High-Field Magnetoresistance Measurements of the Surface States of Samarium Hexaboride using Corbino Structures

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Abstract

The recent conjecture of a topologically-protected surface state in SmB$_6$ and the verification of robust surface conduction below 4 K have prompted a large effort to understand the surface states. Conventional Hall transport measurements allow current to flow on all surfaces of a topological insulator, so such measurements are influenced by contributions from multiple surfaces of varying transport character. Instead, we study magnetotransport of SmB$_6$ using a Corbino geometry, which can directly measure the conductivity of a single, independent surface. Both (011) and (001) crystal surfaces show a strong negative magnetoresistance at all magnetic field angles measured. The (011) surface has a carrier mobility of 122 cm$^2$/V-s with a carrier density of $2.5 \times 10^{13}$ cm$^{-2}$, which are significantly smaller than indicated by Hall transport studies. Analysis of the angle-dependence of conductivity on the (011) surface suggests a combination of a field-dependent enhancement of the carrier density and a suppression of Kondo scattering as the likely origin of the negative magnetoresistance.

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1. INTRODUCTION

Samarium Hexaboride (SmB$_6$) has captured renewed interest due to recent theoretical predictions\cite{1,2} suggesting that it is a strong 3D topological insulator (TI), and also due to subsequent experimental verifications\cite{3–5} of a conducting surface state consistent with TI surface states predicted for the material. As a result, there has been a large volume of theoretical calculations\cite{6} and experimental works providing strong evidence that the surface states have a TI surface state contribution; these experiments include the observation of 2D surface states in Angle-Resolved Photoemission Spectroscopy (ARPES)\cite{7–10} (which in one case measures a helical spin dispersion\cite{10}), the observation of de Haas-van Alphen (dHvA) oscillations on distinct 2D crystallographic surfaces\cite{11}, the suppression of surface states through bulk magnetic impurity doping\cite{12,13}, and reports of weak anti-localization\cite{14,15}. SmB$_6$ stands out from the known 3D semiconductor TIs because its topological band structure arises from strong electron correlation effects. Furthermore, because SmB$_6$ has a fully insulating bulk below $\sim 4$ K, the electrical properties of the surface states can be probed directly and easily by transport measurements; this is not possible for the known 3D semiconductor TIs due to polluting conductivity from the bulk.

Although a Hall bar structure is typically used to characterize magnetotransport of both 2D and 3D conductive states, 3D TIs pose particular difficulties for this conventional geometry. All surfaces of the Hall bar contribute to the total conduction, including any edges or corners that are not perpendicular to the magnetic field, and may vary in surface condition due to preparation procedures such as polishing. For example, this can lead to an effective “edge channel” that would short the quantum Hall insulator state of the surfaces perpendicular to the field. Another complication arises if the surface states exhibit ambipolar conduction, as is indicated in calculations by Lu et al.\cite{6}. The Hall coefficient is sensitive to charge sign, and in a multi-channel scenario with both electron and hole conduction, the contributions of one to the Hall coefficient can compensate the other. Our own Hall bar measurements on SmB$_6$ indicated\cite{3} carrier densities that were unphysically large for a 2D system, perhaps because any or all of these complications reduced the measured value of the Hall coefficient. Unfortunately, these complications also now make a large volume of detailed low-temperature transport work in SmB$_6$ (which assumed the low-temperature resistivity plateau to be a bulk effect) very difficult to interpret, especially since details about crystal
size and geometry are usually not reported.

In this paper, we avoid these particular difficulties by fabricating Corbino disks on single surfaces of SmB$_6$. This geometry is not sensitive to the sign of the charge(s), and is sensitive only to the surface on which it is fabricated. The longitudinal conductivity, $\sigma_{xx}$, of the surface can be directly obtained from the 2-terminal resistance and the geometry of the disk. There is a geometrical diminution of $\sigma_{xx}$ under a perpendicular magnetic field due to resistive losses in the circulating current arising from the transverse conductivity, $\sigma_{xy}$. The conductivity of a single-carrier system is then given by

$$\sigma_{xx}(B) = \frac{ne\mu}{1 + \mu^2 B_\perp^2},$$

where $n$ is the carrier density of the surface, $\mu$ is the carrier mobility, and $B_\perp$ is the perpendicular component of the magnetic field. This dependence on the magnetic field allows us to obtain values for $\mu$ and $n$.

