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Magnetotransport in nanocrystalline SmB_6 thin films

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SmB_6 has been predicted to be a prototype of topological Kondo insulator (TKI) but its direct experimental evidence as a TKI is still lacking to date. Here we report on our search for the signature of a topological surface state and investigation of the effect of disorder on transport properties in nanocrystalline SmB_6 thin films through longitudinal magnetoresistance and Hall coefficient measurements. The magnetoresistance (MR) at 2 K is positive and linear (LPMR) at low field and become negative and quadratic at higher field. While the negative part is understood from the reduction of the hybridization gap due to Zeeman splitting, the positive dependence is similar to what is observed in other topological insulators (TI). We conclude that the LPMR is a characteristic of TI and is related to the linear dispersion near the Dirac cone. The Hall resistance shows a sign change around 50K. It peaks and becomes nonlinear around 10 K then decreases below 10 K. This indicates that carriers with opposite signs emerge below 50 K. These properties indicate that the surface states are robust and probably topological in our nanocrystalline films. [http://dx.doi.org/10.1063/1.4927398]

With spin-momentum locked surface states which are robust against impurity scatterings, topological insulators (TIs) are promising in potential spintronic and quantum computing applications.[1,2] The bulk of most well-known topological insulators such as Bi_2Se_3 or Bi_2Te_3 are often not insulating due to lattice vacancies or self-doping.[3] Significant efforts have been made to improve the insulation of such materials,[4] but the prediction[5,6] of SmB_6 as a possible topological insulator with true insulating bulk make it a very attractive material. To date, there have been reports on transport,[7–15] neutron[16,17] and surface sensitive measurements[18–20] on SmB_6 crystals which have indicated the presence of a metallic surface state, but the convincing proof that the surface state is topological is still lacking. It is also well known that SmB_6 is a Kondo insulator which suggests there might be an interesting interplay between correlated physics and topological properties. The fundamental steps of utilizing these possible topological and correlated properties in actual devices require preparation of SmB_6 in thin film forms. In our previous work,[21] we showed how we can grow SmB_6 nanocrystalline films by co-sputtering SmB_6 and B targets and demonstrated their structural, electrical and spectroscopic properties. In this paper, we search for the signature of the topological surface state from electrical transport of the SmB_6 films in a magnetic field applied in both longitudinal and transverse directions. By comparing our data with the data from single crystals, we discuss how structural disorder, which is typical in our films and possible future devices, might alter those transport properties. Although neither weak-antilocalization nor any quantum oscillations are observed, the observation of linearly positive magnetoresistance (LPMR) and an anomalous Hall signal at low temperatures is consistent with the existence of a robust topological surface state at low temperatures.

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A well-known Kondo insulator,[13] upon decreasing the temperature, the resistivity of SmB_6 increases like an insulator but saturates at temperatures below 5K. Recent transport measurements show that the resistance of this saturation is thickness-independent for both longitudinal[10] and transverse[7] directions. This observation supports the idea that the mysterious low temperature saturation is due to conducting surface states. Moreover, weak antilocalization and linear magnetoresistance[8] have been observed in some single crystal SmB_6, and these observations are used to support the presence of spin momentum locked surface states. It has been shown that doping SmB_6 with magnetic impurities diminishes this saturation, while non-magnetic impurities do not.[12] Such findings suggest that its conduction is from a surface state which is protected by time-reversal symmetry, and is robust against non-magnetic scatterings. However, there are also reports that indicate the resistivity saturation might be due to dangling surface bonds[22] or affected by carbon impurities.[23] One ARPES study[19] claims the observation of spin texture structure of surface states while others disagree.[24] Reports on crystals also show a mysterious variety of behaviors in low-field magnetoresistance.[8,25,26] To test whether there are indeed topological surface states and how they behave under high disorder, we performed temperature dependent magnetotransport measurements on our SmB_6 nanocrystalline thin films.

