Quantum Oscillations in a Topological Insulator Bi$_2$Te$_3$Se with Large Bulk Resistivity (6 Ωcm)

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We report the observation of prominent Shubnikov-de Haas oscillations in a Topological Insulator, Bi$_2$Te$_3$Se, with large bulk resistivity (6 Ωcm at 4 K). By fitting the SdH oscillations, we infer a large metallicity parameter $k_F\ell = 41$, with a surface mobility ($\mu_s \sim 2,800$ cm$^2$/Vs) much larger than the bulk mobility ($\mu_s \sim 50$ cm$^2$/Vs). The plot of the index fields $B_\nu$ vs. filling factor $\nu$ shows a $\nu$-shift, consistent with massless, Dirac states. Evidence for fractional-filling states is seen in an 11-T field.

Topological Insulators are predicted to bear current-carrying, massless, Dirac surface states that traverse the bulk energy gap [1–4]. These unusual surface states have been observed by angle-resolved photoemission spectroscopy (ARPES) [5, 8] and scanning tunneling microscopy experiments (STM) [9]. Quantization of the Dirac states into Landau Levels has been demonstrated in STM experiments [10, 11]. Observation of the surface currents by transport has been more challenging [12, 13]. Recently, however, encouraging progress has been achieved. The surface SdH oscillations and surface mobility was measured in Bi$_2$Te$_3$ crystals [14]. The existence of surface states at fractional filling in a pulsed magnetic field were reported in (Bi,Sb)Se$_3$ [13]. Here we report the observation of prominent SdH oscillations in crystals of Bi$_2$Te$_3$Se with very large bulk resistivity. Evidence for fractional-filling states become apparent at relatively low magnetic fields.

Recently, ARPES experiments have shown that Bi$_2$Te$_2$Se displays a single topological surface state [16]. Independent of our experiment, SdH oscillations in this compound have also been reported by Y. Ando’s group [17]. We were motivated to grow crystals of Bi$_2$Te$_2$Se by reasoning that Bi$_2$Se$_3$ crystals grow with a stable density of Se vacancies. Electrons donated by the vacancies pin the Fermi energy $E_F$ to the conduction band, resulting in a negative thermopower $S$ at low temperature $T$. By contrast, as-grown Bi$_2$Te$_3$ typically has a positive thermopower. By growing a series of the hybrid semiconductor Bi$_2$Se$_{1+x}$Te$_{2-x}$, we have found that the low-temperature thermopower varies systematically, reflecting changes in $E_F$. Crystals were grown by a modified Bridgeman method from high purity elemental starting materials. After heating for one day at 850 C in a clean evacuated quartz tube, the melt was cooled in a temperature gradient to 500 C where it was left to anneal for 2 days before cooling rapidly to room temperature. For the composition Bi$_2$Te$_2$Se, $S$ rises to very large values. With a razor blade, we cleaved thin crystals and attached contacts using silver paint. For the sample here, the crystal thickness $d = 110$ µm, while the distance between voltage leads equals 0.5 mm. The resistivity profile measured at $B = 0$ is shown in Fig. 1a. Below 40 K, the value of $\rho$ attains values in the range 5-6 Ωcm, or ~1000 times higher than in non-metallic Bi$_2$Te$_3$. Expressed as an areal resistance $R_D = \rho/d$, the low-$T$ resistance corresponds to $R_D = 400$ Ω. Despite the high resistance, $\rho$ is only weakly $T$-dependent, displaying a log $T$ increase as $T \to 0$. The observed Hall coefficient $R_H$ below 10 K (n-type) implies a very small bulk carrier concentration $n_b \sim 2.6 \times 10^{16}$ cm$^{-3}$. Combining this with the observed

![FIG. 1: (Color online) The resistivity of a cleaved crystal of the Topological Insulator Bi$_2$Te$_2$Se. Panel (a) shows $\rho$ vs. $T$ measured in $B = 0$. Below 10 K, $\rho$ attains the value 5.5 Ωcm, or an area resistance $R_D \approx 400$ Ω. Despite the non-metallic value of $R_D$, sizeable quantum oscillations are observed below 38 K. Panel (b) displays the prominent SdH oscillations observed in the derivative $d\rho_{xy}/dB$ vs. $B$ at 4.4 K.](image-url)
ρ, we infer a low bulk mobility \( \mu_b \sim 50 \text{ cm}^2/\text{Vs} \).

