Trajectory of the anomalous Hall effect towards the quantized state in a ferromagnetic topological insulator

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I. OPTIMIZATION OF FILM STOICHIOMETRY

The sheet carrier density $n_s$ for the $\text{Cr}_x(\text{Bi}_{1-y}\text{Sb}_y)_{2-x}\text{Te}_3$ films studied here for fixed $y$ and film thickness $t$ is found to remain nearly constant with increasing magnetic dopant concentration $x$. A quantitative measure of this constancy is the difference in applied top gate voltage $V_T$ needed to achieve charge neutrality (the charge neutral value denoted as $V_0$ in the main text) for different $x$. For the films studied here, $y = 0.8$ is chosen such that at $x = 0$ devices have $|V_0| < 1$ V (there is some variation from film to film likely due to processing after growth). As reported in the main text, for $x = 0.22$ we find $V_0 \approx 3$ V, corresponding to a change in sheet carrier density $\Delta n_s$ of the order of $10^{12}$ cm$^{-2}$. If Cr were to enter as $\text{Cr}^{2+}$ in analogy with the case of Mn [S1], we would expect $\Delta n_s = x/2V \sim 10^{14}$ cm$^{-2}$ ($V$ is the unit cell volume) for this $x$. Accordingly, it appears more likely that the dopant enters the system as $\text{Cr}^{3+}$, substantially simplifying the optimization process of the films for charge transport studies.

FIG. S1: (a) Gate voltage $V_T$ dependence of magnetoresistance $R_{xx}(B)$ in the vicinity of the coercive field $H_C$ at $T = 0.2$ K. The direction of the hysteresis loop is shown with arrows. The peak in $R_{xx}(B)$ marks $H_C$, which appears to be independent of $V_T$. (b) $H_C(V_T)$ for two films with $x = 0.22$ and 0.27 at $T = 0.2$ K. (c) $x$ dependence of $H_C$ and the remnant contribution of the Hall resistance $R_{yx}^A$ at $T = 2$ K.
An additional simplification arises due to the carrier independent nature of the magnetism in this system that has been previously predicted and reported [S2, S3, S4]. While this aspect of the magnetic ordering is different than that predicted for surface state mediated ferromagnetism [S5, S6], it nonetheless represents bulk ferromagnetism in close proximity to the surface modes that ensures breaking of time reversal symmetry in the surface states. We can probe the strength of the magnetic order by studying the coercive field $H_C$ as measured by either the longitudinal magnetoresistance $R_{xx}(B)$ or the Hall resistance $R_{yx}(B)$, the former being marked by a maximum in the “butterfly” magnetoresistance and latter a sign change in the anomalous Hall resistance with magnetic reversal, as seen in Fig. 2 of the main text. Focusing on $R_{xx}(B)$, the top gate voltage $V_T$ dependence (relative to the charge neutral point $V_0$) for $x = 0.27$ in the vicinity of the peak region is shown in Fig. S1(a). $H_C$ derived from the peak position for this doping and $x = 0.22$ shown in the main text are shown in Fig. S1(b). There appears to be no systematic dependence of $H_c$ on $V_T$ and therefore the carrier density $n$, though there is a clear shift between different $x$. This allows us to control the magnetic properties of the films with a single parameter $x$ (though the transport properties are additionally affected by the thickness $t$, see the next section).

Focusing on the $x$ dependence at $T = 2$ K, $H_C(x)$ shown in Fig. S1(c) is an increasing function of $x$. In the same films, the magnitude of $R_{yx}$ at $T = 2$ K instead shows a peak in the vicinity of $x = 0.22$. We can understand this behavior by considering that the ferromagnetic order in the films is a monotonically increasing function of $x$, but that the anomalous Hall effect has an enhanced contribution in the topologically non-trivial phase. The relevance of the latter can be understood by considering the evolution of the topologically non-trivial band structure upon replacement of the large spin-orbit material Bi with Cr- eventually the bands well revert to an un-inverted structure at large enough $x$ through a quantum phase transition and the topologically non-trivial contribution to $R_{yx}$ will be lost. Such a phase transition driven by the strength of the spin-orbit interaction has been observed previously in BiTl(S$_{1−δ}$Se$_δ$)$_3$ [S7]. From these observations we conclude that the films with strongest magnetic order which retain a topologically non-trivial band structure have $x = 0.22$ and are near charge neutrality for $y = 0.8$. 

