Electric- and magnetic-field-controlled topological quantum phase transitions in defect-suppressed Bi$_2$Se$_3$ films

Nikesh Koirala$^{1+\ast}$, Maryam Salehi$^2$, Jisoo Moon$^1$, Seongshik Oh$^{1\ast}$

$^1$Department of Physics & Astronomy, Rutgers, The State University of New Jersey, Piscataway, New Jersey 08854, U.S.A.

$^2$Department of Materials Science and Engineering, Rutgers, The State University of New Jersey, Piscataway, New Jersey 08854, U.S.A.

$^\ast$Correspondence should be addressed to nikes@mit.edu and ohsean@physics.rutgers.edu

$^\dagger$Present Address: Department of Physics, Massachusetts Institute of Technology, Cambridge, MA 02139 U.S.A.

Despite many years of efforts, attempts to reach the quantum regime of topological surface states (TSS) on an electrically tunable topological insulator (TI) platform have so far failed on binary TI compounds such as Bi$_2$Se$_3$ due to high density of interfacial defects. Here, utilizing an optimal buffer layer on a gatable substrate, we demonstrate the first electrically tunable quantum Hall effects (QHE) on TSS of Bi$_2$Se$_3$. On the $n$-side, well-defined QHE shows up, but it diminishes near the charge neutrality point (CNP) and completely disappears on the $p$-side. Furthermore, around the CNP the system transitions from a metallic to a highly resistive state as the magnetic field is increased, whose temperature dependence indicates presence of an insulating ground state at high magnetic fields.

Key words: Bi$_2$Se$_3$, quantum Hall effect, topological insulator, electric field effect, charge neutrality point
TSS provides a rich playground for a number of topological quantum effects such as topological magnetoelectric effect, Majorana fermions and QHE\textsuperscript{1,2,3,4,5,6,7,8}. However, due to high level of surface Fermi level originating from unintended native dopants, it has been challenging to access the quantum regime of TSS. In particular, even if QHE is one of the most intensely studied phenomena in 2D systems, gate tuned studies of QHE of TIs has been so far limited only to ternary or quaternary compounds such as Bi\textsubscript{2-x}Sb\textsubscript{x}Te\textsubscript{3} and (Bi\textsubscript{x}Sb\textsubscript{2-x}Se\textsubscript{y}Te\textsubscript{3-y})\textsuperscript{4,5,9}. For the binary compounds such as the prototypical TI Bi\textsubscript{2}Se\textsubscript{3}, tracking evolution of QHE as a function of gate voltage has not been possible due to high density of bulk and interfacial defects\textsuperscript{10,11}. Recently, however, utilizing a structurally and chemically-matched buffer layer that solves the defect problem, QHE was observed in Bi\textsubscript{2}Se\textsubscript{3} thin films\textsuperscript{12,13}. Here, by adapting this buffer layer scheme to a gatable SrTiO\textsubscript{3}(111) substrate\textsuperscript{14}, we present the first gate-dependent study of QHE in Bi\textsubscript{2}Se\textsubscript{3}.

Low-carrier density Bi\textsubscript{2}Se\textsubscript{3} thin films were grown on an electrically insulating buffer layer, which comprises a heterostructure of 5 QL In\textsubscript{2}Se\textsubscript{3} – 4 QL (Bi\textsubscript{0.5}In\textsubscript{0.5})\textsubscript{2}Se\textsubscript{3} grown on SrTiO\textsubscript{3} (111) substrate following the recipe of ref. 9 (See Supporting Information). The films were then capped \textit{in situ} either by a 100 nm Se or a 50 nm MoO\textsubscript{3}/50 nm Se layer to protect against ambient contamination\textsuperscript{15}. Then, a ~100 nm thick Cu layer was deposited \textit{ex situ} on the back surface of SrTiO\textsubscript{3} substrate as a back gate. The films were then scratched into millimeter sized Hall bars using a metal mask and a tweezer, and indium leads were used to make electrical contacts (See Supporting Fig. S1).

On these Hall bar patterns, Hall (R\textsubscript{H}) and sheet resistance (R\textsubscript{S}) were initially measured at magnetic field (B) up to ± 0.6 T in a cryostat at T = 5 K. The measured raw data were symmetrized or anti-symmetrized to eliminate mixing of longitudinal and Hall resistances due to imperfection in the measurement geometry (See Supporting Information). Sheet carrier density (n\textsubscript{S}) = \[ e \cdot (dR_{H} \]
and mobility ($\mu$) = ($R_0 \cdot n_S \cdot e$)$^{-1}$ were then calculated, where $dR_H/dB$ is the slope of low field linear part of Hall resistance (unless otherwise stated), $e$ is the electronic charge and $R_0$ is the zero-field sheet resistance. These films have $n$-type carriers with $n_S \approx 5 \times 10^{12}$ cm$^{-2}$ and $\mu \approx 1,000$ – 3,000 cm$^2$V$^{-1}$s$^{-1}$ (See Supporting Fig. S2). Compared to the films grown directly on SrTiO$_3$(111), where $n_S$ is typically $\sim 4 \times 10^{13}$ cm$^{-2}$, the buffer-layer grown films exhibit an order of magnitude decrease in the defect density$^{16}$, which is consistent with our previous report$^{12}$. This low sheet carrier density obtained with the buffer layer was essential for reaching the quantum regime of TSS via gating as we present below.