Such a low \( \mu_b \) should not produce SdH oscillations for \( B < 14 \text{ T} \). Surprisingly, however, the Hall resistivity \( \rho_{yx} \) displays prominent SdH oscillations that may be resolved up to 40 K. To date, we have detected SdH oscillations in 4 crystals of Bi\(_2\)Te\(_3\)Se with \( \rho-T \) profiles similar to that in Fig. 1a. Figure 1b shows the trace of the derivative \( d\rho_{yx}/dT \) vs. \( B \) at \( T = 4.4 \text{ K} \). Independently, SdH oscillations were also observed in Bi\(_2\)Te\(_3\)Se by Y. Ando et al. [17].

The surface conductance and bulk conductance act as parallel channels for charge transport. The observed conductivity \( \sigma_{ij} \) is then the sum

\[
\sigma_{ij} = \sigma_{ij}^b + G_{ij}^s/d,
\]

where \( \sigma_{ij}^b \) is the bulk conductivity and \( G_{ij}^s \) the conductance matrix of the surface states. To exploit the additivity, we have converted the measured resistivity matrix \( \rho_{ij} \) to the conductivity matrix \( \sigma_{ij} \). To amplify the surface contribution to \( \sigma_{xy} \), we define \( \Delta\sigma_{xy} = \sigma_{xy} - \langle \sigma_{xy} \rangle \) where \( \langle \sigma_{xy} \rangle \) is a smoothed background.

Figure 2 displays the subtracted quantity \( \Delta\sigma_{xy} \) versus \( 1/B \) over the temperature interval \( 0.3 < T < 38 \text{ K} \). With increasing \( T \), the amplitude of the SdH oscillations decreases rapidly as \( T \) increases above 10 K.

To analyze the SdH oscillations, we have fitted the curves using the standard expression \[18\]

\[
\frac{\Delta\sigma_{xy}}{\sigma_{xy}} = \left( \frac{\hbar\omega_c}{2E_F} \right)^{1/2} \frac{\lambda}{\sinh \lambda} e^{-\lambda D} \cos \left[ \frac{2\pi E_F}{\hbar\omega_c} + \frac{\pi}{4} \right],
\]

with \( \lambda = 2\pi^2 k_B T/\hbar\omega_c \) and \( \lambda_D = 2\pi^2 k_B T_D/\hbar\omega_c \), where \( \omega_c \) is the cyclotron frequency and the Dingle temperature is given by \( T_D = \hbar/(2\pi k_B T) \), with \( T \) the lifetime. Compared with the SdH expression for the conductivity \( \sigma_{xx} \), the phase \( \phi \) in the Hall conductivity is shifted by \( \pi/2 \) (\( -\pi/4 \to \pi/4 \)). For 2D systems, we may write the SdH frequency as \( 2\pi E_F B/(\hbar\omega_c) \), which simplifies to \( 4\pi^2 k_F^2 n_s/e \), with the 2D carrier density \( n_s = k_F^2/4\pi \) (per spin). As shown in Ref. 19, Eq. 2 may be employed in a Dirac system if we write the cyclotron mass as \( m_c = E/v_F^2 \).