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II. ROLE OF FILM THICKNESS

The optimization of the film thickness $t$ concerns the tendency of films in the ultra-thin limit ($t < 5$ nm) to become insulating at low $T$, which prevents meaningful transport measurements from being performed, while at the same time thinner films allow for more significant suppression of bulk conduction both geometrically and with our field effect geometry configuration. Using the optimized conditions from the previous section, we have grown a series of films varying $t$ from 5 nm to 12 nm and measured $R(T)$, shown in Fig S2. As described in the main text, while we attribute the high $T > 100$ K behavior to thermal excitation of the bulk electronic bands, the low $T$ behavior reflects the charge transport of the surface modes. As $t$ is reduced below 7 nm, the onset of insulating behavior below $T = 10$ K becomes particularly clear. We also note that the observed peak in $R(T)$ which appears in the vicinity of the ferromagnetic transition temperature $T_C$ shifts down with decreasing $t$, though for $t = 5$ nm the associated $T_C$ is still larger than previously reported [S4].

FIG. S2: Film thickness $t$ dependence of resistance as a function of temperature $R(T)$ for films with $x = 0.22$ and $y = 0.8$ shown in a semi-log scale. The films become electrically insulating at low $T$ as the thickness is decreased.

There are different possible mechanisms for the insulating behavior, including island formation in very thin films due to surface roughness as well as electronic gap formation.
(which would compete with the QAHE gap) in the surface modes due spatially driven hybridization [S8]. While the specific mechanism underlying the behavior in Fig. S2 are not clear, it suggests the use of films above a critical $t \approx 7$ nm. We chose to work use $t = 8$ nm, so that the films are near but above the critical crossover regime between metallic and insulating behavior and remain conducting even at significantly lower temperatures $T < 100$ mK.

![Graph](image)

**FIG. S3:** Current voltage characteristic $I(V)$ across current leads taken in the vicinity of the QAHE state with $V_T = -1$ V shown in Fig. 3(a) of the main text (thickness $t = 8$ nm, patterned device) showing a linear response.

Films of 8 nm and thicker remain conducting to low $T$ and display linear $I(V)$ behavior; a typical example is shown in Fig. S3. Here, measurements are taken with a DC voltage applied and measured across the current terminals of the sample (a 2-probe configuration, a 4-probe configuration also shows a linear response) while the current flowing through the circuit is measured with a current-voltage converting amplifier. It is interesting to note that for films slightly thicker than 8 nm we still observe signatures of the underlying QAHE state. Fig S4 shows a film with $t = 10$ nm with $R_{yx}$ reaching above $0.9h/e^2$ at $T = 50$ mK. Contaminant with this, a minimum in $R_{xx}$ is seen to develop, deepening as $T$ decreases from 200 mK to 50 mK. This suggests that if $T$ could be reduced further, the QAHE state would likely develop. The decrease in temperature scale may be due the increased strength
of bulk conduction or the reduced homogeneity of the chemical potential in the film along the vertical direction. The latter could presumably be addressed with the use of both top and bottom electrostatic gates.

![Graph](image)

**FIG. S4**: Transport result for a film of thickness \( t = 10 \) nm. The characteristic features of the QAHE are present in the form of the maximum in \( R_{yx} \) and simultaneous dip in \( R_{xx} \), but these values do not develop into the fully quantized/dissipationless values at 50 mK.

### III. FLOW BEHAVIOR WITH APPLIED MAGNETIC FIELD

In addition to the flow of \((\sigma_{xy}(V_T), \sigma_{xx}(V_T))\) to the fixed stable points observed for \( B = 0 \), we observe qualitatively similar behavior in an applied magnetic field where the dissipationless behavior in \( R_{xx} \) is more closely realized. Trajectories for \( B = 14 \) T are shown in Fig. S3. In this case, flow toward the \( \nu = 1 \) critical point is clear, though there is still a deviation from a perfectly quantized response.

