

Disorder-induced linear magnetoresistance in (221) topological insulator Bi_2Se_3 films

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A linear magnetoresistance (LMR) with strong temperature dependence and peculiar non-symmetry with respect to the applied magnetic field is observed in high-index (221) Bi_2Se_3 films. Different from the LMR observed in the previous studies which emphasize the role of gapless linear energy dispersion, this LMR is of disorder origin and possibly arises from the electron surface accumulation layer of the film. Besides, an abnormal negative magneto-resistance that shows a non-monotonic temperature dependence and persists even at high temperatures and in strong magnetic fields is also observed. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4816078>]

A three-dimensional (3D) topological insulator (TI) is a new state of quantum matter characterized by gapless topological surface states (SSs) crossing the bulk band gap.^{1,2} Angle-resolved photoemission spectroscopy measurement has clearly identified bismuth compounds Bi_2Se_3 and Bi_2Te_3 as 3D TIs, with a single spin-helical Dirac cone on each surface.^{3,4} Due to the exotic Dirac fermion physics of such SSs, TIs are of great importance in both condensed matter physics and technological applications.

Transport studies of TIs have revealed many interesting quantum phenomena associated with the topological SSs, such as the Aharonov-Bohm oscillation in Bi_2Se_3 nanoribbons,⁵ the weak anti-localization (WAL) in Bi_2Se_3 and Bi_2Te_3 thin films,^{6–8} and the two-dimensional (2D) SdH oscillations in Bi_2Te_3 .⁹ Recently, a non-saturating linear magnetoresistance (LMR) has also been observed in TIs.^{10–18} The transport measurement of Bi_2Se_3 nanoribbons in tilted magnetic fields reveals the 2D nature of the LMR.¹⁰ This LMR can be enhanced by tuning the Fermi level into the bulk band gap of Bi_2Se_3 or $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3$ using a gate voltage.^{11,12} Furthermore, the tunneling between the top and bottom SSs in ultrathin TI films could cause the LMR to disappear.¹³ All these results seem to indicate that the observed LMR in TIs is a possible transport signature of topological SSs. Although several quantum^{17–20} or classical^{21,22} mechanisms have been proposed to explain the LMR, its physical origin in TIs is still undetermined. Previous transport measurements were mainly obtained from the (111) surface of TIs. But since the Fermi surface of other high-index SSs is elliptical,²³ it has been predicted theoretically that such anisotropic surface Dirac fermion would exhibit interesting refractive transport phenomena.²⁴ It is therefore of scientific interest to investigate the transport properties of high-index surfaces of TIs.

In this work, magneto-transport properties of (221) Bi_2Se_3 thin films are systematically investigated. In

particular, a quasi-2D LMR is revealed in these high-index films. This LMR is greatly enhanced at higher temperatures and exhibits peculiar non-symmetry with respect to the applied magnetic field. Detailed analysis further indicates that this LMR is of disorder origin and possibly arises from an electron surface accumulation layer of the (221) TI film. Besides this LMR, an abnormal negative MR (NMR) that exhibits a non-monotonic temperature dependence and persists even at high temperatures and in high magnetic fields is also observed. Our work offers more insight into magneto-transport in TIs, especially the physical origin of LMR, which might find potential application in magnetic sensors.

The (221) Bi_2Se_3 thin films with a thickness (t) of 200 nm were grown on specially prepared (001) InP substrates by molecular beam epitaxy.²³ Different from the previous (111) Bi_2Se_3 films, the quintuple layers (QLs) of (221) films form an angle of about 54.7° with the substrate surface, as shown schematically in the inset of Fig. 1(a). The QLs and the (221) crystal plane intersect in the $[1\bar{1}0]$ direction. As in our previous study of the same (221) Bi_2Se_3 films,²³ L-shaped Hall bar devices were fabricated by photolithography. Such an L-shaped device consists of two connected Hall bars aligned in the $[11\bar{4}]$ and $[1\bar{1}0]$ orthogonal directions. We can thus investigate the transport properties of the films in two different measurement configurations, i.e., the current (I) can be transverse or parallel to QLs, as indicated in the inset of Fig. 1(b). A magneto-transport study of the devices was conducted in a Quantum Design 14T PPMS system. As similar MR behaviors were observed in the two measurement configurations, only the results obtained with I being parallel to the QLs are presented in the following.