In the rest of the Letter, we focus on the gate voltage dependence of $R_H$ and $R_S$ in MoO$_3$/Se-capped films. As reported previously, MoO$_3$ capping further reduces the $n$-type Fermi level of Bi$_2$Se$_3$ films toward the charge neutrality point (CNP)$^{12,17}$. We measured films of three different thicknesses: 8, 10 and 15 QL. 8 QL film was measured at $T = 1.5$ K and $B$ up to $\pm 9$ T and 10 QL and 15 QL films were measured at $T = 5$ K and $B$ up to $\pm 0.6$ T.
Gate-dependent magneto-transport data for Bi$_2$Se$_3$ films. a-c, Zero-field sheet resistance ($R_o$, upper panel) and sheet carrier density ($n_s$, lower panel) as a function of back-gate voltage, $V_G$, for 8, 10 and 15 QL films respectively. Solid black lines are a guide to the eye. The insets show magnetoresistance ($R_s$, upper panel) and corresponding Hall resistance ($R_H$, lower panel) as a function of magnetic field, $B$, taken at several representative back-gate voltage values from which $R_o$ and $n_s$ were extracted. Note the different magnetic field range and temperature for 8 QL compared to 10 and 15 QL films. Ambipolar behavior is observed in (a-b).

Figures 1a-c show $R_o$ (upper panel) and $n_s$ (lower panel) as a function of back-gate voltage ($V_G$) of 8, 10 and 15 QL films, respectively. In all three samples, the $n$-type carrier density is less than $3 \times 10^{12}$ cm$^{-2}$ at $V_G = 0$ V, which is well below the maximum carrier density ($\sim 1 \times 10^{13}$ cm$^{-2}$) required to make the bulk state of TIs insulating$^{18}$. For the 15 QL film, we were able to modulate $n_s$ from $2.8 \times 10^{12}$ cm$^{-2}$ to $1.5 \times 10^{12}$ cm$^{-2}$ and $R_o$ from 1.9 kΩ/sq to 3.0 kΩ/sq, as $V_G$ is tuned from 0 to -20 V. However, ambipolar transport was not observed in this film, presumably because 15 QL is too thick for its top surface to be electrostatically modulated by the bottom gating. For 8 and
10 QL films, $R_o$ increases with $V_G$, reaches a maximum value (for example at $V_G \approx -37$ V for 10 QL film) and then decreases with further increase in $V_G$. Concurrently, ($n$-type) $n_S$ decreases with $V_G$ and eventually changes to $p$-type (for example at $V_G \approx -45$ V for 10 QL film). Such an ambipolar behavior not only confirms the TSS conduction, but also the tunability of chemical potential across the CNP\textsuperscript{19}. Therefore, we focus on 10 QL films below and carry out more in-depth measurements up to much higher magnetic fields (34.5 T).

Figure 2 | QHE as a function of magnetic field at several gate voltage values for a 10 QL film. a-e, sheet resistance ($R_S$, upper panel) and Hall resistance ($R_H$, lower panel) up to $|B| = 34.5$ T at $V_G = 0$, -30, -40, -70 and -100 V respectively. Change in the sign of Hall effect and corresponding maxima in (f) zero field sheet resistance $R_o$ at $V_G \approx -70$ V indicates that $p$-type carriers dominate the transport for $V_G < -70$ V. For $n$-type carriers, $\nu = 1$ QHE is clearly observed at high magnetic fields. (g) Evolution of QHE with carrier
density, where QHE disappears when carriers change from $n$- to $p$-type. $p$-type carrier density for $V_G = -100$ V was estimated from the average slope of Hall effect in (e).

Figures 2a-e show $R_S$ (upper panel) and $R_H$ (lower panel) of another identically prepared 10 QL film as a function of $B$ up to ±34.5 T for various $V_G$ values at $T = 0.35$ K (See Supporting Fig. S3 and related text for more data and discussion). At low magnetic fields, magnitude of the negative slope of $R_H$ increases as $V_G$ changes from 0 to -40 V, indicating decreasing $n$-type carrier density from $1.5 \times 10^{12}$ cm$^{-2}$ to $5.9 \times 10^{11}$ cm$^{-2}$. At $V_G = -70$ V, $R_H$ fluctuates strongly around zero indicating mixed transport from electrons and holes and at $V_G = -100$ V the slope becomes positive albeit non-linear. The nonlinear Hall effect possibly indicates multi-carrier transport likely due to the effect of electron-hole puddles$^{20}$. However, the overall positive slope of $R_H$ indicates that conduction is now dominated by $p$-type carriers. Figure 2f shows corresponding change in $R_o$, where it increases till $V_G = -55$ V, shows a maximum at $-70 \leq V_G \leq -55$ V and then decreases below $V_G = -70$ V. This indicates that the film goes through CNP at $V_G \approx -70$ V.

