Evidence for topological surface states in metallic single crystals of Bi$_2$Te$_3$

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Abstract

Bi$_2$Te$_3$ is a member of a new class of materials known as topological insulators which are supposed to be insulating in the interior and conducting on the surface. However, experimental verification of the conductive qualities of the surface states has been hindered by parallel bulk conductions. We report low temperature magnetotransport measurements on single crystal samples of Bi$_2$Te$_3$. We observe metallic character in our samples and large and linear magnetoresistance from 1.5 K to 290 K with prominent Shubnikov–de Haas (SdH) oscillations whose traces persist up to 20 K. Even though our samples are metallic, we are able to obtain a Berry phase close to the value of $\pi$, which is expected for Dirac fermions of the topological surface states. This indicates that we have obtained evidence for the topological surface states in metallic single crystals of Bi$_2$Te$_3$. Other physical measurements obtained from the analysis of the SdH oscillations are also in close agreement with those reported for the topological surface states. The linear magnetoresistance observed in our sample, which is considered as a signature of the Dirac fermions of the surface states, lends further credence to the existence of topological surface states.

Keywords: topological insulator, shubnikov de haas oscillations, linear magnetoresistance

(Some figures may appear in colour only in the online journal)

1. Introduction

Topological insulators constitute a new phase in condensed matter physics and have attracted considerable attention due to the interesting physics underlying them and the potential for their practical application. Topological insulators are different from ordinary insulators in the topology of their band structure. The band structure of topological insulators have states confined to the surface which span the band gap of the bulk band structure and as a result the surface of a topological insulator is conductive despite having an insulating bulk [1, 2]. The interesting physics does not end here; these surface states are protected from opening of a gap at the $\Gamma$ point in the Brillouin zone due to Kramer’s degeneracy as long as time reversal symmetry is not broken and the dispersion is linear near this point. Moreover the spin is locked to the direction of momentum in these states and as a result backscattering is prohibited unless the impurities are magnetic, which can cause a spin flip. Bi$_{1-x}$Sb$_x$, Bi$_2$Se$_3$, Bi$_2$Te$_3$, Sb$_2$Te$_3$ and Bi$_{1-x}$Sb$_x$Te$_{y-z}$Se$_z$ (BSTS) are the main topological insulators that have been discovered. However, most of the reported data on these materials has a conducting bulk due to defects and vacancies and as a result the detection of surface states in transport measurements has been rather difficult [3–11]. A way to circumvent this problem has been to study Shubnikov–de Haas (SdH) oscillations in the magnetoresistance of these samples and to see their dependence on the tilted magnetic field and then to single out oscillations due to two-dimensional (2D) Fermi surfaces corresponding to the surface states. The magnitude of the oscillations in case of a 2D Fermi surface are expected to be dependent on the perpendicular component of the magnetic field [7, 10]. One clear feature of the magnetoresistance of these topological insulators is its magnitude, which has been noted in a host of reports [3–6, 12–16] and came to the fore when Tang et al [17] Wang et al [18] and He et al [19] emphasized it in their publications. It was also noted that the magnetoresistance was linear and non-saturating as well as being of a significant magnitude [17–22].
Such large and linear magnetoresistance, which does not saturate even at high fields, was first observed in polycrystalline bismuth and other metals by Kapitza [23, 24]. Lifshits [25] et al explained that this was due to the open Fermi surfaces of the metals. However, bismuth has a small and closed Fermi surface. Abrikosov then formulated a theory that predicted linear magnetoresistance would be possible at high fields, although he considered the attainment of such high fields to be physically impossible [26]. However, the rediscovery by Yang et al [27] of the linear magnetoresistance in bismuth led to the establishment of a connection between Abrikosov’s theory and linear magnetoresistance [26]. The large magnetoresistance observed in non-magnetic silver chalcogenides by Xu et al [28] was explained using Abrikosov’s [29] quantum linear magnetoresistance theory. Then Hu and Rosenbaum [30] showed that it was possible to obtain linear magnetoresistance in InSb using both quantum and classical calculations. Quantum linear magnetoresistance was reported in multilayer graphene by Friedman et al [31]. Qu et al [13] reported linear magnetoresistance in single crystals of Bi$_2$Te$_3$. However, they reported magnetoresistance of both metallic and non-metallic samples. Linear magnetoresistance was observed only in the case of insulating crystals. The metallic crystals showed a quadratic dependence on the magnetic field at low fields. The linear magnetoresistance in topological insulators has mainly been explained on the basis of Abrikosov’s theory, attributing its origin to the surface states with Dirac dispersion [17–19]. However, alternate explanations also attribute it variously to the bulk states [20] and sample homogeneity [22]. We have found large, linear and non-saturating magnetoresistance in metallic crystals of Bi$_2$Te$_3$. Tang et al [17] reported linear magnetoresistance in nanoribbons of topological insulator Bi$_2$Se$_3$ and later He et al [19] reported similar linear magnetoresistance in Bi$_2$Se$_3$ thin films.