Fig. 1 shows the MR of the (221) films at different temperatures (T) in a perpendicular magnetic field (B). At $T = 2$ K, the MR shows a distinctive dip in resistance in low magnetic fields, indicating the occurrence of the WAL effect in the TI films.^{6–8} As temperature increases, the WAL gradually weakens, while a NMR emerges in moderate magnetic fields. At $T = 65$ K, the WAL disappears entirely. With temperatures further increased, a high-field positive MR is

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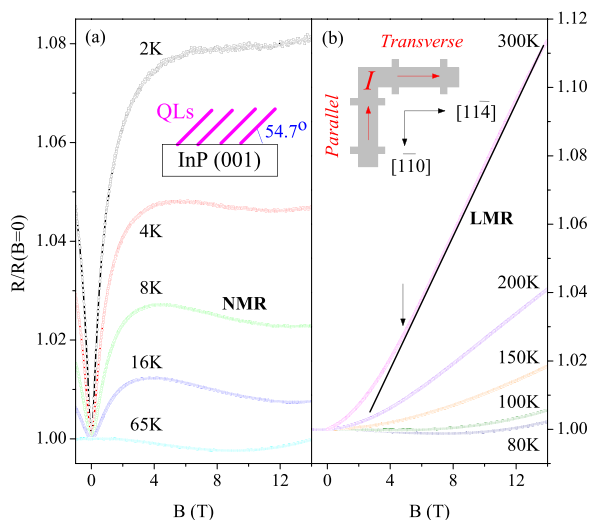


FIG. 1. Magnetoresistance measured at different temperatures in a perpendicular magnetic field. Inset: (a) Schematic drawing of inclined quintuple layers with respect to the (001) InP substrate; (b) Geometry of an L-shaped Hall bar.

enhanced and gradually extends to lower magnetic fields at the expense of the NMR. The positive MR finally evolves into a prominent non-saturating LMR at high temperatures, as shown in Fig. 1(b).

To gain more insight into the LMR observed at high temperatures, we investigated its dependence on magnetic field directions. As illustrated in Fig. 2, the tilting angle θ is defined as the angle between the film surface normal [221] and the magnetic field direction. The magnetic field is perpendicular (or parallel) to Bi_2Se_3 films when $\theta = 0^\circ$ (or 90°). Fig. 2 shows the MR measured with $\theta = 0^\circ$ and that measured with $\theta = 90^\circ$ at $T = 300\text{ K}$. It can be seen that the LMR is greatly suppressed when the magnetic field is in plane, indicating that the LMR depends mainly on the normal component of B and is of quasi-2D nature. The inset of Fig. 2 also shows the angular dependence of LMR at $B = 14\text{ T}$. Different from those in the previous LMR studies,^{10,13} this angular dependence cannot be described by a simple $\cos 2\theta$ function. The MR measured with $\theta = 0^\circ$ and that measured with $\theta = 180^\circ$ differ by ΔR as indicated in the inset. This shows that reversing the field direction will give a different MR, i.e., the observed LMR is not an even function of B .

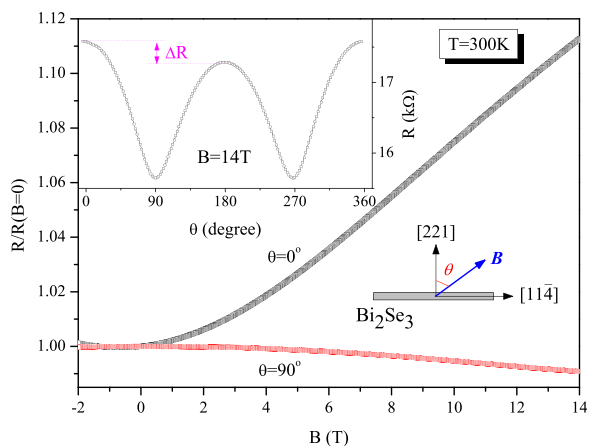


FIG. 2. Magnetoresistance measured at $T = 300\text{ K}$ but at two different tilting angles of the magnetic field. The definition of the tilting angle is illustrated in the figure. Inset: angular dependence of the MR at $T = 300\text{ K}$ and $B = 14\text{ T}$.