In this work, we measure $\sigma_{xx}(B)$ using a Corbino geometry on the (001) and (011) surfaces of SmB$_6$ at 0.3 K. Our results are consistent with a picture where Kondo scattering due to Sm$^{3+}$ ion magnetic moments in an outermost surface oxide layer dominates the low-field magnetoresistance (MR) of the surface states, and where the high-field MR is due to increases in $n$, accompanied by a small decrease in $\mu$ via short-range disorder scattering.

II. EXPERIMENTAL METHODS

We measured single-crystal bars of SmB$_6$ grown in an Al flux. We prepared individual (001) and (011) crystallographic surfaces by polishing with SiC abrasive pads (grit size P4000). We lithographically patterned the Corbino disks with an inner diameter of 300 $\mu$m and an outer diameter of 500 $\mu$m. We ashed the surfaces with oxygen plasma and evaporated 50/1500 $\AA$ Ti/Au contacts. We attached wires to the contacts using Au wirebonding and silver paint methods. Contact resistances were Ohmic both at 300 K and at 4 K. We performed AC resistance measurements using standard lock-in techniques in multiple magnet systems at the National High Magnetic Field Laboratory (NHMFL). We took additional measurements in a $^3$He cryostat with an 8 T superconducting magnet.
III. EXPERIMENTAL RESULTS

Figure 1 shows MR traces obtained at 0.3 K for multiple field angles, measured with respect to the surface normal, in the NHMFL 35 Tesla system for the (011) and (001) surfaces of SmB$_6$, respectively. The most apparent feature for both surfaces is the strong negative MR at all measured angles. We also note that we do not observe Shubnikov-de Haas (SdH) oscillations for either surface up to 45 T, which is perhaps surprising in light of the observation of dHvA oscillations at lower field values by our collaborators.[11]

One of the most striking features of the traces is their angle-dependence, which is primarily a result of the perpendicular field-dependence of $\sigma_{xx}$ arising from the Corbino geometry.
and included in the denominator of Equation 1. Taking the ratio of traces for in-plane magnetic field and magnetic field with arbitrary angle $\theta$ with respect to the surface normal eliminates $n$ and gives

$$\frac{\sigma(B_\parallel)}{\sigma(B)} = 1 + \mu^2 B^2 \cos^2(\theta), \tag{2}$$

from which we can directly obtain $\mu$. The ratio associated with each surface is plotted for different magnetic fields as a function of angle in Figure 1(c) and (d), and for each angle as a function of magnetic field in Figure 1(e) and (f). Both sets of ratios exhibit an apparent $\cos^2 \theta$ dependence, which is the expectation for a surface conduction in the Corbino geometry (Eq. 2), and the (011) ratios also approximately exhibit the expected $B^2$ dependence. Simple quadratic fits of the (011) field-dependent curves in Figure 1(e) yield a carrier mobility of 123 cm$^2$/V s and a carrier density of $2.5 \times 10^{13}$ cm$^{-2}$. Both of these values are much lower than previously reported for Hall bar transport measurements, which may suffer from the problems discussed earlier. However, they are both more consistent with values from ARPES measurements and other Corbino experiments, and the carrier density value is physically plausible. Such a low mobility suggests that SdH oscillations will not be detectable below 81 T, which explains why we do not observe them. Meanwhile, the (001) ratios do not exhibit a simple $B^2$ dependence, most likely due to the presence of multiple carrier channels which may have different MRs. A two-carrier formulation in which the channels have similar conductivities but very different carrier mobilities will yield a total $\sigma(B_\parallel)/\sigma(B)$ with a shape similar to the data ratios in Figure 1(f), but it will not quite fit the data without additional MR-related contributions to each channel. However, our data does not sufficiently constrain the parameters of such a multiple-carrier fit with MR. Thus, in the analysis that follows, we will limit our focus to the (011) surface, except where noted.