2. Experimental details

We performed electrical transport measurements on Bi$_2$Te$_3$ samples obtained from a natural single crystal. Thin samples were cleaved from this single large crystal using a razor blade and further thinned down by peeling off successive layers with Scotch tape. Since Bi$_2$Te$_3$ has a layered crystal structure, it cleaves easily perpendicular to the c-axis with the cleaved single crystals having their flat surfaces perpendicular to the c-axis. In figure 1(a), we show an x-ray diffraction pattern obtained for one of the cleaved single crystals using a PANalytical-X’Pert PRO diffractometer with Cu Kα radiation and in figure 1(b) we show the energy dispersive x-ray (EDX) spectrum for our sample which gives the chemical composition of our sample as 41.5 ± 0.1% Bi and 58.5 ± 0.1% Te by atomic percentage. The cleaved crystals were shaped in the form of rectangular bars and the resistivity measurements were made using a standard four-probe technique. The Hall measurement data reported for some of the samples were compiled at the same time as the resistivity measurements. The resistivity versus temperature, magnetoresistivity and Hall measurements were all performed in an ICEOxford make closed cycle refrigerator $^{DRY}$ICE$^{VTI}$ which has a temperature range of 1.3 K to 300 K and provides a magnetic field of up to 9 Tesla.

3. Results

The results of the resistivity versus temperature measurements for two different single crystal samples cleaved from the same bulk single crystal of Bi$_2$Te$_3$ are shown in figure 2. The temperature dependence of the resistivity shows that our samples are metallic. The residual resistivity ratio (RRR) in the case of the first sample (sample 1) is about 14 and for the second sample (sample 2) is about 16; these lie within the range reported in existing literature for this material [32, 33] with a higher RRR generally signifying greater crystallinity [34]. We also fit the resistivity data for both sample 1 and sample 2 to the equation $\rho = a + bT^n$ in the temperature range 18–230 K and we obtain values of $n = 2.085 \pm 0.005$ and 2.129 ± 0.001 respectively, which suggests Fermi liquid behavior. The uncertainty in the power of temperature $n$ was estimated using the bootstrap method of finding the estimates of uncertainty in the parameters of a fit [35]. In the inset of figure 2(b), we show the results of the Hall measurement performed on sample 2. The results show that our single crystal is p-type in
Figure 2. (a) Resistivity versus temperature for cleaved single crystal of Bi$_2$Te$_3$ (sample 1). The solid orange curve is a fit to the resistivity in the temperature range 18–230 K which yields a $T^{2.085}$ dependence of resistivity on temperature. (b) Resistivity versus temperature for cleaved single crystal of Bi$_2$Te$_3$ (sample 2). The solid pink curve is a fit to the resistivity in the temperature range 18–230 K which yields a $T^{2.129}$ dependence of resistivity on temperature. Inset: Hall resistivity against magnetic field at 1.5 K for the same sample.

nature, while a linear fit to the Hall voltage versus magnetic field above 1 Tesla yields a bulk charge carrier concentration $n_{\text{Hall}}^{\text{bulk}} = 5.87 \times 10^{17} \text{ cm}^{-3}$. Bi$_2$Te$_3$ crystals are known to be p-type due to anti-site defects of Bi on Te sub-lattice sites [33, 36, 37].

In figure 3 we show the magnetoresistance at 1.5 K for samples 1 and 2, for which the temperature variation of resistivity was shown in figure 2. The resistivity shows clear oscillations at high magnetic fields in both cases. However in the case of sample 2 the oscillations are more prominent and the background magnetoresistivity deviates from a linear dependence and saturates at high fields. These kind of oscillations in the magnetoresistance are commonly seen in metals and are known as the SdH oscillations [38]. SdH oscillations have previously been reported in this context [13, 21, 37, 39]. One can extract the size of the Fermi surface from the SdH oscillations and also its shape from the dependence of the oscillations on the angle between the magnetic field and different crystal axes using the semiclassical quantization formula

$$2\pi(n + \gamma) = \pi k_F^2 \frac{\hbar}{eB}$$

where $\hbar$ is Planck’s constant, $e$ is electron charge, $n$ is the Landau level index, $B$ is the magnetic field, $\pi k_F^2$ is the cross-sectional area at the extremum of the Fermi surface and $\gamma$ is a phase correction [13, 38, 40–42]. The value of $\gamma$ is different for normal fermions with parabolic dispersion and for Dirac fermions with linear dispersion because of the Berry phase [40–42]. However, there is an inconsistency in the literature regarding the relationship of $\gamma$ with the Berry phase and hence in the values of $\gamma$ for the normal fermions case and for the Dirac fermions case, which we discuss later in the section on the Berry phase.