At high magnetic fields, we observe increasingly developed dips in $R_S$ and plateaus at $h/e^2$ in $R_H$ for $-30 \leq V_G \leq 0$ V indicating $v = 1$ QH state. Additionally, we observe developing plateau-like features at $R_H \approx h/3e^2$ in the same voltage range, which is consistent with top and bottom surfaces having similar carrier density in this gate voltage range (See Supporting Information and Supporting Fig. S3). At $V_G = -40$ V both the dip in $R_S$ and the plateau in $R_H$ are less well developed than at $V_G = -30$ V indicating that the $v = 1$ QH state weakens as the Fermi level is lowered toward CNP. For $V_G = -70$ V (-100 V), QH signature is completely gone and $R_S$ increases monotonically with $B$ and reaches ~878 kΩ/sq (~90 kΩ/sq) at $B = 25$ T (32 T), corresponding to ~10,000% (~1000%) of magnetoresistance as defined by $\text{MR} = \frac{R_S(B) - R_S(0)}{R_S(0)} \times 100\%$. In Fig. 2g, we summarize our observation, where $v = 1$ QHE emerges with decreasing $n$-type carrier density until
$V_G \approx -30$ V, then diminishes while approaching CNP and gives way to a highly resistive state in the $p$-regime. We note that the $p$-type carrier density at $V_G = -100$ V in Fig. 2g was estimated by taking the average, rather than the low-field, slope of the Hall resistance.

In order to observe continuous evolution of transport with $V_G$, we have measured $R_S$ and $R_H$ as a function of $V_G$ at various magnetic fields $B$ from 0 to 44.5 T and various temperatures $T$ from 0.35 K to 9 K on another 10 QL thick film. Figures 3a and 3b show $R_S$ and $R_H$, respectively, at different magnetic fields and at $T = 0.35$ K (See Supporting Fig. S4 for data at additional $B$ fields). For all magnetic fields we observe a peak in $R_S$ and a change in the sign of $R_H$ from negative to positive at $-30 \, V \lesssim V_G \lesssim -26$ V, indicating that $p$-type carriers dominate the transport below these gate voltages. For higher magnetic fields ($B > 23$ T), we observe a developing dip in $R_S$ and a plateau in $R_H \approx -25.8 \, k\Omega$ for $-20 \, V \lesssim V_G \lesssim -15$ V indicative of $\nu = 1$ QH state for $n$-type carriers. Near CNP and for $p$-type carriers, we observe neither the dips in $R_S$ nor the plateaus in $R_H$ in magnetic field up to 44.5 T. Inset of Fig. 3a shows the magnetic field dependence of $R_S$ at CNP ($R_{CNP}$) and at $V_G = -76$ V ($R_{-76V}$, where $p$-type carriers dominate) indicating that $R_S$ increases monotonically with $B$ for both CNP and $p$-type carriers, reaching as high as $\sim 250 \, k\Omega$ (44 k$\Omega$) and $\sim 44 \, k\Omega$ (18 k$\Omega$) at $B = 44.5$ T (11.5 T), respectively, which correspond to $\sim 3,000\%$ (600%) and 650% (100%) of magnetoresistance.
Gate voltage dependent transport properties of a 10 QL film at $T = 0.35$ K and $B$ field up to 44.5 T. a, sheet resistance ($R_s$) and b, Hall resistance ($R_H$) at $T = 0.35$ K as a function of $V_G$ at several $B$ values from 0 to 44.5 T. Corresponding c, sheet ($\sigma_{XX}$) and d, Hall ($\sigma_{XY}$) conductance. $\nu = 1$ QHE is observed at $V_G \approx -15$ V to -20 V and non-saturating magnetoresistance ($MR$) = $\frac{R_s(B) - R_s(0)}{R_s(0)} \times 100\%$ with $B$ is observed for $V_G \lesssim -21$ V as plotted in inset of (a) for CNP and $V_G = -76$ V.

