High-field linear magneto-resistance in topological insulator Bi$_2$Se$_3$ thin films

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Linear magneto-resistance is observed in high magnetic field in topological insulator Bi$_2$Se$_3$ films. As revealed by tilted magnetic field measurement, this linear magneto-resistance is associated with the gapless topological surface states and of quantum origin. In the ultra-thin limit, the inter-surface tunneling induced surface state gap opening quenches the linear magneto-resistance. Instead, weak negative magneto-resistance is observed in high magnetic fields in ultra-thin films. © *2012 American Institute of Physics.* [doi:10.1063/1.3677669]

Three dimensional topological insulators (TIs) are band insulators but with gapless surface states crossing the bulk band gap.[1,2] As protected by time reversal symmetry, these surface states consist of odd number of Dirac cones on the surface of TIs. Angle-resolved photoemission spectroscopy (ARPES) studies have confirmed the existence of such topological surface states in Bi$_2$Se$_3$ and Bi$_2$Te$_3$ with only one massless Dirac cone on each surface.[3,4] Various quantum transport phenomena associated with these surface states have been revealed in relevant magneto-transport studies such as quantum conductance fluctuation,[5] Aharonov-Bohm oscillations,[6] and weak antilocalization.[7–9] Besides these, a high-field non-saturating linear magneto-resistance (LMR) was recently observed in 25-nm-thick Bi$_2$Se$_3$ nanoribbons, which was also believed to be of quantum origin and attributed to the topological surface states.[10] Previous ARPES study of Bi$_2$Se$_3$ film has shown that in the ultra-thin limit (<6 nm), the inter-surface coupling would give rise to a surface state gap at the Dirac point,[11] greatly suppressing the weak anti-localization effect in low magnetic fields.[12] However, the quantum LMR has not been studied in this ultra-thin limit so far. Besides this, the investigation of this LMR in TIs is also of potential importance in the field of magnetic field sensor technology.[13] In this letter, we study the LMR in a series of Bi$_2$Se$_3$ films with different thickness. We first show the observation of LMR in 8-nm-thick Bi$_2$Se$_3$ films which is of 2D nature, consistent with previous study of Bi$_2$Se$_3$ nano-ribbons.[10] But, as the film thickness is reduced below 6 nm, the high-field LMR disappears. A weak negative magneto-resistance (MR) is observed in high magnetic fields instead of the LMR in these ultra-thin films. We associate the disappearance of LMR with the surface state gap opening induced by inter-surface coupling in the ultra-thin limit.

The Bi$_2$Se$_3$ films were grown by molecular beam epitaxy on (111) silicon wafers using In$_2$Se$_3$ buffer.[14,15] Hall measurement at 2 K shows that the electron densities of these films range between 1.6 and 2.7E19 cm^{-3}. Standard Hall bar devices were fabricated from these films with the bar dimension of 200 μm long and 100 μm wide. MR of these devices

was then measured at low temperatures (T) in a Quantum Design 14 T physical property measurement system (PPMS) with a rotational sample holder, which allows for the variation of the tilting angle (θ) of the magnetic field (B) respective to the film plane.

Fig. 1 shows the MR curves of an 8-nm-thick Bi$_2$Se$_3$ film at $T = 2$ K and different tilting angles (θ) of magnetic fields. When the magnetic field is perpendicular to the film,
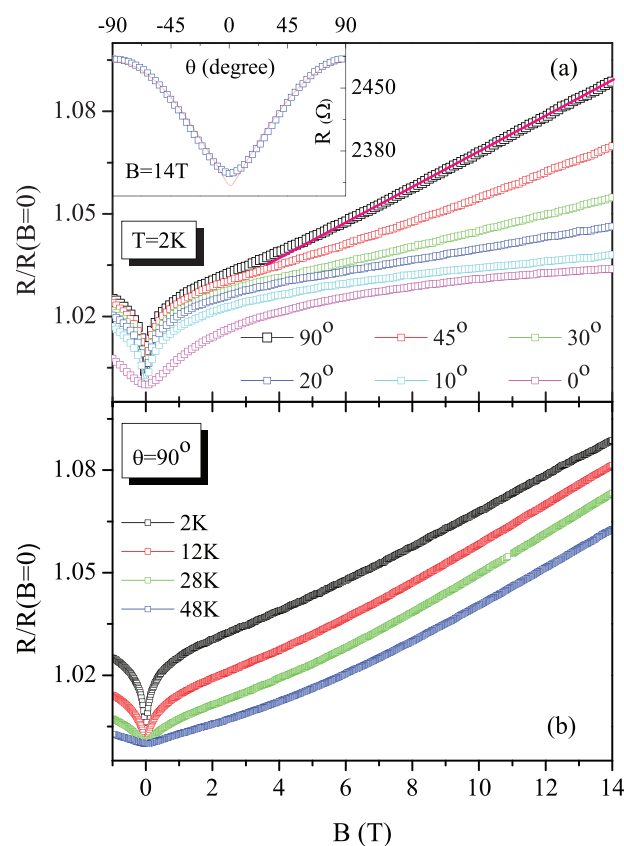