Since for sample 1 the oscillations are not very prominent we took the derivative of resistivity with respect to magnetic
field and plotted it against the inverse of magnetic field as shown in the inset of figure 3(a). Clear SdH oscillations are visible at fields above 2 T in the derivative of resistivity. The minima in resistivity correspond to integer values \( n \) and the maxima to \( n + 1/2 \) [13]. Thus, the minima in the derivatives of the resistivity with respect to the magnetic field will correspond to \( n + 1/4 \) and the maxima to \( n + 3/4 \) [7]. A plot of the inverse field values at which maxima and minima in oscillations occur against the integer values known as the Landau indices which are used to label these peaks, will be a straight line and is known as a Landau level (LL) fan diagram. An LL fan diagram for sample 1 is shown in the bottom inset of figure 3(a) where we have plotted the inverse field values at which maxima and minima in the derivatives of resistivity occur against the Landau indices after labeling the peaks and troughs with the proper \( n + 3/4 \) and \( n + 1/4 \) values. The slope of this straight line, which is also equal to the frequency \( f \) of these oscillations, is given by

\[
\frac{1}{f} = \frac{2\pi e}{\hbar} \frac{1}{\pi K_f^2} \tag{2}
\]

Upon performing a linear fitting of the LL fan diagram in figure 3(a) we obtain a slope of 0.066 and the value obtained for the intercept on the \( n \) axis is 0.242. The SdH oscillations can give further information about the Berry phase of the charge carriers, while the topological surface state charge carriers are supposed to have a Berry phase of \( \pi \) or a Berry phase factor of 1/2 because of their Dirac dispersion relation [7]. We will comment further on the extraction of the Berry phase from the LL fan diagram in the section discussing the Berry phase. Additionally, a calculation from the slope of the fit to the LL fan diagram shows the Fermi wave vector \( k_F \) to be 0.021 \( \text{Å}^{-1} \). If we assume that this corresponds to a two-dimensional (2D) Fermi surface (FS), then the surface carrier concentration [7] is \( n_s = k_F^2/4\pi = 3.5 \times 10^{11} \text{ cm}^{-2} \) while, on the other hand, if we assume a three-dimensional (3D) FS, we get the bulk carrier concentration [38] as \( n_{BH}^{\text{SdH}} = k_F^2/3\pi^2 = 3.1 \times 10^{17} \text{ cm}^{-3} \).

In the case of sample 2, as the oscillations were much more pronounced, it was possible to use the maxima and minima in magnetoresistivity itself to index the Landau levels with the maxima labelled with proper \( n + 1/2 \) values and minima with \( n \) values. In the bottom inset of figure 3(b) we show the Landau level plot for sample 2. From the slope of the fit we obtain a \( k_F \) value of 0.0203 \( \text{Å}^{-1} \) and the intercept on \( n \) axis is 0.308. From the \( k_F \) value obtained in the case of sample 2 we get a surface carrier concentration of \( n_s = 3.28 \times 10^{11} \text{ cm}^{-2} \) if we assume a 2D FS and a bulk carrier concentration of \( n_{BH}^{\text{SdH}} = 2.83 \times 10^{17} \text{ cm}^{-3} \) if we assume a 3D FS. If we compare this with the Hall concentration obtained for sample 2 then we see that the SdH carrier concentration is smaller by a factor of 2.07. In the work done by Rischau et al the carrier concentration obtained from SdH oscillations had to be multiplied by a factor of 6 to obtain the total carrier concentration and this was accounted for by taking into consideration the six-valley model for the lowest valence band and highest conduction band of \( \text{Bi}_2\text{Te}_3 \) [37]. Kulbachinski et al also reported that the carrier concentration obtained from SdH was less than that obtained from the Hall effect and this was explained as being due to the filling of a second lower valence band [43]. We could not make a similar comparison for sample 1 as we had not performed a Hall measurement for that sample. A third sample for which we carried out both Hall and resistivity measurements gave Hall concentration values of 1.74 \( \times 10^{18} \text{ cm}^{-2} \) and from its Landau level index plot we got an SdH carrier concentration of 3.1 \( \times 10^{17} \text{ cm}^{-3} \) which is smaller by a factor of 5.3. This would suggest that the SdH oscillations in our sample originate from the bulk or it could also be the case that the SdH oscillations have a surface origin as is discussed in the section on mobility. The variation in Hall concentration in different samples cleaved from the same bulk single crystal has also previously been observed in \( \text{Bi}_2\text{Te}_3 \) and \( \text{Bi}_2\text{Te}_2\text{Se} \) single crystals [13, 44].