In order to get an additional perspective, we plot sheet conductance $\sigma_{XX} = R_s/(R_s^2 + R_H^2)$ and Hall conductance $\sigma_{XY} = R_H/(R_s^2 + R_H^2)$ in Fig. 3c and 3d, respectively. Consistent with resistance plots, we observe a $\sigma_{XY}$ plateau at $\sim h/e^2$ and a minimum in $\sigma_{XX}$ at $-20$ V $\lesssim V_G \lesssim -15$ V indicative of $\nu = 1$ QHE. Apart from a plateau-like feature at $\sigma_{XY} \approx 0$ and corresponding minimum in $\sigma_{XX}$ for $-35 \lesssim V_G \lesssim -30$ V, which can possibly indicate the $\nu = 0$ state, no features resembling QHE are observed in conductance plots for $V_G \lesssim -30$ V$^{4,5,9,21}$, implying that QHE is lost when $p$-type carriers dominate the transport. The lack of QHE on the $p$-side is consistent with both the recent transport result on compensation-doped Bi$_2$Se$_3$ films and the lack of Landau levels (LL) on the $p$-side of Bi$_2$Se$_3$ in scanning tunneling spectroscopy measurements$^{13,22,23}$. It can be explained
by the proximity of the Dirac point to the bulk valence band and the much broader surface band on the $p$-side of Bi$_2$Se$_3$\textsuperscript{24,25}. This is in marked contrast with Sb-based TIs, which exhibit QHE and LLs for both $n$- and $p$-sides due to relatively symmetric surface bands with a well-exposed Dirac point\textsuperscript{4,26,27,26}.

Next, we discuss temperature dependence of $R_S$ versus $V_G$ at different magnetic fields in order to understand the behavior of $p$-type carries in Bi$_2$Se$_3$. As shown in Fig. 4a, zero-field $R_S$ is $< 7.5$ kΩ/sq, which is much lower than the quantum resistance (25.8 kΩ), and increases only slightly (~4% at CNP) at $T = 0.35$ K compared to $T = 2$ K for all $V_G$. At higher temperatures ($T \approx 6$ to 12 K), we observe similarly small variation in $R_S$ with temperature in a different but identically prepared 10 QL film (See Supporting Fig. S5). In the inset of Fig. 4a we show $R_S$ versus $T$ at $V_G = 0$ V, where small upturn at low temperatures is observed, consistent with previous studies on TI films. In addition, we observe weak anti-localization at low fields for all $V_G$ values and in all samples (see Fig. 2 and 3). Both of these observations are consistent with gapless Dirac band transport in the presence of disorder\textsuperscript{4,28}. 
Figure 4 | Temperature dependence of resistance at high magnetic field. Sheet resistance ($R_s$) at $B = a$, 0 T $b$, 11.5 T $c$, 20 T and $d$, 44.5 T as a function of $V_G$ at several different temperatures. Note that the film was not measured at 9 K in (c). Inset in (a) shows semi-log plot of $R_s$ versus $T$ at $V_G = 0$ V indicating a small upturn at low temperatures, which fits log($T$) dependence as indicated by the blue line. Insets in (b- $d$) show behavior of $R_s$ around $V_G = 0$ V in greater detail. $e$, 2D plot of ratio $R_s(0.34 \text{ K})/R_s(1 \text{ K})$ plotted
as a function of $V_G$ and $B$. Metallic-like behavior is observed for $V_G > -21$ V while, for $V_G \lesssim -21$ V insulating-like behavior is observed. Semi-log plot of temperature dependence of normalized $R_{CNP}$ at $B = 11.5$ T and 44.5 T, where larger slope at 44.5 T indicates stronger insulating tendency: the solid lines are least-square fits using $R_{CNP} \sim \log(T)$.

In order to see how transport changes at higher fields, we plot $R_S$ versus $V_G$ at 11.5 T, 20 T and 44.5 T for $T = 0.35$ K – 9 K in Fig. 4 b-d, respectively (See Fig. S5 for more extensive data and details). A slight shift in $V_G$ corresponding to CNP at $T = 9$ K compared to lower temperatures is observed, but does not affect our analysis. In Fig. 4 b-d, $R_S(T_1) \approx R_S(T_2)$ for $V_G > -21$ V indicating metallic behavior, while for $V_G \lesssim -21$ V, $R_S(T_1) \gg R_S(T_2)$ suggesting an insulating behavior for these gate voltages for all three magnetic fields, where $T_1 < T_2$ are temperatures. Figure 4e summarizes this observation, where we show a 2D plot of the ratio of $R_S$ at $T = 0.3$ K to that at $T = 1$ K ($R_{0.3 K}/R_{1 K}$) as a function of $V_G$ and $B$. $n$-type carrier region shows metallic-like behavior (i.e. $R_{0.3 K}/R_{1 K} \approx 1$) along with QHE at high magnetic fields, while an insulator-like highly resistive state (i.e. $R_{0.3 K}/R_{1 K} > 1$) is observed near CNP and for $p$-type carrier region at high fields.

