High-precision observation of nonvolatile quantum anomalous Hall effect

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Detailed Terms
High-precision observation of nonvolatile quantum anomalous Hall effect

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Abstract

The discovery of the integer quantum Hall (QH) effect in 1980 led to the realization of a topological electronic state with dissipationless currents circulating in one direction along the edge of a two dimensional electron layer under a strong magnetic field. The quantum anomalous Hall (QAH) effect shares a similar physical phenomenon as the QH effect, whereas its physical origin relies on the intrinsic spin-orbit coupling and ferromagnetism. Since the QAH effect does not require an external field and the associated Landau levels, it is believed that this effect has unique potential for applications in electronic devices with low-power consumption. Recently, the QAH effect has been experimentally observed in thin films of the ferromagnetic topological insulators (FMTI), Cr-doped (Bi,Sb)2Te3. However, in this system, a large residual longitudinal resistance \(0.098h/e^2(\sim2.53k\Omega)\) remains, and thus the Hall conductance is only around \(0.987e^2/h\) at zero magnetic field, hampering the precise realization of this dissipationless topological state. Here we report
the experimental observation of the QAH effect in V-doped \((\text{Bi,Sb})_2\text{Te}_3\) films with the zero-field longitudinal resistance down to 0.00013±0.00007\(\frac{h}{e^2}\) (~3.35±1.76\(\Omega\)), Hall conductance reaching 0.9998±0.0006\(\frac{e^2}{h}\) and the Hall angle becoming as high as 89.993±0.004° at \(T=25\text{mK}\), thus realizing the anomalous Hall transport with negligible dissipation in the absence of any initial magnetic field. The advantage of this system comes from the fact that it is a hard ferromagnet with a large coercive field \((H_c>1.0\text{T})\) and a relative high Curie temperature. This realization of nonvolatile QAH state in hard FMTIs is a major step towards dissipationless electronic applications without external fields.

The quantum anomalous Hall (QAH) effect describes the dissipationless quantized Hall transport in ferromagnetic materials in the absence of external magnetic fields.\(^3\) The realization of the QAH effect in realistic materials requires two conditions: ferromagnetic insulating materials and topologically non-trivial electronic band structures.\(^5,12,13\) It has been proposed that these two conditions can be satisfied by introducing ferromagnetism into topological insulators (TIs).\(^12,13\) Ferromagnetism can be achieved by doping TIs with transition metal atoms, such as Cr, V and Mn.\(^17,18,19\) Indeed, the QAH effect has been reported in Cr-doped \((\text{Bi,Sb})_2\text{Te}_3\) system.\(^14,15,16\) Among the various transition metal atoms, V-doped \(\text{Sb}_2\text{Te}_3\) exhibits the most stable ferromagnetism with a high Curie temperature \((T_c)\).\(^17,18\) This fact motivated us to explore the QAH effect in V-doped TI thin films.

Epitaxial thin films with various concentrations of V-doped \(\text{Sb}_2\text{Te}_3\) were prepared by coevaporation in a molecular beam epitaxy (MBE) system. Excellent ferromagnetic response was found for all samples, consistent with the early experiments.\(^17,18\) To achieve the insulating phase, a small amount of Bi was added to tune the chemical potential (see Supplementary Information).\(^20,21\) We note that \(T_c\) and \(H_c\) varies little with Bi doping, even in the rather insulating
samples around the $p$-$n$ crossover region, which indicates that there is a truly insulating ferromagnetic state in V-doped Sb$_2$Te$_3$.\textsuperscript{21} In order to realize accurate and continuous tuning of the Fermi energy and carrier density ($n_{2D}$), electric-field gating was also employed. The heat-treated SrTiO$_3$(111) substrate with a large dielectric constant serves as an effective bottom gate dielectric. The magneto-transport was measured on two samples, denoted as S1 and S2, using the standard six-terminal Hall bar configuration. (see Methods and Supplementary Information).\textsuperscript{14,21}