The magnetoresistance at various temperatures is plotted in figure 4. From the magnetoresistance of sample 1 in figure 4(a) we note two significant observations. Firstly, there is a large change in the magnitude of the resistivity of magnetic field which is as high as 267% at low temperatures up to 25 K and 20% at 290 K. Such large magnetoresistance is seen in very few materials and has great potential for practical application. The second significant observation is that the resistivity increases linearly with the magnetic field after an initial parabolic increase and also does not saturate at high fields. In the case of sample 2, as seen in figure 4(b), at low temperatures where the SdH oscillations are very prominent, the magnetoresistance tends to saturate. However the linear nature returns as the temperature rises and maximum magnetoresistance is obtained at intermediate temperatures, with the magnitude of magnetoresistance decreasing as the temperature rises to room temperature.

### 4. Discussion

The study of the SdH oscillations can give further information regarding the mobility, Berry’s phase and the effective mass of the charge carriers. In order to study the SdH oscillations, a linear background was subtracted from the magnetoresistance data in the case of sample 1 and a parabolic background in the case of sample 2. The SdH oscillations after these subtractions at different temperatures are shown in figure 5. One standard expression for the SdH oscillation is

\[
\Delta \rho / \rho_0 = \frac{5}{2} \sum_{r=1}^{\infty} b_r \cos \left( \frac{2\pi e B}{\hbar \omega_c} r - \frac{\pi}{4} \right) \tag{3}
\]

with the amplitude \( b_r \) given by

\[
b_r = \frac{(-1)^r}{\sqrt{r}} \frac{\hbar \omega_c}{2E_F} \frac{2}{\sin h(2\pi r k_BT/\hbar \omega_c)} \times \exp \left( -\frac{2\pi^2 r k_BT_D}{\hbar \omega_c} \right) \cos \left( \frac{\pi r g m^*}{2m_e} \right)
\]

where \( k_B, g, m^*, \omega_c = eB/m^* \) and \( T \) are Boltzmann-constant, Landé g factor, cyclotron frequency and temperature respectively [45]. \( T_D \) is the Dingle temperature and takes into account the broadening of the Landau levels. It is related to the lifetime \( \tau \) of a state expressed by \( T_D = \hbar /2\pi k_B \tau \). A similar expression that has been used for SdH oscillations in the case of

\[
\frac{D}{\rho_0} = \frac{5}{2} \sum_{r=1}^{\infty} b_r \cos \left( \frac{2\pi e B}{\hbar \omega_c} r - \frac{\pi}{4} \right) \tag{3}
\]

with the amplitude \( b_r \) given by

\[
b_r = \frac{(-1)^r}{\sqrt{r}} \frac{\hbar \omega_c}{2E_F} \frac{2}{\sin h(2\pi r k_BT/\hbar \omega_c)} \times \exp \left( -\frac{2\pi^2 r k_BT_D}{\hbar \omega_c} \right) \cos \left( \frac{\pi r g m^*}{2m_e} \right)
\]
in equation (3) through the $\omega_1^{1/2}$ term, was necessary in order to obtain a good fit. The parabolic contribution, $\alpha = -\gamma B + \delta B^2$, takes care of any remnant background in the oscillations and was found to improve the quality of the fit. This is evident from the fact that the coefficient of determination ($R^2$) for the fit with parabolic contribution is 0.9811 while that for the fit without the parabolic contribution is 0.8635.

We fitted the oscillations in resistivity at 1.5 K to this equation and the resulting fit, along with the original data, are shown in figure 6. The values of $A$, $\mu$, $\beta$ and $B_F$ thus obtained for both samples, along with the uncertainty in these parameters obtained by the bootstrap method of finding estimates of uncertainty in the parameters of a fit, are given in the caption for figure 6 [35].

### 4.1. Magnitude of SdH oscillations

As pointed out earlier in the results section, the magnitude of SdH oscillations in sample 2 is much larger than that of sample 1. This can be explained on the basis of equation (3) from which we see that the magnitude of oscillations is proportional to the background resistivity $\rho_0$. Thus it will be higher in case of sample 2 where resistivity at zero field is of the order of 1 m$\Omega$cm which is about 5 times higher than that of sample 1 where the resistivity at zero field is of the order 0.2 m$\Omega$cm. In fact the magnitude of oscillations at a particular field and temperature is about five times higher for sample 2 compared to sample 1. That the magnitude of oscillation is proportional to the zero field resistivity at a particular temperature is verified by the fact that the ratio of magnitude of oscillations to zero field resistivity $\Delta \rho/\rho_0$ for sample 1 and sample 2, which are 0.071 and 0.067 respectively, is of similar magnitude. We note a similar observation in the work of Schneider et al [45] who point out that the amplitude of SdH oscillations decreases across samples with increasing electron concentration. However, we also note that resistivity decreases across those samples. Additionally, Xiong et al [46] mention that although SdH oscillations were observed in all of their samples of Bi$_2$Te$_2$Se, they were the largest only for those samples where resistivity exceeded 4 Ohm cm.