The MR, which is not explicitly included in Equations 1 or 2, is due to $B$-dependence of $n$, $\mu$, or both. A more detailed analysis allows us to investigate the relative contributions of $n(B)$ and $\mu(B)$ to the MR. The coupling between the orbital motion of 2D surface electrons and the external magnetic field is expected to show a $\cos^2 \theta$ dependence similar to that of Eqs. 1 and 2 and would not affect $\sigma(B_\parallel)$. Meanwhile, other mechanisms (e.g. contributions from the Zeeman splitting) are expected to be independent (or weakly dependent) on $\theta$. Because most of the $\theta$-dependence in the data comes from the Corbino geometry, and because $\sigma(B_\parallel)$ exhibits large MR, we proceed with the assumption that $n(B)$ and $\mu(B)$ are independent of the field angle $\theta$. (A small $\theta$-dependent contribution is expected to arise
FIG. 2. (Color online) (011) surface carrier density (filled squares) and mobility (open diamonds) obtained from angle-dependent fits of the data. Shaded areas represent uncertainty in the parameters of the angle-dependent fits. Best-fit curves for polynomial \( n(B) \) (blue) and corresponding \( \mu(B) \) (red) using the \( \theta = 25^\circ \) and \( \theta = 85^\circ \) data (solid lines), and using the \( \theta = 5^\circ \) and \( \theta = 85^\circ \) data (dotted lines). The vertical log scale allows direct comparison of the relative magnitudes of changes in \( n \) and \( \mu \).

from the weakening of TI backscattering suppression due to the magnetic field’s influence on the helical spin dispersion\(^{17}\). We calculate that this effect is negligible at the field values measured here.) We plot the carrier densities and mobilities obtained from \( \cos^2 \theta \) fits at constant \( B \) (e.g., Fig. 1(c) and (d)) as a function of magnetic field (symbols in Figure 2). The quality of the fits\(^{15}\) at large \( B \) supports the assumption that \( n(B) \) and \( \mu(B) \) are independent of \( \theta \), and \( n(B) \) and \( \mu(B) \) can be obtained with good precision. However, the fits (and the analytical form of Eqs. 1 and 2, solved for \( n(B) \) and \( \mu(B) \)) are divergently sensitive to noise near \( B = 0 \), so this method does not work well at low field values, which is evident in the uncertainty of the values in Figure 2.

To overcome this, we assume that \( n(B) \) can be approximated using an even polynomial in \( B \). By treating the polynomial coefficients as fitting parameters, we can determine a best fit for \( n(B) \) and \( \mu(B) \), constrained by two \( \sigma(B) \) traces at different \( \theta \) of our choosing. Solid (dotted) lines in Figure 2 show the best fit for a \( 6^\text{th} \)-order polynomial \( n(B) \) using the \( \theta = 85^\circ \) trace and the \( \theta = 25^\circ (\theta = 5^\circ) \) trace, along with the corresponding \( \mu(B) \). Fits at other angles
change the relative magnitude of $n(B)$ and $\mu(B)$ by $<10\%$, suggesting some small angle-dependence of $n(B)$ and $\mu(B)$ that is not sufficiently expressed in a two-parameter model, but the qualitative dependence on $B$ remains the same. Both the $\theta$-dependent fits and the $B$-dependent fits suggest that changes in carrier density are primarily responsible for the MR of the (011) surface; i.e., the MR is a result of large changes in the carrier density accompanied by small changes in the carrier mobility.

For the (001) surface, a naïve application of single-carrier $\cos^2 \theta$ fits above 25 T yields\textsuperscript{18} a constant mobility of 61 cm$^2$/V s and an increasing carrier density around $2 \times 10^{14}$ cm$^{-2}$. If such fits are believed, this suggests that the (001) surface’s MR is also dominated by changes in carrier density. However, below $\sim 25$ T, the fit residuals start becoming much larger\textsuperscript{18}. Meanwhile, a polynomial best-fit of $n(B)$ fails to reproduce the $B$-dependence of the data, giving credence to the notion that the analysis is complicated by the presence of multiple carrier channels with different MRs or another unknown $\theta$-dependent effect\textsuperscript{19}.