Our magneto-transport on a four quintuple-layers (QL) $(\text{Bi}_{0.29}\text{Sb}_{0.71})_{1.89}\text{V}_{0.11}\text{Te}_3$ film (sample S1) as shown in Fig. 1, exhibits nearly ideal QAH behavior. At the charge neutral point $V_g^0$, the Hall resistance ($\rho_{yx}$) at zero magnetic field (labeled as $\rho_{yx}(0)$) displays the quantized value of $1.00019\pm0.00069\hbar/e^2(25.8178\pm0.0177\text{k\Omega})$, while the longitudinal resistance ($\rho_{xx}$) at zero magnetic field (labeled as $\rho_{xx}(0)$) is only $0.00013\pm0.00007\hbar/e^2$ ($\sim3.35\pm1.76\text{\Omega}$) measured at $T=25\text{mK}$,\textsuperscript{22} shown in Figs 1b and 1c. The ratio $\rho_{yx}(0)/\rho_{xx}(0)$ corresponds to an anomalous Hall angle of $89.993\pm0.004^\circ$. The corresponding Hall conductance at zero magnetic field (labeled as $\sigma_{yx}(0)$) is up to $0.9998\pm0.0006e^2/\hbar$ and longitudinal conductance at zero magnetic field (labeled as $\sigma_{xx}(0)$) is down to $0.00013\pm0.00007e^2/\hbar$. For comparison, $\rho_{xx}(0)$, $\sigma_{yx}(0)$, $\sigma_{xx}(0)$ and the anomalous Hall angle in the Cr-doped system are $0.098\hbar/e^2(\sim2.53\text{k\Omega})$, $0.987e^2/\hbar$, $0.096e^2/\hbar$ and $84.40^\circ$ at $T=30\text{mK}$, respectively,\textsuperscript{14} noticeably different from the expected values. The precise values of these quantities at zero-field were not reported systematically in ref. 15 and 16. Additional measurement about temperature and gate voltage dependence has also been performed, showing that the QAH behavior with an anomalous Hall angle of $84.40^\circ$ (the largest Hall angle in Cr doped system) in V-doped system is seen up to ~$130\text{mK}$. (see Supplementary Information).
What we have found in V-doped system is clearly a more precise experimental confirmation of the ideal QAH effect than that in Cr-doped system. To understand the differences between these two materials, we carried out direct comparison of the two systems. Our studies showed three main differences. (1) Figure 2a shows the dependence of $T_C$ for 6QL films of Sb$_{2-x}$Cr$_x$Te$_3$ and Sb$_{2-x}$V$_x$Te$_3$ as a function of the Cr or V doping concentration $x$. The $T_C$ of a FM material can be determined from the Arrott plot (see Supplementary Information). At the same concentration $x$, the $T_C$ for V doping is twice value than that for Cr doping. Having a higher $T_C$ with low $x$ is in the right direction for enhancing the temperature to obtain QAH state. (2) The Hall traces for $x=0.13$ films are shown in Fig. 2b. In both cases, the square-shaped loops indicate the long-range ferromagnetic order with out-of-plane magnetic anisotropy. The coercive field $H_c$ for the Sb$_{1.87}$V$_{0.13}$Te$_3$ film is ~1.3T at $T$=2K, an order of magnitude larger than that of Sb$_{1.87}$Cr$_{0.13}$Te$_3$ with $H_c$~0.1T. The large $H_c$ of V-doped Sb$_2$Te$_3$ is clearly advantageous for a nonvolatile FMTI system. (3) Remarkably, the $n_{2D}$ of 6QL Sb$_{1.87}$V$_{0.13}$Te$_3$ is ~ 2.5×10$^{13}$ cm$^{-2}$, about one half of $n_{2D}$ in Sb$_{1.87}$Cr$_{0.13}$Te$_3$ ($n_{2D}$~4.9×10$^{13}$ cm$^{-2}$). The low $n_{2D}$ in the parent material is beneficial because it reduces the amount of Bi doping required to drive the system towards a charge neutral state.