SmB_6 films are grown by co-sputtering of using SmB_6 and B targets in ultrahigh vacuum at 800C. The films are annealed in situ at 800C for 3 hours and Wavelength Dispersive Spectroscopy (WDS) is used to make sure the samples we measure are stoichiometric SmB_6. They are 100 nm thick on Si/SiO_2 substrates. X-ray diffraction and transmission electron microscopy indicate the films are nanocrystalline with a typical grain size ranging from 2 to 5 nm. Details of fabrication and characterization are given in Ref. 21. Magnetotransport measurements are carried out in a Quantum Design PPMS system with the field up to 9 T and temperature down to 2 K. Magnetoresistance and the Hall resistance are symmetrized as $R'_{xx}(H) = [R_{xx}(H) + R_{xx}(-H)]/2$ and antisymmetrized as $R'_{xy}(H) = [R_{xy}(H) - R_{xy}(-H)]/2$, respectively to avoid mixing of the longitudinal and transverse signals. We have measured more than five samples. Since the data are similar for the five samples, we only display one set of data here.

Typical magnetoresistance at 300 K, 50 K and 2 K, normalized to their zero field value, is shown in Fig. 1. At room temperature, there is very little MR. At 50 K, the MR develops a negative parabolic dependence. This has been observed previously and is attributed to the reduction of the hybridization gap from Zeeman splitting.[27] When the sample is further cooled down to 2 K, an additional linearly positive MR is found for $B < 4$ T. Our overall MR behavior is similar to the one reported in one crystal study,[8] although our overall MR change is two orders of magnitude smaller.

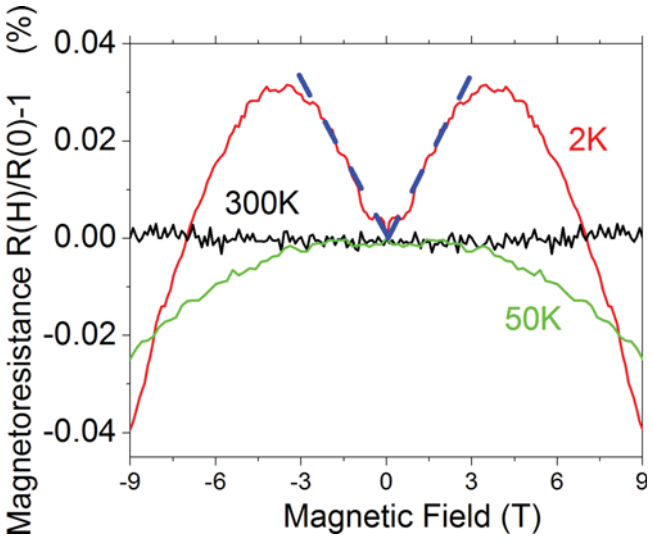

FIG. 1. Magnetoresistance change in SmB_6 films defined by $[R(H)/R(0)-1)]\times 100\%$ as a function of magnetic field at 300 K (in black), 50 K (in green) and 2 K (in red). The blue dashed lines are linear guide to the eye.

This is probably because our films are nanocrystalline, so the overall resistance is dominated by field-independent grain-boundary scatterings. This LPMR does not show any angular field dependence as seen in single crystals because the nanograins in our films have random orientations. The fact that our data is qualitatively similar to the single crystal data indicates that our temperature dependence arises primarily from the intra-granular component of the films.