\section*{IV. DISCUSSION}

We now address the possible physical origins of the negative MR. Past measurements\textsuperscript{20} at 4 K have also observed strong negative MR. These researchers, assuming they were measuring fully bulk properties, attributed the negative MR to closure of the bulk gap $\Delta$ and an increase in $n_{bulk}$. Indeed, 4 K is very near the reported\textsuperscript{31,4} crossover temperature between surface-dominated and bulk-dominated conduction for similar flux-grown crystals. However, our own data is taken well below this transition temperature in a regime where the bulk is electrically dead, and the conduction we measure is purely due to the surface states. In this regime, the carrier density of the bulk bands is not related to the surface conduction, and a change in activated bulk transport with gap reduction is unable to explain the negative MR we observe\textsuperscript{21}. It is, however, possible that a change in the bulk structure could have some effect on the surface states at the Fermi level (especially a change in the Dirac point relative to the Fermi energy), causing a change in the surface state carrier density. Because the fits of our data indicate that $n(B)$ is the dominant source of the negative MR, it seems reasonable to attribute the negative MR to such a bulk-driven ($\theta$-independent) picture. However, our collaborator’s dHvA measurements\textsuperscript{11} suggest that the carrier density does not change significantly up to 45 T for any $\theta$. This disagreement, along with the large variations
FIG. 3. (Color online) Resistivity vs. temperature for the (a) (011) surface and the (b) (001) surface. The solid black line is data, the long-dashed red lines are logarithmic fits on a linear temperature background, and the short-dashed green line is a logarithmic fit on a quadratic temperature background.

among reported\textsuperscript{7–9,11} values for \( n, \mu, \) and \( k_F, \) remains to be resolved.

If we take the \( B \)-dependence of \( n \) as a given, we can investigate weaker features of the MR that are apparent in \( \mu(B) \). Motivated by the observation of magnetic hysteresis at low fields\textsuperscript{22}, we investigate magnetic impurity scattering as a likely contribution to the negative MR. We measured the Corbino resistances as a function of temperature with \( B = 0 \) (Figure 3). On both surfaces, as the temperature is reduced, we observe a logarithmic increase of the resistance, the coefficient of which is far from the quantum conductance. This, taken together with the low-field increase in \( \mu(B) \), suggests a TI surface Kondo scattering mechanism\textsuperscript{23,24}. The origin of magnetic scatterers is likely the formation of moments in Sm\(_2\)O\(_3\) at the surface of the crystal, which is exposed to ambient air during much of its preparation. Two recent X-ray studies have demonstrated an increase in magnetic Sm\(^{3+}\) at the surface\textsuperscript{25,26}, along with its expected net magnetic moment (in contrast to the non-magnetic bulk)\textsuperscript{25}, and a modest oxygen signal, indicating the existence of a thin (\( \approx 20 \) Å) oxide film\textsuperscript{26}. Regardless of the origin of the impurities, we assess Kondo scattering as a possible mechanism to explain both the temperature dependence and the low-field enhancement of \( \mu(B) \).

The logarithmic \( T \)-dependence of the electron scattering rate we observe can be fit using the following formula\textsuperscript{24}, developed for a 3D TI system with dilute magnetic impurities:

\[
\frac{1}{\tau} \propto 3 + J\rho \ln \frac{T}{T_K},
\]

where \( J \) is the coupling constant, \( \rho \) is the density of states at the Fermi energy, \( T \) is the
temperature, and \( T_K \) is the Kondo temperature calculated using the renormalization group approach. Here, \( \bar{\tau} \) represents the scattering-angle-averaged scattering time, since the spin-momentum locking of the TI surface states causes \( \tau \) to depend on the scattering angle. For the SmB\(_6\) surface, \( J \) might represent the coupling (hybridization) between the surface states and the paramagnetic Sm\(_2\)O\(_3\) \( f\)-states. The Kondo scattering produces negative MR according to the formula\(^{23,24,27}\)

\[
\frac{1}{\mu} = \frac{1}{\mu_d} + \frac{1}{\mu_M} \left( 3 + J \rho \ln \frac{T}{T_K} \right) \cos^2 \left( \frac{\pi}{2} M(B) \right),
\]