In order to further understand the nature of the highly-resistive state, we have plotted $R_{CNP}(T)/R_{CNP}(9$ K) as a function of temperature at $B = 11.5$ T and 44.5 T in Fig. 4f. $R_{CNP}(T)/R_{CNP}(9$ K) increases logarithmically with decreasing temperature for both $B = 11.5$ T and 44.5 T, with stronger insulating behavior observed at higher field as indicated by greater slope of $R_{CNP}$ vs. log($T$) for $B = 44.5$ T. For comparison, insets of Fig. 4 b-d show an enlarged view of $R_S$ versus $V_G$ at $V_G \approx 0$ V, where $R_S$ either decreases or does not increase significantly with decreasing temperature indicating a metallic behavior. Such an increasingly insulating behavior near CNP at higher magnetic fields indicates presence of a magnetic-field-induced insulator phase, whose
origin remains unknown at present. Local and non-local measurements at lower temperatures and higher magnetic fields could elucidate the nature of this ground state, which we leave for future work.

In conclusion, we have studied gate-dependent QHE on low-carrier density Bi$_2$Se$_3$ thin films for the first time by employing a novel buffer layer growth method on gate-amenable SrTiO$_3$ substrates. At low fields we observe ambipolar transport for thinner films, and at high fields we observe $v = 1$ QHE for $n$-type carriers. On the other hand, for CNP and $p$-type carriers we observe non-saturating magnetoresistance up to $B = 44.5$ T, whose temperature dependence point to the existence of a magnetic-field-induced insulating state. Further experimental and theoretical efforts are necessary to clarify its origin.

**Acknowledgements:**

This work is supported by the Gordon and Betty Moore Foundation’s EPiQS Initiative (GBMF4418) and National Science Foundation (NSF) grant EFMA-1542798. A portion of this work was performed at the National High Magnetic Field Laboratory which is supported by NSF Cooperative Agreement No. DMR-1644779 and the State of Florida.

**Author contributions:**

N.K. and S.O. conceived the experiment. N.K., M.S. and J.M. synthesized the samples and performed transport measurements. N.K. and S.O. wrote the manuscript with inputs from all the authors. All authors contributed to the scientific analysis and manuscript revisions.

**References:**

1. Essin, A. M., Moore, J. E. & Vanderbilt, D. Magnetolectric Polarizability and Axion Electrodynamics in Crystalline Insulators. *Phys. Rev. Lett.* **102**, 146805 (2009).
2. Wu, L. et al. Quantized Faraday and Kerr rotation and axion electrodynamics of a 3D topological insulator. Science 354, 1124–1127 (2016).

3. Fu, L. & Kane, C. L. Superconducting Proximity Effect and Majorana Fermions at the Surface of a Topological Insulator. Phys. Rev. Lett. 100, 96407 (2008).

4. Yoshimi, R. et al. Quantum Hall effect on top and bottom surface states of topological insulator (Bi$_{1-x}$Sb$_x$)$_2$Te$_3$ films. Nat. Commun. 6, 6627 (2015).

5. Xu, Y. et al. Observation of topological surface state quantum Hall effect in an intrinsic three-dimensional topological insulator. Nat. Phys. 10, 956–963 (2014).

6. He, Q. L. et al. Chiral Majorana fermion modes in a quantum anomalous Hall insulator–superconductor structure. Science 357, 294-299 (2017).

7. Xiao, D. et al. Realization of the Axion Insulator State in Quantum Anomalous Hall Sandwich Heterostructures. Phys. Rev. Lett. 120, 56801 (2018).

8. Chang, C.-Z. et al. Experimental Observation of the Quantum Anomalous Hall Effect in a Magnetic Topological Insulator. Science 340, 167–170 (2013).

9. Xu, Y., Miotkowski, I. & Chen, Y. P. Quantum transport of two-species Dirac fermions in dual-gated three-dimensional topological insulators. Nat. Commun. 7, 11434 (2016).

10. Zhang, H. et al. Topological insulators in Bi$_2$Se$_3$, Bi$_2$Te$_3$ and Sb$_2$Te$_3$ with a single Dirac cone on the surface. Nat. Phys. 5, 438–442 (2009).

11. Cao, H. et al. Quantized Hall Effect and Shubnikov–de Haas Oscillations in Highly Doped Bi$_2$Se$_3$: Evidence for Layered Transport of Bulk Carriers. Phys. Rev. Lett. 108, 216803 (2012).

12. Koirala, N. et al. Record Surface State Mobility and Quantum Hall Effect in Topological Insulator Thin Films via Interface Engineering. Nano Lett. 15, 8245–8249 (2015).
13. Moon, J. et al. Solution to the hole-doping problem and tunable quantum Hall effect in Bi$_2$Se$_3$ thin films. *Nano Lett.* **18**, 820–826 (2018).

14. Neville, R. C., Hoeneisen, B. & Mead, C. A. Permittivity of strontium titanate. *J. Appl. Phys.* **43**, 2124–2131 (1972).

15. Salehi, M. et al. Stability of low-carrier-density topological-insulator Bi$_2$Se$_3$ thin films and effect of capping layers. *APL Mater.* **3**, 91101 (2015).

16. Chen, J. et al. Gate-voltage control of chemical potential and weak antilocalization in Bi$_2$Se$_3$. *Phys. Rev. Lett.* **105**, 176602 (2010).