The main difference between these two materials lies in different magnetic moments and valence of V and Cr atoms. The saturation magnetic moment per V ion was determined to be 1.5μ$_B$ (Bohr magnetron) for $x=0.13$ V-doped Sb$_{2-x}$V$_x$Te$_3$ (shown in Fig. S6). This suggests that the valence state of V is a mixture of 3+ and 4+ (or/and 5+) as it is expected to substitute for the Sb$^{3+}$ ion on the Sb sub-lattice. The extra free electrons resulting from V$^{4+}$ (or/and V$^{5+}$) go to neutralize the $p$-type carriers, so the $n_{2D}$ of V-doped Sb$_2$Te$_3$ is expected and measured to be lower than that of Cr-doped samples, in which Cr takes the valence state 3+.17,21
The magnetic moment of V atom is smaller than that of Cr atom (~3μB)\(^{17,21}\) and thus with exceptionally high \(H_c\). The \(H_c\) in FM films, in addition to extrinsic influences such as defects that pin domain walls, depends on the magnetic anisotropy (both crystallographic and shape) and saturation magnetization moment (\(M_s\)) of the films. For magnetization reversal by the process of magnetic domain rotation, \(H_c \propto \frac{K}{M_s}\), where \(K\) is the total magnetic anisotropy constant, so a higher \(K\) and/or lower \(M_s\) can lead to a high \(H_c\). Since the demagnetization energy (\(E_d\)) determines the number of magnetic domain walls, and \(E_d \propto M_s^2\), materials with few domains have a high \(H_c\) while those with many domains have a low \(H_c\)\(^{26,27}\). Therefore it is clear that V-doped Sb\(_2\)Te\(_3\) with a lower \(M_s\) and a higher \(H_c\) have fewer magnetic domains than Cr-doped systems, indicating that the magnetic domains in V-doped Sb\(_2\)Te\(_3\) are much larger in size than in Cr-doped films. The two main sources of magnetic anisotropy, magnetocrystalline anisotropy and surface anisotropy, appear to be very different for V- and Cr-doped Sb\(_2\)Te\(_3\) films. The magnetocrystalline anisotropy is usually the most effective means of impeding the magnetization reversal by rotation processes, while the surface anisotropy is a major contributor in nm thick films. In high-quality epitaxial V-doped Sb\(_2\)Te\(_3\) with large atomically flat areas, a near-perfect layered structure, and the slight expansion of c-axis lattice parameter may lead to the large magnetic anisotropy in the present films (see Supplementary Information)\(^{26,27}\).

Due to the more robust ferromagnetism, V-doped Sb\(_2\)Te\(_3\) films exhibit QAH state spontaneously without any magnetic field training, in sharp contrast to the Cr-doped systems\(^{14,15,16}\). Figure 3a shows the temperature-dependence of \(\rho_{xx}\) and \(\rho_{yx}\) for a different 4QL (Bi\(_{0.29}\)Sb\(_{0.71}\))\(_{1.89}\)V\(_{0.11}\)Te\(_3\) film(sample S2), using a bottom gate bias \(V_g=0\) and no applied magnetic field and cooled from room temperature under zero-field. \(\rho_{xx}\) shows semiconducting behaviors from room temperature down to ~2.5K, where a drop occurs, signaling that it is entering the
QAH state. $\rho_{yx}$ remains constant from room temperature down to ~23K where it increases rapidly with further decreasing in temperature. This indicates that the system enters the FM state below ~23K. $\rho_{yx}$ continues to increase and $\rho_{xx}$ continues to decrease with decreasing temperature, $\rho_{yx}$ approaches ~0.99$h/e^2$, and $\rho_{xx}$ reaches ~0.30$h/e^2$ at the lowest temperature of the measurement at ~80mK. This behavior signifies the film spontaneously progressing toward the QAH state. The net magnetization of virgin state in the film, i.e. not induced by any external field, and the reduced thermal activation at lower temperature drives the film from the diffusive transport regime towards the conducting QAH state. The inset of Fig. 3a shows the low-temperature data at $V_g$=0V below 3K, for magnetic fields $\mu_0H$=0 and 2T. This spontaneous decrease of $\rho_{xx}$ at zero-field seen for V-doping is absent in Cr-doped TIs.\textsuperscript{14,15,16} In Cr-doped TI films, unless the magnetic domains are aligned with an external applied perpendicular magnetic field, $\rho_{xx}$ increases with decreasing temperature and no QAH state can be achieved even at very low temperatures.\textsuperscript{15,16} Applying a field of 2T to the V-doped TI film, on the other hand, did not change the qualitative temperature dependence in $\rho_{xx}$ and $\rho_{yx}$, from that of the zero-field cooling behavior. This clearly demonstrates that the macroscopic net magnetization of the virgin state in V-doped TIs is spontaneous and highly developed. The fact that virgin $\rho_{yx}$ and $\rho_{xx}$ curves overlap with the hysteresis loop (Fig. 3b) and the butterfly structure (Fig. 3c), also confirms the spontaneous self-magnetization of V-doped TI samples in the QAH regime. The high self-magnetization also appears in the direct magnetization measurements (see Supplementary Information).