Usually non-magnetic metals have small, quadratic positive magnetoresistance, and it quickly saturates at some small field.[28] Linearly positive magnetoresistance (LPMR) is unusual and has been observed in several systems such as multilayer graphene,[29] semimetal Bi,[30] doped semiconductor InSb[31] and other topological insulators.[32] It is usually a much larger effect which makes it more interesting for possible applications. It also saturates at a much larger field or does not saturate at all. There is currently no consensus theory for its origin. Theorists have proposed two mechanisms to explain this unusual effect. First is a classical one proposed by Littlewood and Parish[33] who suggest that inhomogeneities create tails in both the conduction and valence bands in small-gap semiconductors and cause them to overlap. Distorted current paths misalign with the driving voltage and mix in the off-diagonal components of the magnetoresistivity tensor. As a result, the magnetotransport can be then dominated by the magnitude of the fluctuations in the mobility near the band edge. This has been demonstrated in intentionally doped InSb[31] and AgSe.[34] Another mechanism is a quantum one proposed by Abrikosov,[35,36] and it only applies to rather extreme conditions, i.e., under a very large magnetic field, for a small effective mass, and also for a small carrier density. We do not believe that the LPMR we observed satisfies these conditions because it is observed at relatively high temperatures and in small magnetic fields. The LPMR we observe might be in the classical scenario since the Kondo band gap in SmB_6 is only ~20 meV and the nanocrystalline nature of our films makes them behave effectively as a disordered metal.

It is worthwhile to note that most materials with linear band dispersion (for example, near a Dirac cone) display a LPMR. This includes topological insulators such as Bi_2Te_3[32,37] and Bi_2Se_3[38] and new semi-metallic materials such as $LaAgBi_2$,[39] $SrMnBi_2$[40] and Cd_3As_2.[41] Therefore, it seems that this is an intrinsic and universal property of 3-D topological materials. This is then an indirect evidence that SmB_6 is also a topological material. More theoretical work is needed to confirm this empirical observation.

The Hall resistance as a function of external magnetic field at different temperatures is shown in Fig. 2. At 50 K and higher, R_{XY} is linear with a negative slope. Below 40 K, R_{XY} becomes positive and non-linear, and it peaks at around 10 K. Below 10 K, R_{XY} becomes smaller and more linear as the temperature is lowered to 2 K. To quantify the linearity of the $R_{XY}(H)$, we introduce a parameter $\alpha \equiv R_{XY}(4\,T)/4R_{XY}(1T)$ and plot it as function of temperature in Fig. 3. Above 40 K,

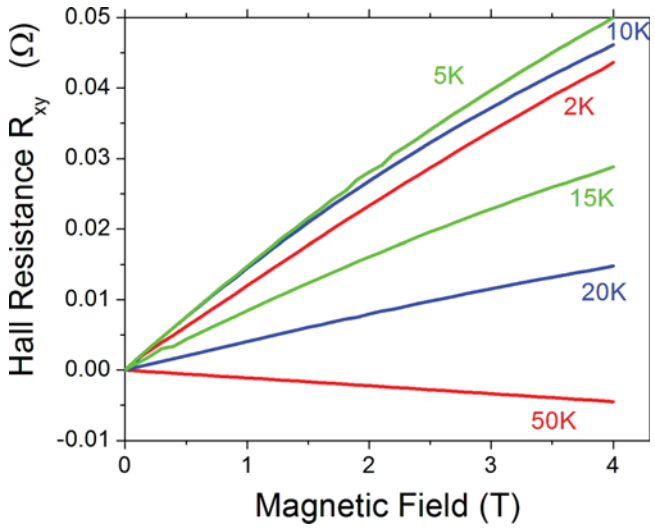

FIG. 2. Hall resistance R_{xy} of SmB_6 films as a function of magnetic field at selected temperatures. The field is perpendicular to the film surface, and the data are anti-symmetrized.