where \( \mu_M \) is the coefficient of the contribution from Kondo scattering, \( \mu_d \) is the mobility from disorder scattering alone, and \( M(B) \), whose relationship to \( B \) can be exactly calculated at low temperatures\(^{23} \), is the normalized magnetization of the impurities. Motivated by the experimental signatures of Kondo scattering, we apply this theory to SmB\(_6\), even though the surface magnetic moments from the Sm\(_2\)O\(_3\) might not be in the dilute limit. At zero magnetic field \((B, M = 0)\), the logarithmic fits shown in Figure 3, which include a linear background resistance of unknown origin, allow us to experimentally determine \( \mu_d \) and \( \mu_M \). The dependence on magnetic field \((B, M \neq 0)\), which arises from the suppression of spin-flip scattering due to Zeeman splitting, can then be predicted as a function of \( T \) and \( T_K \). Using the values from our logarithmic fits and for our value of \( n(B = 0) \), we plot computed values for \( \mu(B) \) for several different Kondo temperatures, alongside our fit of our experimental \( \mu(B) \) for comparison, in Figure 4. The low-field increase in \( \mu(B) \) fits quite well with Kondo scattering for \( T_K = 40 \) K. We note that if we were to ignore the evidence for \( B \)-dependent \( n \), and instead attribute the MR solely to changes in \( \mu(B) \), such a naïve fit would yield a much larger negative MR than can be explained by Kondo scattering alone\(^{18} \); the theoretical prediction and the experimental curve agree only by combining the Kondo effect together with the \( B \)-dependence of the carrier density.

The downturn of \( \mu(B) \) at higher magnetic fields is not a feature of Kondo scattering, but is qualitatively consistent with short-range disorder scattering mechanisms. It has long been known\(^{28} \) that in the high carrier density limit (in which the SmB\(_6\) surface states live), the mobility is partially determined by short range disorder scattering mechanisms (e.g., surface roughness scattering) and scales inversely with the carrier density, e.i., \( \mu \propto n^{-\alpha} \), where \( \alpha \) is determined by the particular scattering mechanism(s). This behavior has been observed in several semiconductor heterostructures\(^{20} \). In SmB\(_6\), as \( n(B) \) increases with increasing \( B \),
the short-range scattering time (and thus $\mu$) decreases with $B$. Typical values for $\alpha$ between $\frac{1}{2}$ and 2 are consistent with our data, but a precise determination of $\alpha$ from the data is problematic, since $T_K$ is not known (the model works over a range of values for $T_K$), and since $n(B)$ only varies by 15% over the fields measured (the dynamic range for determining a power-law relation is too small). However, this effect, together with the Kondo scattering, gives a picture that is qualitatively consistent with the $\mu(B)$ we extract from our fits, where the low-field negative MR is due mostly to Kondo scattering, and the high-field negative MR is due mainly to an enhancement in $n(B)$, which then causes a much weaker diminution of $\mu(B)$ via an increase in short-range scattering.

V. CONCLUSION

We have performed transport measurements of individual crystallographic surfaces of SmB$_6$. Both (001) and (011) surfaces display strong negative MR. The (011) surface exhibits a carrier density and mobility at values which are significantly lower than previously reported from transport methods, but which are more consistent with ARPES data. For both (001) and (011) surfaces, the temperature dependence suggests Kondo scattering from magnetic surface impurities. Fits of the angular dependence of our data suggest that the negative MR is primarily due to an increase in carrier density, especially at high field, but with some additional contribution from the suppression of Kondo scattering.
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See Supplemental Material at [URL] for further discussion of the (001) surface data, as well as an evaluation of Kondo scattering alone to explain the negative magnetoresistance.

Multiple channels giving rise to visible MR features at distinguishable magnetic field values is an indication that the channels likely have carrier densities and mobilities that differ by orders of magnitude, but have resistivities of the same order. In fact, this is a reasonable expectation in a system that exhibits both large and small Fermi pockets, as has been observed on the (001) surface of SmB$_6$ both by ARPES and dHvA measurements. The large pocket, which is centered about the X point and has a large carrier density, may suffer from short-range disorder scattering and have a comparatively small mobility, as discussed later. Meanwhile, the small pocket, which is centered around the Γ point and has a smaller carrier density, may be dominated by long-range impurity scattering, which allows a much higher mobility.

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Although we cannot rule out a bulk gap closure mechanism, our measurements are taken at a temperature at least one full order of magnitude below the crossover temperature. Pollution from bulk conduction at even 0.01% does not arise in an activated transport model until the band gap is only 13% of its zero-field value, which can happen no lower than $\sim 80$ T. It is possible that at 4 K (near the crossover temperature), transport measurements will be sensitive to any MR coming from the surface states.
We caution the reader that this formulation may be quantitatively different from the 2D Kondo scattering description appropriate for surface magnetic impurities. However, the qualitative behavior will be the same, which is sufficient here because we take the magnitudes of $\mu$, $\mu_d$, and $\mu_M$ as fitting parameters in the subsequent analysis.