The QAH state observed in V-doped TIs is unexpected from the view of first-principle calculations that points to the absence of an insulating FM state in V-doped TI due to the presence of $d$-orbital impurity bands in the bulk band gap.\textsuperscript{12} Furthermore, in the QAH state
regime, the gate-voltage dependence of $H_c$ of V-doped film (see Supplementary Information) is much larger than that of Cr-doped system,\textsuperscript{14} also indicating the possible existence of FM induced from impurity bands, in addition to that from the van Vleck mechanism.\textsuperscript{12} A possible explanation why there exists an insulating FM state could be that the $d$-orbital impurity bands become localized at lower temperatures, so that it does not participate in the charge transport and the whole system becomes an Anderson insulator instead of a band insulator. The localization physics in the QAH regime has been studied numerically in Ref. 10, which confirms that the QAH state can indeed be achieved in a metallic system by introducing disorder to localize bulk carriers. Thus, our experiments indicate the role of the localization physics of impurity bands in V-doped systems.

Our results demonstrate the high-precision confirmation of the QAH effect with the zero-field longitudinal resistance down to $0.00013 \pm 0.00007h/e^2$ ($\sim 3.35 \pm 1.76\Omega$) and zero-field Hall conductance reaching up to $0.9998 \pm 0.0006e^2/h$ in V-doped $(\text{Bi,Sb})_2\text{Te}_3$, a hard FMTI. It is particularly remarkable considering that mobility of the device measured near 80K is only $\sim 130\text{cm}^2/\text{Vs}$ (see Supplementary Information), orders of magnitude smaller than that found in semiconductor hetero-structures with two dimensional electron gas (2DEG) that reached the QH state.\textsuperscript{1,2} This FMTI is found to be in the QAH state without the aid of a polarizing external magnetic field, making it a promising candidate for dissipationless electronic applications.

**Methods**

**MBE growth.** Thin film growth was performed using a custom-built ultrahigh vacuum MBE system. Semi-insulating etched Si(111), insulating heat-treated sapphire(0001) and SrTiO$_3$(111) substrates were outgassed before the growth of TI films. High-purity Bi(99.999%), Sb(99.9999%), and Te(99.9999%) were evaporated from Knudsen effusion cells, whereas the transition metal dopants Cr(99.999%) and
V(99.995%) were evaporated by e-guns. During the growth, the substrate was maintained at 230°C. The flux ratio of Te per Bi and Sb was set to approximately ~8 to prevent Te deficiency in the films. The Sb, Bi, and V concentration in the films were determined by their ratio obtained in situ during growth using separate quartz crystal monitors and later confirmed ex situ by inductively coupled plasma atomic emission spectroscopy (ICP-AES). The growth rate for the films was approximately 0.2 quintuple layers per minute. Epitaxial growth was monitored by in situ reflection high energy electron diffraction (RHEED) patterns, where the high crystal quality and the atomically flat surface were confirmed by the streaky and sharp “1×1” patterns (see Supplementary Information).

**Transport measurements.** The transport measurements were performed ex situ on the magnetically-doped TI thin films. To avoid possible contamination, a 10-nm thick epitaxial Te capping layer was deposited at room temperature on top of the TI films before taken out of the growth chamber for transport measurements (see Supplementary Information). The Hall effect and longitudinal resistance were measured using both a Quantum Design Physical Property Measurement System (PPMS) (50mK, 9T) and a dilution refrigerator (Leiden Cryogenics, 10mK, 9T) with the excitation current flowing in the film plane and the magnetic field applied perpendicular to the plane. The bottom gate voltage was applied using the Keithley 6430. All the resistance meters were calibrated by a standard resistor. The QAH results reported here have been reproduced on several samples using the above two cryostats.