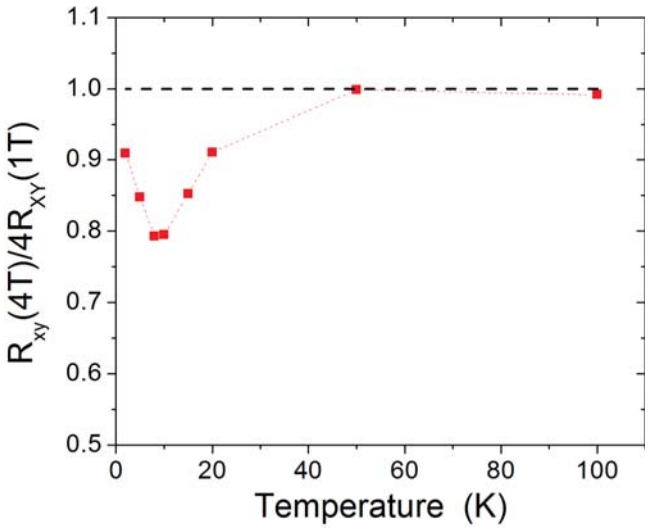

FIG. 3. Hall resistance linearity parameter defined as $R_{XY}(4T)/[4\times R_{XY}(1T)]$ as a function of the temperature. The dashed black line shows a perfectly linear Hall resistance.

α is almost 1.00 exhibiting the linear behavior. Then its value decreases to 0.80 at 10 K followed by an increase to above 0.90 at 2 K. The non-linearity in Hall resistance usually signifies the presence of two different types of carriers with different mobilities. This fact, together with the sign change, indicates that the low temperature carriers in our films have opposite signs and different origins from high temperature carriers.

To gain more insights on the Hall data, the Hall coefficients R_H (slope in small H limit) as function of temperature for two films are displayed in Fig. 4. Upon cooling down, the Hall coefficients of both samples show a sign change near 40 K. Both peak at around 10 K, then decrease at lower temperatures. Qualitatively speaking, both the sign change and the small peak are also observed in single crystal samples.[7,13] They are related to the opening of the hybridization gap at 40 K and two carrier competitions at intermediate temperatures (~10 K). One surprise is that the sign of our Hall data is opposite of that in crystals. (We checked our measurement configuration carefully to make sure we are measuring the correct sign.) This sign discrepancy might be due to the semiconducting-like properties of SmB_6. The modest increase of R_H (in absolute value) from

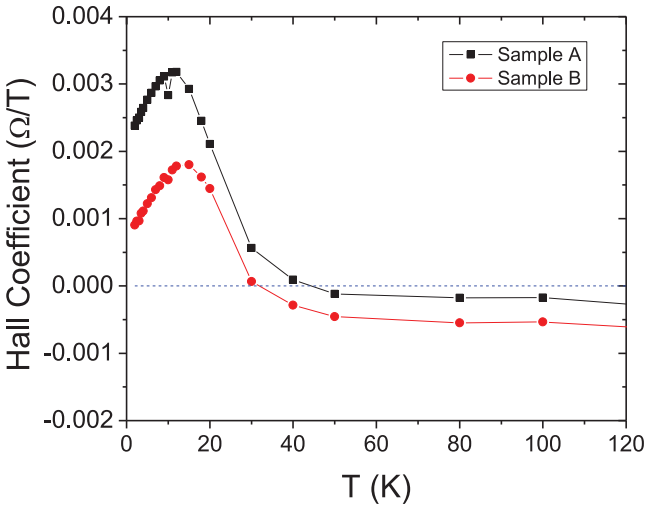

FIG. 4. Hall coefficient $R_H = R_{XY}/B$ as a function of temperature for two samples with different thicknesses and patterned Hall bar geometries. The dashed line indicates where the Hall coefficient changes sign.

300 K to 10 K that we observe is in direct contrast to that of crystals,[7] where R_H increases by orders of magnitude. This is similar to the smaller longitudinal resistance R_{xx} increase that we found previously,[21] which is ascribed to a much larger surface-to-bulk ratio for our films (by roughly three orders of magnitude). Also due to the much larger surface-to-bulk ratio, the surface state dominates the bulk states faster than in crystals when cooling down. R_H peaks at 10 K compared to the peak observed in 5 K in single crystals.[7,13] The temperature dependence of R_H clearly demonstrates the existence of surface carriers at low temperatures.