Another interesting feature here is the result at \( V_T = +6.75 \) V, which appears to be very nearly tuned to the boundary between the two stable phases. Only in this critically balanced condition does the trajectory flow toward \((\sigma_{xy}, \sigma_{xx}) = (e^2/2h, e^2/2h)\), consistent with the existence of an unstable fixed point associated with delocalization [S9, S10].
FIG. S5: Flow behavior with magnetic field $B = 14$ T for temperature $T$ between 50 mK and 700 mK. The flow direction is toward lower $\sigma_{xx}$ with decreasing $T$.

As the flow lines here and in the main text are indicative of a phase transition between an insulating and quantum Hall phase, a natural question is if the $B$ dependence also shows a divergence of the localization length $\xi$ associated with the delocalization transition as has been observed in the integer quantum Hall case [S11]. The analysis of the $B$ driven transition is typically done by examining the slope of the plateau to plateau transition in $R_{yx}$, which evaluated at a critical magnetic field $B_C$ diverges as a power law in temperature $T^{-\kappa}$, with $\kappa = 0.42$. We note that $\kappa$ itself is not a universal value but connects to the divergence in $\xi \propto |B - B_c|^\nu$ through the inelastic scattering length temperature exponent $p$ as $\kappa = p/2\nu$. Upon analyzing the slope in the present context, we find that we cannot identify a $B_C$ in the same manner as was done in the integer quantum Hall case. As can be seen for sample 2 in Fig. S6, as a function of $T$ the position of the maximum in $dR_{yx}/dB$ shifts. We speculate that this difficulty is due to the dynamics of magnetization reversal affecting the observed transition. The behavior is different from sample to sample and seems unlikely to be probing a universal feature of the transition. Recent theoretical work has suggested that in fact the relevant regime for seeing the universality is between $R_{yx} = 0$ and $\hbar/e^2$ [S12]- the present samples do not clearly show a regime where the former develops and thus may indicate why
FIG. S6: Field derivative of the Hall response $R_{yx}$ in the vicinity of the ferromagnetic transition in sample 2. We observe a systematic shift of this critical $B$.

we are unable to connect with scaling in $B$. Improving the samples to observe this effect is the subject of ongoing work.

IV. SYMMETRIZATION

We briefly comment on the use of field symmetrization in our analysis. The only data presented in the manuscript that are symmetrized to remove the contamination between longitudinal and transverse voltages are in Fig. 1(e) and 1(f) of the main text. As described in methods, those measurements were performed on unpatterned films with hand made electrical contact that left them prone to such effects. For completion, in Fig. S7 we show the unsymmetrized data in which the same trends can be discerned, but a clear mixing is observed.

V. MEASUREMENT CONFIGURATION

A schematic of the circuit used for measurement in the main text is shown in Fig. S8. Three lock-in amplifiers (SR830, Stanford Research Systems, synchronized at 1-3 Hz) are
FIG. S7: Unsymmetrized $R_{xx}$ and $R_{yx}$ for unpatterned films. There is a clear contamination of the signals that we remove in the main text by field symmetrizing the data.

used to detect the current and voltages in the circuit, with a current/voltage converter (Model 1211, DL Instruments, typically set to $10^{-6}$ A/V) and high impedance (> 1 GΩ) voltage pre-amplifiers (Model 1201, DL Instruments, typically set to 50 times gain) used for amplification, respectively. Filtering is performed with $\pi$-filters with a cutoff frequency of 1 MHz on each line at room temperature and with $RC$ filtering at low temperature. The current in the circuit is limited to 1 nA by a series resistor. The virtual ground of the current/voltage converter provides a low impedance path to ground.


FIG. S8: Configuration of measurement circuit. The components in the blue shaded region are located within the cryostat. The current to voltage converter feeds in to lock-in 1 while the voltage pre-amplifiers feed in to lock-ins 2 and 3.

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