17. Edmonds, M. T. et al. Air-stable electron depletion of Bi$_2$Se$_3$ using molybdenum trioxide into the topological regime. *ACS Nano* **8**, 6400–6406 (2014).

18. Brahlek, M., Koirala, N., Salehi, M., Bansal, N. & Oh, S. Emergence of decoupled surface transport channels in bulk insulating Bi$_2$Se$_3$ thin films. *Phys. Rev. Lett.* **113**, 026801 (2014).

19. Kim, D. et al. Surface conduction of topological Dirac electrons in bulk insulating Bi$_2$Se$_3$. *Nat Phys* **8**, 459–463 (2012).

20. Beidenkopf, H. et al. Spatial fluctuations of helical Dirac fermions on the surface of topological insulators. *Nat. Phys.* **7**, 939 (2011).

21. Morimoto, T., Furusaki, A. & Nagaosa, N. Charge and spin transport in edge channels of a $\nu = 0$ quantum Hall system on the surface of topological insulators. *Phys. Rev. Lett.* **114**, 146803 (2015).

22. Hanaguri, T., Igarashi, K., Kawamura, M., Takagi, H. & Sasagawa, T. Momentum-resolved Landau-level spectroscopy of Dirac surface state in Bi$_2$Se$_3$. *Phys. Rev. B* **82**, 81305 (2010).
23. Cheng, P. et al. Landau Quantization of Topological Surface States in Bi$_2$Se$_3$. *Phys. Rev. Lett.* **105**, 76801 (2010).

24. Xia, Y. et al. Observation of a large-gap topological-insulator class with a single Dirac cone on the surface. *Nat. Phys.* **5**, 398–402 (2009).

25. Menshchikova, T. V et al. Band structure engineering in topological insulator based heterostructures. *Nano Lett.* **13**, 6064–6069 (2013).

26. Zhang, J. et al. Band structure engineering in (Bi$_{1-x}$Sb$_x$)$_2$Te$_3$ ternary topological insulators. *Nat. Commun.* **2**, 574 (2011).

27. Pauly, C., Saunus, C., Liebmann, M. & Morgenstern, M. Spatially resolved Landau level spectroscopy of the topological Dirac cone of bulk-type Sb$_2$Te$_3$(0001): Potential fluctuations and quasiparticle lifetime. *Phys. Rev. B* **92**, 85140 (2015).

28. Lang, M. et al. Competing weak localization and weak antilocalization in ultrathin topological insulators. *Nano Lett.* **13**, 48–53 (2013).
Supporting Information

Content:

1. Growth of Bi$_2$Se$_3$ thin films grown on In$_2$Se$_3$ based buffer layer on SrTiO$_3$ (111) substrate.
   Contains Fig. S1.

2. (Anti) symmetrization of sheet (Hall) resistance and electrical properties of Se-capped films.
   Contains Fig. S2.

3. Extended data set at more $V_G$ values and plateau-like features at $R_H \approx h/3e^2$ in the 10 QL film measured in Fig. 2 as well as explanation of limited $B$ field data for $V_G \leq -70$ V. Contains Fig. S3.

4. Data at additional $B$ field values related to Fig. 3 of the main text. Contains Fig. S4.

5. Temperature dependence of $R_S$ versus $V_G$ at extended $T$ and $B$ values for the 10 QL film shown in Fig. 4 of the main text. Contains Fig. S5.