FIG. 1. (Color online) (a) Magneto-resistance of 8 nm Bi$_2$Se$_3$ films measured at $T = 2$ K with different tilting angles indicated. Solid line in the $\theta = 90°$ curve is a linear fit. Inset: measured angular dependence of magneto-resistance obtained with $B = 14$ T and $T = 2$ K (symbols) and a $|\sin(\theta)|$ function (solid line). (b) Magneto-resistance of 8 nm Bi$_2$Se$_3$ films measured at $\theta = 90°$ with different temperatures indicated.

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i.e., $\theta = 90°$, a linear and non-saturating magneto-resistance is clearly observed in high B fields ($B > 7\,\text{T}$), as indicated by the straight line in Fig. 1(a). With decreasing the tilting angle, this LMR weakens. When the B field is in plane ($\theta = 0°$), the LMR is greatly suppressed, as shown in Fig. 1(a). The inset in Fig. 1(a) shows the tilting angle dependence of MR obtained at $T = 2\,\text{K}$ and $B = 14\,\text{T}$. As shown by the solid fitting curve in the inset, this angular dependence can be well fitted by a $|\sin(\theta)|$ function. It shows that the LMR only depends on the normal component of magnetic fields, i.e., $B|\sin(\theta)|$, indicating the 2D nature of this LMR. Besides this high field LMR, weak antilocalization (WAL) is also observed at low B fields, which decreases with decreasing the tilting angle. Our previous study of WAL in tilted B field in Bi_2Te_3 has shown that this low field WAL arises from the π Berry phase of topological surface states.[8] Therefore, the 2D LMR observed in high fields is also most likely associated with the topological surface states. This is in good agreement with a recent transport study of Bi_2Se_3 nanoribbons, where the high field LMR is also observed and ascribed to topological surface states.[10] It is worth noting that the 2D nature of the LMR does not likely arise from the anisotropic bulk transport of the Bi_2Se_3 films with layer-by-layer crystal structure. In our previous study of WAL in tilted B field in 50 nm Bi_2Te_3, we have observed WAL even with the magnetic field in plane. This not only reveals the bulk contribution to the WAL but also indicates the 3D nature of the bulk transport.[8] We also studied the temperature dependence of the LMR. Fig. 1(b) shows the MR data of the 8 nm film measured at $\theta = 90°$ with different temperatures as indicated. With temperatures increasing from 2 to 48 K, the slope of the high-field LMR remains unchanged. This is in sharp contrast to the low B field WAL, which decreases and disappears quickly with increasing temperatures.[8]

According to the theory by Abriskov,[16] a quantum LMR would be expected to occur in the quantum limit in gapless semiconductor with linear energy spectrum, such as the surface states of topological insulators we studied here. The quantum limit requires that the applied magnetic field should be so large that only one Landau level is populated with electrons. But, later experimental studies revealed that the quantum LMR could appear in relatively smaller B field, with a few Landau levels populated.[13] The STM study of Landau quantization in 50-quintuple-layer Bi_2Se_3 film with surface carrier density of $2 \sim 3\text{E}12\,\text{cm}^{-2}$ (or bulk density of $4 \sim 6\,\text{E}19\,\text{cm}^{-3}$) at $T = 4.2\,\text{K}$ shows that the formation of surface-state Landau levels can be clearly observed with the B field above 6 T.[17] Considering the comparable carrier density of our films ($1.6 \sim 2.7\text{E}19\,\text{cm}^{-3}$), it is thus reasonable to ascribe the LMR observed with $B > 7\,\text{T}$ in our Bi_2Se_3 films to the quantum origin proposed by Abriskov. Furthermore, the theory also predicts weak temperature dependence of the quantum LMR. This is in good agreement with our experimental observations as shown in Fig. 1(b).