### 4.2. Berry phase

There are two ways of extracting the Berry phase of the charge carriers from the SdH oscillations; one is from the LL fan diagram and the other is by fitting the SdH oscillations to equation (5). However, we have noticed in the existing literature an inconsistency in obtaining the Berry phase from the LL fan diagram using the semiclassical quantization method given by equation (1). Some authors report that the phase correction $\gamma$ in equation (1) is related to the Berry phase factor $\beta$ as $\gamma = 1 - \beta$ and that $\gamma = 1/2 (\beta = 0)$ for normal fermions and $0 (\beta = 1/2)$ for Dirac fermions [13, 41, 42]. If this is correct, then from equation (1) we see that the intercept on the $n$ axis is equal to $-\gamma$ and not $\beta$. However, in most experimental papers, it is assumed that the intercept on the $n$ axis is equal to $\beta$ itself [7, 16, 21, 46–51]. The same expression given in equation (1), may be interpreted in a contrasting way, as the intercept on the $n$ axis is related to the Berry phase factor $\beta$ as $\gamma = 1 - \beta$. This interpretation is consistent with the results of Schneider et al [45] who point out that the amplitude of SdH oscillations decreases across samples with increasing electron concentration. However, we also note that resistivity decreases across those samples. Additionally, Xiong et al [46] mention that although SdH oscillations were observed in all of their samples of Bi$_2$Te$_2$Se, they were the largest only for those samples where resistivity exceeded 4 Ohm cm.

**Figure 4.** (a) Relative change in magnetic field-dependent resistivity at various temperatures for sample 1. The magnetoresistivity shown for 240 K and 290 K are from a different sample cleaved from the same single crystal. (b) Relative change in magnetic field dependent resistivity at various temperatures for sample 2. The inset shows the temperature variation of Hall mobility for sample 2.
the convention in most experimental papers, then we see that $\beta$ for sample 2. The values of 
\begin{align*}
\beta_1 & = 0.242 \\
\beta_2 & = 0.308
\end{align*}
respectively, at 1.5 K for sample 2. Inset: fit of the temperature dependence of the SdH oscillation amplitude in the magnetoresistivity of a magnetic field of 5.028 T. (a) SdH oscillations at different temperatures after subtracting linear background in the case of sample 2. Inset: fit of the temperature dependence of the SdH oscillation amplitude in the magnetoresistivity of a magnetic field of 6.87 T. 

\begin{align*}
\gamma_{1,007} & = 0.242 \\
\gamma_{1,009} & = 0.308
\end{align*}
the values of $\beta$ from the LL fan diagram are simply 0.242 and 0.308 for samples 1 and 2 respectively and in that case they are closer to the values obtained from the SdH fit.

\subsection*{3.3 Mobility of charge carriers}
From the fit of SdH oscillations at 1.5 K we obtain a mobility of 
\begin{align*}
(2210 \pm 40) \text{ cm}^2 \text{Vs}^{-1}
\end{align*}
and $\mu_{SdH}$ to be 
\begin{align*}
9833 \text{ cm}^2 \text{Vs}^{-1}
\end{align*}
Hall mobility of 8800 cm$^2$Vs$^{-1}$ was higher than the SdH mobility of 1300 cm$^2$Vs$^{-1}$ for the in-plane field and these SdH oscillations with the in-plane field were attributed to the topological surface states of the 2D sidewalls \cite{53}. Furthermore, like in sample 2, in the case of the Bi$_2$Se$_3$ nanoplates also, the Hall concentration of $5 \times 10^{18}$ cm$^{-3}$ is about 2.8 times higher than the SdH concentration of $1.8 \times 10^{18}$ cm$^{-3}$ which can be obtained from the reported Fermi wave vector value, assuming a 3D FS. Thus the different Hall and SdH concentrations, along with the different Hall and SdH mobility in our samples, could imply that the SdH oscillations originate from the surface states, especially since the Berry phase extracted from the SdH oscillations points to the presence of Dirac charge carriers which are supposed to be in the surface states. However it should be noted that the SdH mobility can be smaller than the Hall mobility even if both have a bulk origin, because in the former the quantum lifetime $\tau$ plays an important role whereas in the latter the transport lifetime $\tau_t$ is the important physical quantity and generally
\[\tau_N > \tau.\] This is due to the fact that \(1/\tau_N\) acquires a factor of \((1 - \cos \phi)\) due to spatial averaging, where \(\phi\) is the scattering angle, while \(1/\tau\) does not and at low temperatures small angle scattering can dominate, which will make the factor \((1 - \cos \phi)\) small and in turn make \(\tau_N\) larger \[44\]. Nevertheless, this does not necessarily rule out the possibility that the SdH oscillations have a surface origin, as even in topological surface states the small angle scattering will dominate since backscattering is forbidden and hence \(\tau_N\) will be larger \[54\].