**References**


18. Dyck, J. S. et al. Diluted magnetic semiconductors based on Sb$_{2-x}$V$_x$Te$_3$ (0.01≤x≤0.03). Phys. Rev. B 65, 115212 (2002).


22. $\rho_{xx}$ and $\rho_{xy}$ results shown in Fig. 1 were carried out with excitation current of 1 nA and with the mixing chamber of the dilution refrigerator anchored at 10 mK. However, systematic measurements of $\rho_{xx}$ as functions of temperature and (higher) excitation current indicate the electron temperature of the sample is ~25±5mK due to the stray electromagnetic noise.


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**Author contributions**

C. Z. C., M. H. W. C. and J. S. M. conceived and designed the research. C. Z. C. grew the material with the help of J. S. M.. C. Z. C. performed characterization studies of the samples with the help of B. A. A. and D. H.. W. Z. made the devices and performed the transport measurements with the help of C. Z. C., D. K. and M. H. W. C.. C. X. L., H. J. Z. and S. C. Z. provided theory support. C. Z. C., C. X. L., and M. H. W. C. analyzed the data and wrote the manuscript with contributions from all authors.

**Additional information**

Supplementary information is available in the online version of the paper. Reprints and permissions information is available at [www.nature.com/reprints](http://www.nature.com/reprints).

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**Competing financial interests**

The authors declare no competing financial interests.
Figures and Figure captions:

Figure 1 | The QAH effect in a 4QL \((\text{Bi}_{0.29}\text{Sb}_{0.71})_{1.89}\text{V}_{0.11}\text{Te}_3\) film (sample S1) measured at 25mK. a, Magnetic field dependence of the longitudinal resistance \(\rho_{xx}\) (red curve) and the Hall resistance \(\rho_{yx}\) (blue curve) at charge neutral point \(V_g=V_g^0\). b, c, Expanded \(\rho_{yx}\) (b) and \(\rho_{xx}\) (c) at low magnetic field. \(\rho_{xx}\) at zero magnetic field exhibits a value of \(1.00019\pm0.00069h/e^2\), while \(\rho_{yx}\) at zero magnetic field is only \(\sim0.00013\pm0.00007h/e^2\) (~3.35±1.76Ω).

Figure 2 | Ferromagnetic properties comparing Cr- and V-doped \(\text{Sb}_2\text{Te}_3\). a, The Curie temperature \(T_C\) of 6QL \(\text{Sb}_{2-x}\text{V}_x\text{Te}_3\) (blue circles) and \(\text{Sb}_{2-x}\text{Cr}_x\text{Te}_3\) thin films (red circles). Note the factor of 2 increase in \(T_C\) for V doping. b, The Hall traces of 6QL \(\text{Sb}_{1.87}\text{V}_{0.13}\text{Te}_3\) and \(\text{Sb}_{1.87}\text{Cr}_{0.13}\text{Te}_3\) thin films, measured at \(T=2K\). Note the order of magnitude increase in \(H_c\) for V doping.
Figure 3 | The self-driven QAH state in a different 4QL (Bi$_{0.29}$Sb$_{0.71}$)$_{1.89}$V$_{0.11}$Te$_3$ film (sample S2). a, Temperature dependence of longitudinal resistance $\rho_{xx}$ (red curve) and Hall resistance $|\rho_{yx}|$ (blue curve) of sample S2 without magnetic training. Inset: $T$-dependent $\rho_{xx}$ (red curve) and $|\rho_{yx}|$ (blue curve) under magnetic field $\mu_0H=0$ (solid) and 2T (dashed). The opposite $T$-dependent behaviors of $\rho_{xx}$ and $|\rho_{yx}|$ reveal the QAH state at low $T$. b, c, The magnetic field-dependent $\rho_{yx}$ (b) and $\rho_{xx}$ (c) of sample S2 measured at 120mK. The red curves in b and c were obtained by first cooling the sample under zero-field and then measured from zero to higher field.