---

1. Growth of Bi$_2$Se$_3$ thin films on In$_2$Se$_3$ based buffer layer on SrTiO$_3$(111) substrate.

Films were grown on a custom-built molecular beam epitaxy system from SVTA Inc. with a base pressure of $2 \times 10^{-10}$ Torr. SrTiO$_3$ substrates were prepared by $ex$ $situ$ cleaning in a UV-ozone system for 5 minutes and $in$ $situ$ at 630 °C for 10 minutes in oxygen pressure of $2 \times 10^{-6}$ Torr. High purity (>99.99%) elemental sources of bismuth, indium and selenium were evaporated using effusion cells. Beam fluxes were calibrated $in$ $situ$ using quartz crystal microbalance and $ex$ $situ$ using Rutherford backscattering, which together give the accuracy of film thickness and composition to within 1%. Growth rate is determined by cation flux (i.e. Bi or In). Selenium flux is always maintained at more than 10 times the cation flux and Se shutter is kept open at all times during the growth. For (Bi$_{0.5}$In$_{0.5}$)$_2$Se$_3$ growth, both Bi and In shutter were opened/closed simultaneously. Reflection high energy electron diffraction (RHEED) was used to monitor the crystal quality during the film growth.
For In\textsubscript{2}Se\textsubscript{3} based buffer layer, we first grew a 3 QL Bi\textsubscript{2}Se\textsubscript{3} seed layer at 150 °C followed by annealing to 300 °C at which temperature an additional 47 QL Bi\textsubscript{2}Se\textsubscript{3} was deposited followed by deposition of 5 QL In\textsubscript{2}Se\textsubscript{3}. This entire structure was then heated to 600 °C, during which process 50 QL Bi\textsubscript{2}Se\textsubscript{3} evaporates leaving only 4 QL In\textsubscript{2}Se\textsubscript{3} behind. The sample temperature was then lowered to 275 °C, where we deposited 4 QL (Bi\textsubscript{0.5}In\textsubscript{0.5})\textsubscript{2}Se\textsubscript{3} layer. This 5 QL In\textsubscript{2}Se\textsubscript{3} – 4 QL (Bi\textsubscript{0.5}In\textsubscript{0.5})\textsubscript{2}Se\textsubscript{3} structure forms the buffer layer and is electrically insulating as indicated by immeasurably high resistance at room temperature. On top of this buffer layer, desired thickness of Bi\textsubscript{2}Se\textsubscript{3} layer was then deposited at the same temperature. Figure S1a shows the RHEED image of a representative Bi\textsubscript{2}Se\textsubscript{3} film grown on the buffer layer, where the streaks indicate a highly crystalline and smooth film. The sample was then allowed to cool below 50 °C before depositing either 100 nm Se or 50 nm MoO\textsubscript{3} followed by 50 nm Se (referred to as MoO\textsubscript{3}/Se capping in the main text) for capping purpose. Se capping provides protection against surface ambient contamination, while MoO\textsubscript{3}/Se also lowers the Fermi level due to charge transfer doping from MoO\textsubscript{3}. ~100 nm thick copper was sputtered onto the backside of the samples to form the back gate electrode. The samples were then fabricated into Hall bars by placing them under a metal mask and scratching the exposed area with a tweezer. Pressed indium wires were used to make all electrical contacts. Figure S1b shows the film structure and the measurement geometry.
Figure S1 | Film structure and measurement geometry. a, RHEED streaks indicating highly crystalline and smooth surface of a Bi$_2$Se$_3$ film grown on top of buffer layer. b, Schematic of the film structure and the Hall bar geometry. Optical image of a representative hand-patterned Hall bar device is also shown.

2. (Anti) symmetrization of sheet (Hall) resistance and electrical properties of Se-capped films

Magnetoresistance is an even function of magnetic field i.e. $R_S(B) = R_S(-B)$ and Hall resistance is an odd function i.e. $R_H(B) = -R_H(-B)$. In measurements, Hall and longitudinal voltages can mix due to misalignment of measurement leads leading to cross-contamination. In order to remove this unwanted mixing of sheet resistance to the measured Hall resistance, we perform post-measurement anti-symmetrization in order to extract the true Hall resistance, $R_H(B) = [R_{Hall}(B) - R_{Hall}(-B)]/2$, where $R_{Hall}(B)$ is the raw Hall resistance. Similarly, symmetrized sheet resistance, $R_S(B)$, can be obtained by $R_S(B) = [R_{sheet}(B) + R_{sheet}(-B)]/2$, where $R_{sheet}(B)$ is the raw sheet resistance. Since, the effect of mixing on sheet resistance tends to be small (usually a few percent), we use the same symbol, $R_S$, to denote both symmetrized and raw sheet resistance in the main text.

We have measured $R_H$ and $R_S$ of Se-capped Bi$_2$Se$_3$ films in Hall bar geometry at $T = 5$ K in $B$ field up to $\pm 0.6$ T as a function of film thickness. Figure S2 shows sheet carrier density ($n_S$) and mobility ($\mu$) obtained using the method described in the main text. $n_S$ of $\sim 4 - 7 \times 10^{12}$ cm$^{-2}$ represents significant improvement in film quality compared to Bi$_2$Se$_3$ films grown directly on SrTiO$_3$ (111) substrates.
Figure S2 | Sheet carrier density and mobility of Se-capped BiSe$_3$ films. a, sheet carrier density ($n_S$) and b, mobility ($\mu$) of Se-capped BiSe$_3$ films for four different film thickness ($t$) at $T = 5$ K. Please note that 1 QL = 1 nm.

3. Extended data set at more $V_G$ values and plateau-like features at $R_H \approx h/3e^2$ in 10 QL film measured in Fig. 2 as well as explanation of limited $B$ field data for $V_G \leq -70$ V.

For $V_G \leq -70$ V in Fig. 2 of the main text, sheet resistance ($R_S$) extends from $B \approx -30$ T to $B \approx 12$ T and anti-symmetrized Hall resistance ($R_H$) extends only up to $|B| \approx 12$ T. Due to limited measurement time at NHMFL, Florida, we only measured sheet and Hall resistance from $B \approx -30$ T to $B \approx 12$ T for $V_G \leq -70$ V. Since, anti-symmetrization procedure requires values at both $+B$ and $-B$ (See Supporting Information 2 above), the anti-symmetrized Hall resistance presented in Fig. 2 extends only to $|B| \approx 12$ T.