4.4. Effective mass of charge carriers from temperature dependence of SdH oscillation

The SdH oscillations are dampened as the temperature rises and as is evident from figures (a) and (b), traces of the oscillations can be seen up to 15 K in sample 1 and up to 20 K in sample 2. The amplitude of the SdH oscillation for a fixed value of the field was plotted for different temperatures for samples 1 and 2 in the inset of figures 5(a) and (b) and these values were calculated with the following expression:

\[\Delta \rho(T) = \Delta \rho(0) - \frac{2\pi^2 k_B T/\hbar \omega_c}{\sin h(2\pi^2 k_B T/\hbar \omega_c)}\]  

We chose the maxima at 6.87 T for sample 1 and at 6.124 T for sample 2. From this fitting the extracted effective mass of the charge carrier turns out to be 0.09 me for both samples, where \(m_e\) is the free electron mass. Qu et al \[13\] reported a value of 0.1 me for Bi2Te3.

4.5. Fermi velocity, Fermi energy and electron mean free path

From the value of effective mass \(m^*\) and Fermi wave vector, the Fermi velocity \(v_F\) using \(v_F = \frac{\hbar k_F}{m^*}\) comes out as \(2.7 \times 10^5\) m/s for sample 1 and \(2.6 \times 10^5\) m/s for sample 2, which is close to the value of \(1.4 \times 10^5\) m/s reported by Veldhorst et al \[13\] and \(3.7 \times 10^5\) m/s reported by Qu et al for Bi2Te3 \[13, 39\]. Similarly, the Fermi energy, \(E_F\) comes out at 37 meV for sample 1 and 35 meV for sample 2 using a relativistic formula \(E_F = m^* v_F^2\) \[13\]. Qu et al reported \(E_F\) in the range of 78–94 meV for the surface Dirac electrons in Bi2Te3. Comparing equations (3) and (4), we see that the Fermi energy is related to the frequency of oscillation in inverse magnetic field as \(E_F = (\hbar e/\mu)^*B_F\). Using this expression we obtain \(E_F\) values of 18 meV and 16.8 meV for samples 1 and 2 respectively. The Fermi energy obtained from the frequency of SdH oscillations is almost half of that obtained from the Fermi velocity using the relativistic formula. We note that this was also true in the case of some other reports when we compared the Fermi energy calculated from the Fermi velocity and the frequency of SdH oscillations, but the reason behind this is not clear \[21, 53\].

The mean free path of an electron \(l_e\) can be calculated from the SdH mobility and Fermi wave vector using the relation \(\mu = e l_e/\hbar k_F\) by which we obtain \(l_e\) of 31 nm and 37 nm for sample 1 and sample 2 respectively. The metalllicity parameter \(k_F l_e\) then turns out to be 6.5 and 7.5 for samples 1 and 2 respectively. As a comparison we note that \(l_e\) values ranging from 105 nm to 219 nm \[13, 21, 39, 49\] and \(k_F l_e\) values ranging from 13.8 to 66 have been reported for the surface states of Bi2Te3 in earlier papers \[13, 21, 49\].

4.6. Saturation of magnetoresistance

As pointed out in the results section, the magnetoresistance of sample 2 differs from that of the sample 1 at low temperatures. The magnetoresistance of sample 1 is large, linear and non-saturating with the magnitude dropping as the temperature rises, while in sample 2 the magnetoresistance at low temperatures has a tendency to saturate. The magnetoresistance however becomes linear at high fields once the temperature rises above 100 K and from then on the magnitude of magnetoresistance decreases as the temperature rises, as in sample 1. This can be explained on the basis of the classical magnetoresistance theory according to which the condition for saturation to set in is that \(\omega_c \tau \gg 1\) where \(\omega_c\) is the Larmor frequency and \(\tau\) is the collision time \[26\]. This condition can be re-written as \(\mu B \tau \gg 1\) where \(\mu\) is the mobility and \(B\) the magnetic field, from which we see that as mobility decreases the condition for saturation to set in is fulfilled at progressively higher values of the magnetic field.

We have shown in the inset of figure 4(b) the temperature dependence of Hall mobility for sample 2 from which it is evident that the change in mobility is very small up to 25 K and then falls drastically as the temperature rises. This coincides with the temperature range where saturation is seen in the magnetoresistance and as temperature rises the mobility decreases leading to an increase in the value of the magnetic field at which saturation will set in thus making it unattainable within the maximum magnetic field obtainable in our experiments.

