities $n \approx 0$,
\[
S_{xx} \approx -\frac{\pi^2 k_B T}{3e} \left( \frac{1}{u/k_B T} - \frac{\partial \Sigma(u)}{\partial \Sigma(u)/k_B T} \right). \tag{16}
\]
Combined with equation (4), so
\[
S \approx -\frac{\pi^2 k_B T}{3e}. \tag{17}
\]
Hence, overall, as $T \to 0$, $S(T) \propto -T$. The final Seebeck becomes
\[
S = -\frac{1}{eT} \frac{L_1}{L_0}. \tag{18}
\]
As shown in figure 2(b), polycrystalline SmB$_6$ exhibits a low-temperature $S$ anomaly. Below 12 K, $|S|$ decreases linearly with temperature and resembles values for metal-like transport. High-resolution ARPES identified that in-gap low-lying states form electron-like Fermi surface pockets within a 4 meV window of the Fermi level [5]. They disappear above 15 K, in correspondence with the complete disappearance of the 2D conductivity channels. Hence, the $S$ anomaly below 12 K in polycrystalline SmB$_6$ is attributable to the 2D conductivity channels.

The efficiency of thermoelectric materials for cooling or power generation is described by the thermoelectric figure of merit $ZT$
\[
ZT = \frac{S^2 \sigma}{k} \tag{19}
\]
where $\sigma$, $S$, and $k$ are the electrical conductivity, the Seebeck coefficient, and the thermal conductivity respectively. The resulting $ZT$ in polycrystalline SmB$_6$ is shown in figure 3. At high temperature, the 3D metallic bulk transport is dominant. The $ZT$ increases with decreasing temperature and reaches a maximum value of up to 0.0073 at 37 K. Below 37 K, the $ZT$ monotonously decreases down to near zero. Although someone reported that topological surface states could provide a route to optimize the $ZT$, it is evident that SmB$_6$ is not promising to be used in thermoelectric devices [15, 16].

In summary, we report the thermoelectric transport in the polycrystalline topological Kondo insulator SmB$_6$. We find that the robust surface states survive in polycrystalline material with a large amount of non-magnetic impurities and disorder. The temperature dependent Seebeck coefficient demonstrates an anomalous transition from $S(T) \propto -1/T$ to $S(T) \propto -T$ at 12 K, where surface states start dominating the conductivity. This anomalous transition demonstrates a transformation from 3D metallic bulk states to completely 2D metallic surface states. Our results provide a new way to detect surface states in topological insulators.

Acknowledgments

This work is partially supported by the Australian Research Council under a Discovery project (ARC Discovery, DP130102956).

References

[1] Hasan M Z and Kane C L 2010 Rev. Mod. Phys. 82 3045
[2] Qi X-L and Zhang S-C 2011 Rev. Mod. Phys. 83 1057
[3] Li G et al 2014 Science 346 1208
[4] Yue Z, Wang X, Wang D, Wang J, Culcer D and Dou S 2015 J. Phys. Soc. Japan 84 044717
[5] Neupane M et al 2013 Nat. Commun. 4 3786
[6] Kim D J, Xia J and Fisk Z 2014 Nat. Mater. 13 466
[7] Nakajima Y, Syers P, Wang X, Wang R and Paglione J 2015 Nat. Phys. 12 213
[8] Ando Y 2013 J. Phys. Soc. Japan 82 102001
[9] Yue Z J, Zhu C B, Dou S X and Wang X L 2012 Phys. Rev. B 86 195120
[10] Yue Z J, Wang X L and Dou S X 2012 Appl. Phys. Lett. 101 152107
[11] Kim D J, Thomas S, Grant T, Botimer J, Fisk Z and Xia J 2013 Sci. Rep. 3 3150
[12] Maher G D 1997 Solid State Physics vol 51, ed H Ehrenreich and S Frans (New York: Academic) p 81
[13] Cutler M and Mott N F 1969 Phys. Rev. 181 1336
[14] Takahashi R and Murakami S 2012 Semicond. Sci. Technol. 27 124005
[15] Chao S-P and Aji V 2011 Phys. Rev. B 84 155430
[16] Takahashi R and Murakami S 2010 Phys. Rev. B 81 161302
[17] Ghaemi P, Mong R S K and Moore J E 2010 Phys. Rev. Lett. 105 166603