Quantum oscillations and nontrivial transport in (Bi$_{0.92}$In$_{0.08}$)$_2$Se$_3$

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Abstract

Quantum phase transition in topological insulators has drawn heightened attention in condensed matter physics and future device applications. Here we report the magnetotransport properties of single crystalline (Bi$_{0.92}$In$_{0.08}$)$_2$Se$_3$. The average mobility of $\sim$1000 cm$^2$V$^{-1}$s$^{-1}$ is obtained from the Lorentz law at the low field ($< 3$ T) up to 50 K. The quantum oscillations rise at a field of $\sim$ 5 T, revealing a high mobility of $\sim$1.4 $\times$ 10$^4$ cm$^2$V$^{-1}$s$^{-1}$ at 2 K. The topological Dirac fermions are evident by the nontrivial Berry phase in the Landau Fan diagram. The properties make the (Bi$_{0.92}$In$_{0.08}$)$_2$Se$_3$ a promising platform for the investigation of quantum phase transition in topological insulators.

Keywords: quantum phase transition, topological insulators, quantum oscillations, topological Dirac fermions, nontrivial Berry phase

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1. Introduction

Three-dimensional (3D) topological insulators (TIs) have attracted worldwide attention in condensed matter physics and future device applications due to the unique surface states protected by time-reversal symmetry\textsuperscript{1-5}. On behalf of the topology of TIs, the surface states come from the band inversion of the bulk with strong spin-orbit coupling (SOC). The topological quantum phase transition (QPT) in TIs can be realized by finite-size effects in ultrathin films\textsuperscript{6,7}, pressure engineering\textsuperscript{8-10} and chemical composition design\textsuperscript{11-22}. Chemical composition design has been regarded as a promising way to compensate the bulk charge in TIs\textsuperscript{23-26}. Besides, due to the effective manipulation of the strength of SOC, chemical composition design can drive the 3D TIs through a QPT\textsuperscript{11-20}.

Successive evolution of the electronic state has been realized when 3D TIs undergoes a QPT by chemical composition design\textsuperscript{11-20}. For example, Bi\textsubscript{2}Se\textsubscript{3} is a topological insulator with bulk energy gap of \( \sim 0.3 \) eV\textsuperscript{27-29}. In\textsubscript{2}Se\textsubscript{3} is a band insulator with energy gap of \( \sim 1.3 \) eV. When these two are mixed, various electronic states have been reported, such as a topological insulator, a Dirac semimetal and a band insulator. The dispersion relation of bulk band should become more linear when the system undergoes a QPT, which may result in an enhanced mobility of bulk carriers. Besides, the bulk band gap closes. Exotic electronic states were observed in the angle-resolved photoemission spectroscopy (ARPES) spectrum, which may offer a unique platform for future electronic devices. However, the magnetotransport properties of (Bi\textsubscript{1-x}In\textsubscript{x})\textsubscript{2}Se\textsubscript{3} have been rarely reported.

In this work, we synthesized high-quality QPT system (Bi\textsubscript{0.92}In\textsubscript{0.08})\textsubscript{2}Se\textsubscript{3} by a simple melting approach. We investigated the magnetotransport properties of the single crystalline sample under a magnetic field up to 13 T. The average mobility of \( \sim 1000 \) cm\textsuperscript{2}V\textsuperscript{-1}s\textsuperscript{-1} was obtained from the Lorentz law at the low field (\(< 3 \) T). The quantum oscillations revealed a high mobility of \( 1.4 \times 10^4 \) cm\textsuperscript{2}V\textsuperscript{-1}s\textsuperscript{-1} at 2 K. The topological Dirac fermions were evident by the nontrivial Berry phase in the Landau Fan diagram.
2. Experimental methods

Single crystals were grown by the melting high-purity (99.99%) elements of Bi, In, and Se at 850 °C with the designed doping concentration for one day in evacuated quartz tubes, and annealed at 850 °C for two days followed by cooling to room temperature for four days. Then, we obtain millimeter-sized samples by mechanical exfoliation with a blade. The thickness of the samples is normally a few micrometers. The structure was characterized by X-ray diffraction (XRD) measurements using a Cu Kα line (Rigaku Ultima III) with 2θ scanned from 5° to 80°. Before transport measurements, four-probe contacts were made by depositing the silver paste on the samples. The transport measurements between 2 and 50 K were conducted in a Quantum Design physical properties measurement system (PPMS) which can sweep the magnetic field between ± 14 T. A typical measurement current of 5 mA was applied for a high signal-to-noise ratio.

3. Results and discussion

In Figure 1(a), we show a profile of the furnace temperature during the growth procedure. The as-prepared crystal can be easily cleaved to the sheets of several millimeters, as shown in the inset of Figure 1(a). The (Bi0.92In0.08)2Se3 shares a common rhombohedral structure with that of Bi2Se3, as revealed by XRD [Figure 1(b)] and Raman spectrum [Figure 1(c)]. Only sharp (00l) reflections are observed in XRD patterns, confirming the excellent crystalline quality of our single crystals. In a typical Raman spectrum taken from a single crystal, three characteristic peaks are found at the position of 71 cm⁻¹, 131 cm⁻¹, and 175 cm⁻¹, which are related to three vibrational modes of $A_{1g}^1$, $E_g^1$, and $A_{1g}^2$, respectively. Therefore, the crystal structure maintains a rhombohedral phase with excellent crystallinity after the doping of In into Bi2Se3. Figure 1(d) shows the temperature-dependent resistance curve of the crystal, which suggests a metallic behavior.
Figure 1. The preparation and structural characterization of (Bi$_{0.92}$In$_{0.08}$)$_2$Se$_3$ single crystals. (a) The temperature profile of the furnace temperature during the growth procedure. The inset is the refined (Bi$_{0.92}$In$_{0.08}$)$_2$Se$_3$ crystals. (b) The XRD patterns of a single crystal. (c) The Raman spectrum of a single crystal. (d) The temperature dependence of the resistance.