4.7. Large and linear magnetoresistance

The magnetoresistance of our Bi2Te3 samples is characterized by a large magnitude which is substantial even at room temperature and a linear dependence of the resistivity on the magnetic field after an initial parabolic rise that does not saturate even at a field of 9 T. The only exception is that the magnetoresistance at low temperatures in sample 2 shows a saturating behaviour. But even in this sample, linear magnetoresistance makes a comeback at temperatures above 100 K and thereafter it is similar to that of sample 1 in terms of linearity and magnitude. Such large and linear non-saturating magnetoresistance has been reported earlier in insulating Bi2Te3 \[13, 18\]. Qu et al reported magnetoresistance studies on metallic crystals of Bi2Te3, but the magnetoresistance was not linear \[13\]. Recently, linear magnetoresistance has been reported in metallic Bi2Te3 by Yue et al and Hamdou et al \[20, 21\]. It should be noted though that Yue et al report metallic samples even though these were taken from the same bulk crystal used in the earlier work of Wang et al who reported insulating samples \[18, 20\]. Thus our report is among one of the few instances of large, linear magnetoresistance in metallic Bi2Te3 bulk single crystals.

From figure 4 it can be seen that the magnitude of the magnetoresistance at a field of 9 T is 267% at low temperatures and 20% at 290 K for sample 1, while for sample 2 it reaches a maximum of 50 K with a magnitude of 109% and at 290 K it is 20%. To compare it with other reports we note that Qu et al \[13\] reported a similar magnitude of 250% for their...
metallic crystals and about 180% for their insulating crystals, while Wang et al [18] reported magnetoresistance up to 625%. The large and linear magnetoresistance generally decreases in magnitude as the temperature rises and this is also observed in our samples [13, 17, 19].

The linear magnetoresistance in topological insulators has been attributed to the presence of topological surface states with a Dirac dispersion relationship [17–19]. The classical theory of magnetoresistance accounts for the quadratic dependence of the magnetic field resistivity at low fields followed by saturation at high fields seen in normal metals [17, 26]. The linear and non-saturating magnetoresistance observed in silver chalcogenides, [28] InSb, [30] multi-layer graphene, [31] and in topological insulators can be explained by the theory of quantum magnetoresistance put forward by Abrikosov [29]. This theory predicts that in the case of a gapless and linear dispersion relation, it is possible to have a linear and non-saturating magnetoresistance. This holds true in the case of topological insulators which have a surface state with a linear dispersion relation. According to this theory, in order to observe linear magnetoresistance the magnetic field should be so high that only the lowest Landau level is populated and also the temperature should not be so high that the thermal energy exceeds the Landau level separation. In terms of the charge carrier concentration, $n_e$ and effective mass of the charge carriers, $m^*$, the conditions to be satisfied by the field $B$ and temperature $T$ are

$$n_e \ll \left( \frac{eB}{\hbar} \right)^{3/2} T < v\sqrt{eBeB/k_B}$$  

(7)

where $v$ is taken to be $10^{16}$ cm s$^{-1}$ and $\hbar$ is the Planck constant.

We now check whether the conditions given in equation (7) for Abrikosov’s theory of linear magnetoresistance to be applicable are fulfilled in our sample. As far as the condition for only the lowest Landau level being filled is concerned, we can see from the Landau index plot of figure 3 that even at high fields more than one Landau level is filled. However Hu et al [30] have shown that linear magnetoresistance can set in at low magnetic fields even when more than one Landau level is populated. We note from figure 4(a) that in our samples there is a crossover at around 2 T from quadratic dependence on magnetic field at low fields to a linear dependence at higher fields. Such a crossover has been seen in almost all instances of linear magnetoresistance reported for topological insulators as well as other materials [13, 17, 19, 30, 31]. Thus beyond the crossover field of 2 T the first condition of equation (7) must be satisfied and for linear magnetoresistance to be seen at a field of 2 T we obtain the condition that the carrier concentration should be less than $1.67 \times 10^{17}$ cm$^{-3}$ which is clearly exceeded by the carrier concentration of $5.87 \times 10^{17}$ cm$^{-3}$ obtained from the Hall measurement. However if we convert the surface carrier concentration $n_s = 3.5 \times 10^{11}$ cm$^{-2}$ obtained from SdH oscillations to bulk concentration using the thickness of the sample which is 136 $\mu$m, then we obtain a bulk concentration of $2.6 \times 10^{13}$ cm$^{-3}$ which is well below the limit. This would suggest that the linear magnetoresistance could originate from the surface states. The second condition of equation (7) gives a temperature of nearly 421 K for a field of 2 T. This agrees with the fact that we observe linear magnetoresistance up to room temperature. Thus the linear magnetoresistance in our sample satisfies the criteria for observing quantum magnetoresistance thereby signifying that it is a signature of the topological surface states since linear magnetoresistance is supposed to rise in the case of gapless linear dispersion states. Since the SdH oscillations have already shown signs of Dirac states, evidence that the linear magnetoresistance is originating from the same surface states would further strengthen the evidence for topological surface states.