We further investigated this quantum LMR in the ultra-thin limit. Fig. 2(a) shows the MR of a series of ultra-thin Bi_2Se_3 films at $\theta = 90°$ and $T = 2\,\text{K}$ with the film thickness (t) as indicated. For comparison, the MR and linear fit at high fields of the 8-nm-thick sample are also shown in Fig. 2(a). It can be clearly seen that the high-field LMR disap-
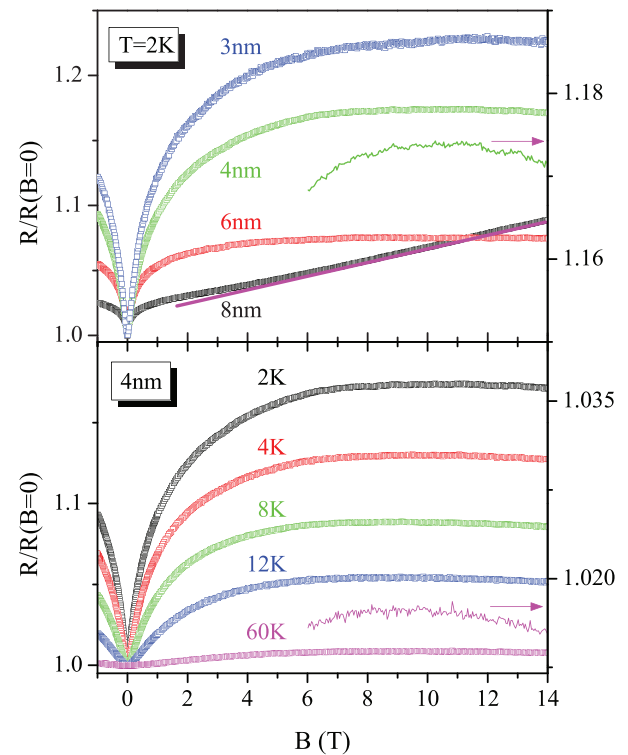

FIG. 2. (Color online) (a) Magneto-resistance of ultra-thin Bi_2Se_3 films measured with magnetic field applied perpendicularly to the film and $T = 2\,\text{K}$. Also shown is the enlarged view of the high-field MR data of the 4 nm film. (b) Magneto-resistance of the 4 nm film obtained with $\theta = 90°$ at different temperatures as indicated. The enlarged view of the high-field MR data measured at $T = 60\,\text{K}$ is also shown.

pears in these ultra-thin films with $t = 6$, 4, and 3 nm, respectively. The disappearance of LMR is likely associated with the gap opening of the surface states at the Dirac point in the ultra-thin limit.[11] Quantum LMR has been investigated in Ag-rich or Ag-deficient silver chalcogenides, the band structure of which could be altered by applying hydrostatic pressure.[18] It is found that the high-field LMR emerges when the band gap closes under certain pressure. This indicates gapless linear dispersion energy spectrum is essential to the observation of LMR in high fields. Due to the tunneling between the top and bottom surface states, a surface state gap would be opened at the Dirac point in ultra-thin Bi_2Se_3 films.[11] Therefore, decreasing the film thickness changes the energy spectrum of topological surface states from gapless in the 8-nm-thick film to gapped in ultra-thin films, giving rise to the disappearance of LMR as shown in Fig. 2(a). The above discussion also excludes the possible bulk contribution to the observed quantum LMR, since there is a bulk band gap for the bulk states. The change in the energy spectrum with film thickness also accounts for the variation of WAL with the film thickness in Fig. 2(a), as pointed out by a recent study of WAL in ultra-thin Bi_2Se_3 films.[12]

It is worth noting in Fig. 2(a) that instead of LMR, a rather weak negative MR is observed in high fields in Bi_2Se_3 films with $t = 4$ and 3 nm, respectively. For clarity, the MR data of the 4 nm film between B = 6 and 14 T are enlarged as shown in Fig. 2(a). Different from the 8 and 6 nm films where positive MR is observed in the entire field range, the 4 and 3 nm films exhibit positive WAL in low B fields but

negative MR in high *B* fields. As proposed by a recent localization theory of topological insulators, the competition between WAL from surface states and weak localization (WL) from bulk channels might give rise to the change of MR from positive in low *B* fields to negative in high *B* fields.[19] This WL-induced negative MR would be expected to occur at low temperatures but disappear at high temperatures. Fig. 2(b) shows the MR of the 4 nm film measured at $\theta = 90°$ with different temperatures indicated. As *T* is increased from 2 to 60 K, the WAL in low *B* fields totally disappears as expected.[8] On the contrary, the negative MR in high *B* fields exhibits weak temperature dependence and still can be observed at high temperatures, as indicated by the enlarged view of the MR data between 6 and 14 T at $T = 60$ K. As a result, the persistence of negative MR at high temperatures seems to exclude the WL contribution from bulk channels. This negative MR is also not likely associated with electron-electron interaction which has been reported of importance in ultra-thin Bi_2Se_3 films and gives rise to positive MR at low temperatures.[12,20] More theoretical or experimental work in the future is needed to clarify the physical origin of this high-field weak negative MR observed in ultra-thin Bi_2Se_3 films.

In conclusion, we have studied the linear magnetoresistance in Bi_2Se_3 films with different thickness in high magnetic fields. The LMR observed in thick Bi_2Se_3 films is believed to be of quantum origin and associated with gapless energy spectrum of surface Dirac fermions. The disappearance of LMR in ultra-thin films further reveals the gap opening of the surface states due to the inter-surface tunneling.

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