Figure 2(a) shows the magnetoresistance (MR) curves measured at $\theta = 90^\circ$ under 2 K and 50 K. As temperature increases from 2 to 50 K, the MR in low magnetic fields ($< 3$ T) is almost unchanged but Shubnikov-de Hass (SdH) oscillations in high magnetic fields ($> 5$ T) seem to disappear at $T = 50$ K. We note that the MR curves show a quadratic magnetic field dependence in low magnetic fields ($< 3$ T). As shown in Figure 2(b), the MR curves at $T = 2$ to 50 K can be well fitted by parabolas within a small magnetic field range. Such a parabolic MR is believed to arise from the Lorentz deflection of carriers and the fitting allows us to deduce the carrier mobility ($\mu$), by $R(B) = R_0[1 + (\mu B)^2]$ $^{33,34}$. The obtained mobility of our sample at different temperatures is shown in Figure 2(c). Below 50 K, the carrier density is almost a constant. At $T = 2$ K, the average mobility reaches 1000 cm$^2$V$^{-1}$s$^{-1}$. On the basis of Drude model of electric conduction, the carrier density ($n$) of our samples can be derived by $n = 1/(e\rho\mu)$, where $e$ is the electron charge, $\rho$ is the resistivity and $\mu$ is the
carrier mobility\(^{33,34}\). Figure 2(d) shows the obtained carrier density as a function of temperature. Below 50 K, the carrier density is almost a constant.

SdH oscillations can be obviously seen from the MR difference ($\Delta R$) versus inverse magnetic field ($1/B$) curve after a smooth subtraction of background in Figure 3(a). The Landau level (LL) fan diagram is plotted in Figure 3(b). The maxima of $\Delta R$ are assigned to be the integer indices. The intercept value of -0.618 indicates the nontrivial $\pi$ Berry phase which can be observed in topological phase of matter, evidencing the Dirac surface state\(^{35}\). Fast Fourier Transformation (FFT) of SdH oscillations shows a single peak around 51 T in the inset of Figure 3(b). By using the semi-classical Onsager relation, the cross-sectional area $S_F = 5.12 \times 10^{-3}$ Å\(^{-2}\) of the Fermi surface is obtained. Assuming a circular cross section, a small Fermi momentum $k_F = 0.0404$ Å\(^{-1}\) can be extracted. To extract more transport parameters of the SdH oscillations, we fit the temperature-dependent SdH oscillation amplitude at different
indices according to the Lifshitz-Kosevich (LK) theory: \[ \Delta R(T)/R(0) = \lambda(T)/\sinh(\lambda(T)), \]
where \( \lambda(T) = 2n^2k_BT_m/\hbar eB \), \( m_c \) is the cyclotron mass, \( \hbar \) is the reduced Planck constant, and \( k_B \) is Boltzmann constant\(^{36,37} \). The cyclotron mass \( m_c \) is extracted to be 0.07 \( m_e \) shown in Figure 3(c). Then we can calculate the Fermi velocity and the Fermi level position to be \( v_F = \hbar k_F/m_c = 6.34 \times 10^5 \) m/s and \( E_F = m_c v_F^2 = 160 \) meV. Dingle temperature \( T_D = 2.06 \) K and surface carrier lifetime \( \tau = 5.6 \times 10^{-13} \) s are extracted from the field dependence of the amplitude of quantum oscillations at fixed temperatures in Figure 3(d). We calculate the \( \mu_{SdH} = e\tau/m_c = 1.4 \times 10^4 \) cm\(^2\)V\(^{-1}\)s\(^{-1}\) and \( l = v_F \tau = 355 \) nm.

\[ \text{Figure 3. The SdH oscillations in a single crystal. (a) The SdH oscillations amplitude } \Delta R \text{ as a function of } 1/B \text{ at different temperatures after subtracting the smooth background. (b) The plot of LL index } n \text{ versus } 1/B. \text{ The intercept is -0.618 by taking the maximum and minimum of } \Delta R \text{ as the integer and half integer, respectively, indicating the Dirac phase. (c) The temperature dependence of } \Delta R \text{ of different LLs, giving the } m_c = 0.07 \text{ } m_e. \text{ (d) The Dingle plots giving the quantum lifetime of } 5.6 \times 10^{-13} \text{ s and Dinge temperature of } 2.06 \text{ K.} \]

4. Conclusions

In conclusion, (Bi\(_{0.92}\)In\(_{0.08}\))\(_2\)Se\(_3\) single crystals have been synthesized through the melting process. The good crystallinity is characterized by XRD and Raman
spectroscopy. The analysis of magnetotransport measurements at the low field (< 3 T) reveals the average mobility of ~1000 cm²V⁻¹s⁻¹. Magnetotransport measurements at the high field (> 5 T) show the SdH oscillations, manifesting the nontrivial transport. Our results are helpful for understanding the magnetotransport properties of the QPT system.

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