We have found that the oscillations cannot be fitted using one SdH frequency. For \( T < 6 \text{ K} \), the sharp decrease of the oscillation amplitude for \( B^{-1} > 0.12 \text{ T}^{-1} \) suggests beating between 2 terms of nearly equal frequencies. Indeed, a good fit is obtained if we add a second term identical to the one in Eq. 2 except for a slight difference in \( n_s \). The measured curve of \( \Delta\sigma_{xy} \) was fitted with the 2 terms (the absolute value of the surface Hall conductance \( G_{xy} \) is not known). There are altogether 5 adjustable parameters \( (n_{s1}, \text{amplitude } A_1, \text{ with } i = 1.2) \) and \( T_D \) (assumed same for both). The best fit (Fig. 4k) is obtained with \( A_1 = A_2 \) and and densities differing by only 5% \( [(n_{s1}, n_{s2}) = (1.79, 1.71) \times 10^{12} \text{ cm}^{-2}] \), corresponding to an average Fermi wavevector \( k_F = 0.047 \text{ Å}^{-1} \). The fit yields \( T_D = 8.5 \pm 1.5 \text{ K} \), which corresponds to a mean-free-path \( \ell = 70-100 \text{ nm} \) and a surface mobility \( \mu_s = e\ell/k_F = 2.800\pm 250 \text{ cm}^2/\text{Vs} \). Interestingly, \( \mu_s \) is strongly enhanced over \( \mu_b \) (by ~60).

By fitting to the decrease in amplitude with \( T \) at fixed \( B = 12.4 \text{ T} \), we obtain an effective mass \( m^* = 0.089 m_e \). Together with \( k_F \), we obtain a Fermi velocity \( v_F \sim 6 \times 10^5 \text{ m/s} \), higher than that in Bi\(_2\)Te\(_3\).

The high mobility provides strong evidence that the SdH oscillations arise from surface states. Suppose for the sake of argument that the oscillations come from bulk states. The SdH period must then be identified with a 3D Fermi sphere of radius \( k_F = 0.047 \text{ Å}^{-1} \), or a 3D density of \( 3.3 \times 10^{18} \text{ cm}^{-3} \). The inferred \( \mu \) then implies a 3D resistivity \( \rho_b \sim 0.7 \text{ mΩcm} \) at 4.4 K. The large discrepancy (factor of 9,000) from the observed value argues firmly against a bulk origin.

Hence we conclude that the SdH oscillations come from high mobility surface carriers. The two periods

![FIG. 2: (Color online) The Hall SdH oscillations versus 1/B at selected T. In Panel (a), we have plotted the subtracted Hall conductivity \( \Delta\sigma_{xy} \) to highlight the decrease of the amplitude over 2 decades in T (0.3 to 38 K). Panel (b) displays traces of the Hall resistivity \( \rho_{yx}/dB \) at 5 temperatures. The minima in \( d\rho_{yx}/dB \) are used to fix the index field \( B_x \) (dashed lines with \( \nu \) indicated). For LL with \( \nu < 6 \), sharp structures are observed between integer filling.](image-url)
likely arise from the large surfaces of the cleaved crystal that are normal to $B$. From the inferred $n_{s1}$ and $\mu$, we find that the conductance of each surface is $G_s = \frac{1}{2}(e^2/h)k_F\ell \simeq 0.72$ mS (or $R_{\square} \sim 1.39$ kΩ).

The prominence of the oscillations allows us to address the question whether the surface states have a Dirac dispersion. In principle, one may plot the “index” field $1/B_\nu$ versus the integers $\nu = 1, 2, \ldots$, and track the intercept in the limit $B \to \infty$. However, away from the quantum Hall effect (QHE) regime at low $B$, it is sometimes uncertain whether one should take the maxima or minima of $\rho_{xx}$ or $\rho_{yx}$ for the index field. To us, the most natural choice is the field defined by the filling factor $\nu = N_e/N_\phi$, viz.

$$B_\nu = n_s\phi_0/\nu,$$

where $N_e$ and $N_\phi$ are the total number of electrons on a surface and the number of flux quanta $\phi_0 = h/e$ piercing the surface (hereafter, we focus on one surface, i.e. one spin degree of freedom). Equation (3) is equivalent to having exactly $\nu$ flux quanta enclosed in the cyclotron orbit for an electron in the LL $\nu$. $B_\nu$ are the fields at which $\zeta$ falls between LLs (arrows). The inset shows the 2D Dirac energy surface in zero $B$. In Panel (b), the measured values of $1/B_\nu$ are plotted versus filling factor $\nu$. The 5 values of $1/B_\nu$ fall on a straight line that intercepts the $\nu$ axis at -0.55, consistent with a Dirac spectrum.