The results shown in Figs. 1 and 2 indicate that the LMR observed in our (221) Bi_2Se_3 films is very different from the LMR in the previous studies of TIs,^{10–14,17,18} where a weak temperature dependence is usually observed and the angular dependence can be fitted quite well by a $\cos 2\theta$ function. One can also infer from the results that this LMR arises from the high-mobility channel revealed in our previous study of the same (221) Bi_2Se_3 films.²³ It has been shown that a high-mobility conducting channel (mobility = $500\text{--}1000\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$) coexists with the ordinary bulk one (mobility = $\sim 60\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$) at high temperatures in the (221) thin films.²³ As T decreases, the carrier density of the high-mobility channel decreases significantly. Below 85 K , free carriers of the extra channel are frozen out, leaving the bulk channel the only conducting in the film. Since the LMR is only observed above 80 K and becomes more evident at higher temperatures, it is very likely attributed to the high-mobility channel. This is also consistent with the tilted magnetic field measurement in Fig. 2, which reveals the quasi-2D character of the LMR and thus rules out the possible role of the bulk channel.

LMR has been studied in several TIs,^{10–18} but its physical origin is still under investigation. According to the theory by Abrikosov,¹⁹ LMR appears in gapless semiconductors with linear energy dispersion under the quantum limit condition, i.e., the applied field must be so strong that only the lowest Landau level is populated. This quantum LMR model also predicts the n^{-2} dependence of LMR, where n is the free carrier density. In contrast to the quantum LMR model, Wang and Lei's recent theoretical study points out that LMR could also arise in 2D systems with linear energy dispersion and a non-zero g factor in the presence of overlapping Landau levels.²⁰ Their model predicts that the LMR is proportional to n^{-1} . But in our work, the LMR is observed at high temperatures, precluding the possibility of Landau level formation in the (221) films. Besides, the LMR is enhanced with higher carrier density, since our previous study has shown that the carrier density of the high-mobility channel related to LMR increases rapidly with temperature. This is distinctively different from the carrier density dependence of LMR predicted by Abrikosov and Wang and Lei. Therefore, the LMR observed here falls outside the scope of the two models. In recent transport studies of (111) Bi_2Te_3 and $\text{Bi}_2\text{Te}_2\text{Se}$ thin films, a LMR was observed at low temperatures and ascribed to WAL.^{17,18} But as Fig. 1 shows clearly, WAL disappears in our (221) Bi_2Se_3 films by the time the temperature reaches 65 K , so this WAL model is also not applicable to our observations.

Besides the above quantum models, Parish and Littlewood proposed that inhomogeneity or mobility disorder is crucial to the explanation of LMR observed in polycrystalline silver chalcogenides.²¹ In this classical model, the LMR results from the admixture of Hall signals due to disorder. A recent Monte Carlo simulation further investigated the microscopic nature of this classical LMR. It is the multiple electron scattering by low-mobility islands in an inhomogeneous conductor that gives rise to the disorder-induced LMR.²² As free carriers are frozen out of the LMR-related high-mobility channel at low temperatures,²³ disorder is considered to play an important role in our (221) TI films.

Structure analysis of the films also reveals the surface roughness of, or even the presence of rotational domains in, these (221) Bi_2Se_3 films which are grown on rough and faceted InP substrates.²³ Furthermore, as the tilted magnetic field measurements show, the observed LMR is not an even function of B , suggesting the possible admixture of Hall signals in the MR measurement due to disorder. Therefore, it is believed at the present stage that the LMR observed in (221) Bi_2Se_3 films is likely of disorder origin, as proposed by the Parish-Littlewood model. According to this model, we can further estimate the width of the mobility disorder $\Delta\mu$ in our (221) films, as the inverse of $\Delta\mu$ gives the crossover field to LMR. At $T = 300$ K, the LMR is only observed above 5 T, as marked by a downward arrow in Fig. 1(b). Therefore, the corresponding $\Delta\mu$ is about $2000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at 300 K. But this is only a rough estimate of $\Delta\mu$, since both the high-mobility and bulk channels coexist at $T = 300$ K. The Parish-Littlewood model might also be applicable to a recent observation of LMR in polycrystalline Bi_2Te_3 films¹⁵ and of giant anisotropic LMR in Bi_2Te_3 bulk single crystals.¹⁶ In the latter case, the observed LMR differs much in opposite magnetic field directions, indicating the possible admixture of odd Hall signals.