However, the existence of the surface conducting layer does not mean the surface states are topological. It is important to point out that we have not observed weak antilocalization or quantum oscillations, which are indicative of the surface states and are observed in most well-known TIs.[42–44] However, we believe this can be reconciled by carefully checking the conditions under which these quantum phenomena occur. It has been shown that even the best single crystals of SmB_6 do not show any quantum oscillations from transport measurements at 2 K.[27] Since the bulk is a Kondo insulator, no oscillations should be expected from the bulk. For the surface states, relatively high carrier mobilities are required to observe oscillations. This is given by a simple condition that $\mu B > 1$ where μ is the mobility of the carriers. One recent study indicates that the surface carrier mobility in SmB_6 crystals is only 133 $cm^2/V/s$ at 2 K.[10] This leads to μB having a maximum value of about 0.1 (this value for our films would be even smaller due to its nanocrystalline nature), too small to observe quantum oscillations.

Indeed, only one torque magnetometry study[9] reports an observation of quantum oscillations in SmB_6 crystals. It was enabled by the sensitivity enhancement due to the H^2 term in the torque formula. They had accordingly identified a Berry phase contribution from the oscillations, similar to ones observed in other 2D Dirac electronic systems such as graphene.[45] A theoretical paper[46] explained why a small effective electron mass is observed and predicted that the surface states are robust against decreasing thickness of the sample, which is consistent with the observation of surface states in our nanocrystalline films.

As for weak antilocalization, there is an ongoing debate over the low field intrinsic magnetoresistance behavior at lowest temperatures. Some attribute the observation of dips at low field and mK temperatures in some samples to weak antilocalization (WAL).[8] It has also been pointed out that the observed dips are sweeping rate dependent and that the observed "WAL" are from surface magnetism.[25] There are also those who observed hysteric behavior and suggest this as quantum transport due to surface magnetic domains.[26] Even if the observed dips are indeed from WAL, it has been found only in a small fraction of single crystal samples and only at mK temperatures. We have also cooled three of our films down to 20 mK, but neither WAL nor any hysteric behavior was observed (data not shown). Our conclusion is that WAL, if it exists, is not a robust phenomenon for SmB_6, at least compared to other topological insulators such as Bi_2Se_3, where WAL is observed by many groups at temperatures as high as 100 K. The reason behind this difference is still unknown.

There are some reports that indicate that the surface states in SmB_6 originate from dangling boron bonds[22] or from carbon contaminations[23]. It is well-known that the electronic potential of a semiconductor surface is usually different from that of the bulk and an internal electric field can develop. Then the band bending effect might lead to a formation of a two-dimensional electron gas (2DEG). This has been found to be the case for Bi_2Se_3,[43,44] where a 2DEG and topological states coexist on the surface. However, if the chemically formed 2DEG or trivial surface states such as dangling boron bonds exist and contribute to the surface states, they should show signatures such as field-dependent magnetoresistance or nonlinear Hall data at room temperature. Instead, those features only appear below 50 K in our samples, where the hybridization gap fully opens. This indicates that the observed surface state is strongly associated with the opening of the hybridization gap and likely to be topological. To definitely distinguish the topological surface state from the trivial surface state, more fabrication effort to grow high single crystalline thin films and more detailed surface-sensitive measurements are needed.

In conclusion, we measured magnetoresistance and Hall coefficients on nanocrystalline SmB_6 films down to 2 K. Although grain boundary scatterings and low carrier mobility in these films can possibly mask both weak-antilocalization and quantum oscillations, a linear positive MR is identified at low temperatures and is likely from the linear dispersion near a Dirac point. The sign

change of the Hall coefficient and its nonlinearity at lower temperatures may also be an indication of a robust surface conduction channel at low temperatures. The temperature dependence of these phenomena indicates that the surface states are bound to the opening of the hybridization gap and is consistent with the surface states being topological.

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