However, one observation that counters the above analysis and that of the surface origin of SdH oscillations is that the values of surface conductance calculated from surface carrier concentration and mobility obtained from the SdH oscillations come to $\approx 1 \times 10^{-4} \Omega^{-1}$ for both our samples and compared to our total conductance values calculated from the resistivity at zero fields these are very small. This makes it difficult to maintain that the large linear magnetoresistance and SdH oscillations originating from the surface states can dominate the magnetoresistance even though the surface conduction is a very small fraction of the total conduction. One thing which is very interesting to note is that the conductance of surface states obtained from the physical quantities extracted from the SdH oscillations is almost universally in the range of $10^{-3}$–$10^{-4} \Omega^{-1}$ for many topological insulators reported in existing research; this is also the case in our samples [6, 7, 12, 13, 16, 17, 21, 39, 46, 47, 49, 50, 53].

Only the total resistance in the majority of earlier papers is generally larger than in our own study. Thus it is interesting to note that although the surface conduction is a small fraction of total conductance in our samples, the surface conduction obtained from the SdH oscillations matches that reported in existing papers on topological surface states. Linear magnetoresistance, which is generally attributed to the surface states and surface SdH oscillations have been seen in topological insulator samples where the percentage of surface conduction ranges from 0.1–0.3% of the total conduction [10, 13, 53]. Even if one could attribute the SdH oscillations in these samples to the surface states it is difficult to do so for the large linear magnetoresistance. In fact in Bi$_{1-x}$Sb$_x$, studies of SdH and the angle-dependent magnetoresistance of de Haas-van Alphen (dHvA) effect showed that bulk and 2D Fermi surfaces existed, but the magnitude of the dHvA oscillations was of an order of magnitude larger than expected for the number of surface state carriers [12]. The authors speculated that a 2D cylindrical Fermi surface inside the bulk Brillouin zone could explain this discrepancy. Since the non-zero Berry phase obtained for the SdH oscillations in our samples point to these coming from non-trivial surface states with Dirac dispersion, one possibility could be the existence of many parallel surface states of which the conductance adds up to make a significant contribution to the total conduction, thus enabling them to dominate the overall conduction. Such layered transport with the bulk behaving as many parallel 2D electron systems has already been reported in highly doped Bi$_2$Se$_3$, although these did not have Dirac dispersion [6].
One is tempted to draw an analogy from the case of graphite where signatures of Dirac fermions were seen in the transport properties from the Berry phase of the SdH oscillations even though their relative phase volume was very small as predicted by the band structure [55, 56]. Thus it could be possible that bulk single crystals of topological insulators behave as many layers of parallel topological insulators with surface states in the fashion of many layers of independent graphene in bulk graphite. Moreover, alternate explanations of the linear magnetoresistance in topological insulators exist, like the recent finding by Yue et al from their angular dependent magnetoresistance studies, that the linear magnetoresistance in Bi$_2$Te$_3$ could have its origin in the bulk states [20]. Also sample inhomogeneity, which can also lead to linear magnetoresistance but was previously considered invalid in the context of topological insulators [17, 18], has recently been shown to be behind the linear magnetoresistance in Bi$_2$Se$_3$ [22]. Thus the issue of the linear magnetoresistance in topological insulators and the dominance of surface SdH oscillations in the magnetoresistance in spite of a more conductive bulk remain open questions and further studies are necessary. The extremely large value of the magnetoresistance and its linearity, along with the fact that it persists up to room temperature, opens up possibilities of this material finding use in practical applications.

5. Conclusion

In summary, we report large, linear and non-saturating magnetoresistance in metallic single crystals of Bi$_2$Te$_3$ with magnetoresistance as large as 267% at low temperatures, which persists up to room temperature although its magnitude is reduced. This is among the very few cases of linear magnetoresistance that have been reported in metallic single crystals of Bi$_2$Te$_3$. We also observe prominent SdH oscillations up to 20 K and the values of Berry’s phase extracted from the Landau plot and from the fit to the SdH oscillations, are close to the value of $\pi$ expected for the Dirac fermions of the topological surface states and point to the existence of such states in our sample. The values of mobility, surface carrier concentration, effective mass and Fermi velocity extracted from the SdH oscillations are close to those reported in existing literature on the topological surface states. The values of Fermi energy and electron mean free path are also comparable to what has been reported earlier for the surface states of Bi$_2$Te$_3$. The conditions for Abrikosov’s quantum magnetoresistance to be seen are fulfilled by the 2D carrier concentration obtained from the SdH oscillations and this may imply that the large and linear magnetoresistance originates from these 2D states, giving further indications of a topological surface state in our samples. One contradiction that remains is how the surface states dominate the magnetoresistance despite the surface conduction being very much smaller than the total conductance. Together, the SdH oscillations and linear magnetoresistance provide evidence for topological surface states in the metallic single crystal of Bi$_2$Te$_3$.

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