So where does the high-mobility channel come from? As the observed LMR arises from disorder and the gapless linear energy dispersion is not a prerequisite for its occurrence, the high-mobility channel related to LMR is thus unlikely to originate from topological SSs. Considering the carrier freeze-out at low temperatures and the quasi-2D nature of the observed LMR, the high-mobility channel is more likely to be from a surface accumulation layer. Note that this channel cannot come from the impurity band either, since its mobility is much higher than that of the bulk channel.²³

Besides the high-field LMR observed at high temperatures, the Bi_2Se_3 (221) films also exhibit NMR as shown in Fig. 1. Although this NMR begins to appear in intermediate field at $T = 4$ K, it actually originates in low magnetic fields. This can be seen from the MR obtained at $T = 80$ K and $\theta = 0^\circ$ (perpendicular magnetic field) shown in Fig. 3(a). With the disappearance of WAL at $T = 80$ K, the MR in the film is completely dominated by NMR in low magnetic field. In order to determine the origin of NMR, we also measured the MR at $T = 80$ K but with $\theta = 90^\circ$ (in-plane magnetic field). It is clear from Fig. 3(a) that the NMR is weakly dependent on the tilting angle θ , indicating the bulk nature of the observed NMR. As Fig. 1 shows, the LMR dominates the NMR at high temperatures in the perpendicular magnetic field. To study the temperature dependence of NMR, we thus measured the high-temperature NMR in an in-plane magnetic field ($\theta = 90^\circ$). As can be seen in Fig. 3(b), the NMR initially weakens as temperature increases from 80 K to 150 K, but it is actually enhanced as temperature rises further. Therefore, the NMR displays a non-monotonic temperature dependence. Note also that at $T = 300$ K, the NMR persists even in a magnetic field of up to 14 T.

At present, the physical origin of this abnormal NMR is still unclear, but it is unlikely identical to the NMR arising from weak localization in magnetically doped TI²⁵ or to that observed in ultra-thin TI films.¹³ NMR is also observed in

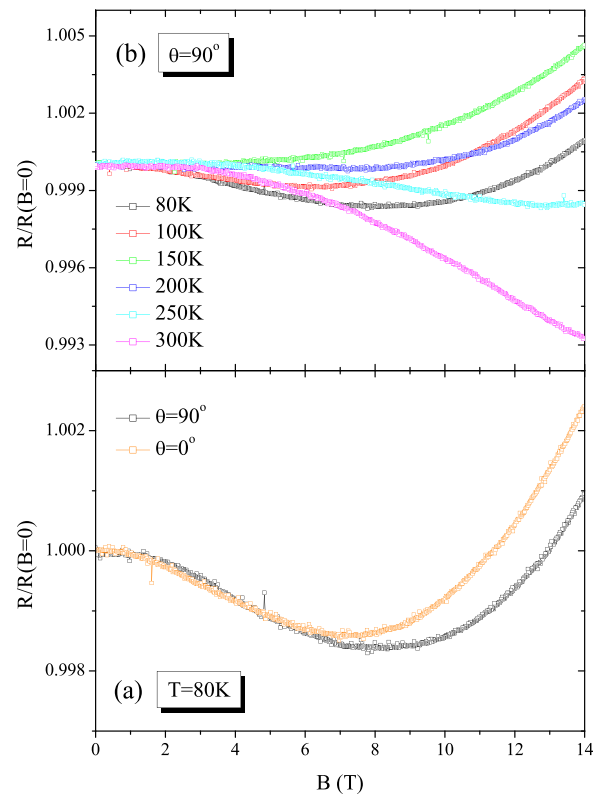


FIG. 3. (a) Magnetoresistance measured at $T = 80$ K with two different tilting angles of 0° and 90° . (b) Magnetoresistance measured at $\theta = 90^\circ$ with different temperatures.

multilayer massless Dirac Fermion systems such as α -(BEDT-TTF)₂I₃ and graphite.^{26,27} In such systems, the vertical transport is governed by tunneling between adjacent weakly coupled layers. In a perpendicular magnetic field, an interlayer NMR would arise due to the increase in carrier density in the zero-mode Landau level of each layer. But as shown in Fig. 3, the NMR in our (221) TI films is of bulk nature, irrespective of the field direction. It can also be observed in low magnetic fields and even at high temperatures, thus, ruling out the formation of Landau levels in the films. Hence the above scenario is not applicable to the abnormal NMR we observed. More work need to be done to reveal the origin of this abnormal NMR in (221) films.

In conclusion, the magneto-transport study of (221) Bi_2Se_3 films has revealed the presence of a quasi-2D LMR in these high-index TI films. This observed LMR has a strong temperature dependence and is not symmetric with respect to the applied magnetic field. Unlike the LMR observed in the previous studies where gapless linear energy dispersion is believed crucial to the observation of LMR, this LMR is more likely of disorder origin. An abnormal NMR which has a non-monotonic temperature dependence and is still observable at high temperatures and in high magnetic fields has also been studied systematically. Following this work, it would be interesting to study the real transport properties of (221) topological SSs in the future by further improving the quality of the (221) Bi_2Se_3 films.

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