In Fig. S3a, we show $R_H$ versus $B$ at several different gate voltage values $V_G$. Data at $V_G = -20$ V and -25 V are in addition to those shown in Fig. 2 of the main text. Along with plateaus at $\frac{h}{e^2}$ ($v = 1$ QHE) observed in Fig. 2b-c of the main text, Hall resistance ($R_H$) shows plateau-like features at moderate $B$ field for several different gate voltage values as seen in Fig. S3a. In order to clearly visualize this feature, we have plotted the derivative of $R_H$ w.r.t. $B$ ($dR_H/dB$) versus $B$ as shown in Fig. S3b. The local minima in the derivative plot, as indicated by arrows for plots corresponding to $V_G = 0, -15$ and -20 V values indicate the centre of the plateau-like features. From the $B$ value corresponding to this arrow we can extract the $R_H$ value of the plateau-like features as shown in Fig. S3c for $V_G = -25$ V (See the black dashed line). Finally, in Fig. S3d, we tally all the plateau-like features, which lie at $\sim h/3e^2$ indicating an incipient $v = 3$ QHE state. For completeness, we also plot the plateau values at $h/e^2$, which correspond to the $v = 1$ QHE state. No plateau-like features were observed at $\sim h/2e^2$, indicating that the $v = 2$ QHE is absent in our film. Such absence of even-valued QHE is expected when both top and bottom surfaces have similar carrier density, such that $v_T = v_B$ resulting in odd-valued $v = v_T + v_B$, where $v_T (B)$ is the top (bottom) surface filling factor and is half-integer valued$^1$. We note that corresponding dip-like features in sheet resistance are not resolvable for the incipient $v = 3$ QHE state.
Figure S3 | QHE in a MoO3/Se-capped 10 QL thick Bi2Se3 film at additional \( V_G \) values compared to Fig. 2 in the main text. a, Hall resistance \( R_H \) and b, derivative of Hall resistance \( dR_H/dB \) as a function of magnetic field, \( B \), at different \( V_G \). In (b) the vertical arrows mark the local minima in \( dR_H/dB \), which correspond to the centre of plateau-like features in \( R_H \). This is indicated by the vertical dashed black line in (c) where \( dR_H/dB \) and \( R_H \) are overlaid for a representative plot at \( V_G = -20 \) V. c, \( R_H \) plateaus at \( \sim \hbar/e^2 \) and plateau-like features at \( \sim \hbar/3e^2 \) are observed for several different \( V_G \) values corresponding to \( v = 1 \) QHE and incipient \( v = 3 \) QHE, respectively, while no plateau (-like) feature is observed at \( \sim \hbar/2e^2 \) indicating absence of \( v = 2 \) QH state.

4. Data at additional \( B \) field values related to Fig. 3 of the main text.

In Fig. S4, we plot \( R_S \) vs. \( V_G \) (left panel) and \( R_H \) vs. \( V_G \) (right panel) at extended \( B \) field values compared to Fig. 3 in the main text.
Figure S4 | Sheet and Hall resistance versus $V_G$ at additional $B$ values compared to Fig. 3 of the main text. Sheet resistance (left) and Hall resistance (right) as a function of the gate voltage, $V_G$, for additional magnetic field values (compared to Fig. 3 of the main text) at $T = 0.35$ K for 10 QL thick film.

5. Temperature dependence of $R_S$ versus $V_G$ at additional $B$ field values for the 10 QL film shown in Fig. 4 of the main text.

In this section we show further temperature dependence of the 10 QL film in support of Fig. 4 of the main text. As shown in Fig. S5a, zero-field sheet resistance ($R_S$) versus $V_G$ of a different (but identically prepared) 10 QL film at $T \approx 6 - 12$ K does not show significant change with temperature. Such small increase or change at all measured $V_G$ values is consistent with previous reports on Dirac-band transport in TIs$^2$. In Fig. S5b, we show temperature dependence of $R_S$ versus $V_G$ at $B = 30$ T. Consistent with the discussions of the main text, we observe metallic-like behaviour for $V_G \gtrsim -21$ V, while insulating-like behaviour for $V_G \lesssim -21$ V.

Figure S5 | Temperature dependence of sheet resistance versus $V_G$ at additional magnetic field values compared to Fig. 4 of the main text. a-b, Temperature dependence of sheet resistance at (a) zero-magnetic field for a different but identically prepared 10 QL film and (b) $B = 30$ T for the film used in Fig. 4 of the main text.

References:

1. Xu, Y. et al. Observation of topological surface state quantum Hall effect in an intrinsic three-dimensional topological insulator. Nat. Phys. 10, 956–963 (2014).

2. Yoshimi, R. et al. Quantum Hall effect on top and bottom surface states of topological insulator (Bi$_{1-x}$Sb$_x$)$_2$Te$_3$ films. Nat. Commun. 6, 6627 (2015).