FIG. 3: (Color online) Fits of $\Delta \sigma_{xy}$ to extract mobility. Panel (a) shows the fit to $\Delta \sigma_{xy}$ at $T = 4.4$ K to Eq. (2). The rapid decrease of the oscillation amplitude for $1/B > 0.09$ T$^{-1}$ reflects interference between 2 terms of equal amplitudes and densities $(n_{s1}, n_{s2}) = (1.8, 1.7) \times 10^{12}$ cm$^{-2}$. The fit yields mobility $\mu = 2.800$ cm$^2$/Vs, and $k_F\ell = 41$. Panel (b) shows the fit of the envelope versus $T$ with $B$ fixed at 12 T. The fit yields $m^* = 0.089$ m$_0$ (free mass). With $k_F = 0.047$ Å$^{-1}$, the inferred velocity $v_F = 6 \times 10^5$ m/s.

FIG. 4: (Color online) Construction used to fix the filling factor for a Dirac spectrum. Panel (a) sketches schematically the step-like increase of $\sigma_{xy} = (e^2/h)(\nu + \frac{1}{2})$ versus energy $E$, where $\frac{1}{2}$ arises from the $\nu = 0$ LL at the Dirac point. The half-ovals are broadened Landau Levels centered at $E_\nu = h\nu^2v_F\sqrt{2\nu}$. $B_\nu$ are the fields at which $\zeta$ lies between LLs (arrows). The inset shows the 2D Dirac energy surface in zero $B$. In Panel (b), the measured values of $1/B_\nu$ are plotted versus filling factor $\nu$. The 5 values of $1/B_\nu$ fall on a straight line that intercepts the $\nu$ axis at -0.55, consistent with a Dirac spectrum.
FIG. 5: (Color online) Panel (a): Expanded view of $d\rho_{yx}/dB$ for $4 < \nu < 6$ showing sharp maxima at non-integer values of $\nu$, at $T$ from 0.35–10 K. The arrows locate the more prominent peaks. In Panel (b), the peak positions plotted against $\nu$ align well with fractional values of $\nu$ (arrows mark the values $\nu = \frac{13}{3}, \frac{9}{2}, \ldots, \frac{17}{3}$).

at much lower fields (11 T). As shown in Fig. 5(a), an intriguing array of sharp maxima in $d\rho_{yx}/dB$ (arrows) is apparent for $4 < \nu < 6$. The peaks become weaker as $T$ increases from 0.35 K, but some are still resolved at 10 K. Interestingly, the peak positions align well with fractional values of $\nu$, as shown in Fig. 5(b).

As in the fractional QHE regime in GaAs and graphene, we interpret the peaks as evidence for many-body states that are stabilized at fractional $\nu$. However, there are several puzzling features (when compared with the standard FQHE phenomenology). First, in Fig. 5(a), we find that the maxima of $|d\rho_{yx}/dB|$ locate the fractional values of $\nu$ whereas $B_x$, for integer $\nu$, is fixed by the minima. We do not have an explanation for this inversion. Second, the fractional-filling peaks are observed at fairly large $\nu$, whereas in GaAs, they are difficult to resolve for $\nu > 2$, despite the much higher $\mu_s$ in GaAs. Finally, unlike in GaAs, the fractions corresponding to $\frac{1}{2}$ are more prominent here (particularly $\frac{11}{2}$) than those corresponding to $\frac{1}{3}$ or $\frac{1}{5}$. In this system, we may incorporate features such as the linear Dirac dispersion, strong spin-orbit interaction, and strong suppression of $2k_F$ scattering to understand the states at fractional filling.

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