Magnetic quantum phase transition in Cr-doped Bi$_2$(Se$_x$Te$_{1-x}$)$_3$

driven by the Stark effect

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A. Experimental methods

The eight-quintuple-layer \((\text{Bi}_{0.89}\text{Cr}_{0.11})_2(\text{Se}_{x}\text{Te}_{1-x})_3\) films are prepared by using an ultrahigh vacuum system with a base pressure of \(8\times10^{-11}\) Torr, which consists of molecular beam epitaxy, scanning tunneling microscopy and angle resolved photoemission spectroscopy. Prior to sample growth, the \(\text{SrTiO}_3(111)\) substrates are first degassed at 550°C for 10 minutes and then heated at 650°C for 25 minutes. High purity Bi (99.9999%), Cr (99.999%), Se (99.999%) and Te (99.9999%) are evaporated from standard Knudsen cells following the same procedure in our previous report\(^1\). A real-time reflection high energy electron diffraction (RHEED) intensity oscillation measured on the (00) diffraction is used to calibrate the growth rate, which is controlled at a typical value of \(~0.125\) QL/minute.

Before being taken out of the growth chamber, the samples are covered with a 20 nm Te capping layer to avoid contamination. Thin films for transport measurements have a typical size of \(2\text{mm} \times 5\text{mm}\), which can be manually scratched into a Hall bar geometry like the one used in a previous report\(^2\). The size of the Indium contacts for voltage probe is around 500 \(\mu\text{m}\). A 17 Hz AC current with amplitude of 10 \(\mu\text{A}\) is used for the Hall effect measurements. The Hall signal is anti-symmetrized with respect to the magnetic field to remove the pickup of longitudinal resistance due to the unavoidable misalignment of the contacts.

B. Temperature-dependent anomalous Hall effect

Figure S1 shows the temperature-dependent anomalous Hall effect measured on these five samples. The two-dimensional Hall resistivity \(R_{yx}\) can be expressed as: \(R_{yx} = R_A M + R_N H\), where \(M\) is the sample magnetization, \(R_A\) and \(R_N\) are the anomalous and normal Hall coefficients respectively\(^3\). In eight-quintuple-layer \((\text{Bi}_{0.89}\text{Cr}_{0.11})_2\text{Te}_3\) \((x = 0\) sample\), the anomalous Hall curve shows nearly square hysteresis loop with positive jump at \(T = 1.5\) K, indicating ferromagnetic ground state. The loop shrinks and coercive field decreases with increasing temperature and finally disappears at 20 K.

On the other hand, the Hall curve of eight-quintuple-layer \((\text{Bi}_{0.89}\text{Cr}_{0.11})_2\text{Se}_3\) \((x = 1\) sample\) develops a negative curvature at low magnetic field, which is gradually smeared out by increasing temperature. This behaviour is the typical signature of anomalous Hall effect in paramagnetic
materials. The isovalent Se substitution of Te systematically reduces the effective spin-orbit coupling strength and drives the observed topological-magnetic phase transition, which is accompanied by the sign reversal of anomalous Hall effect\(^1\).

![Graph showing temperature-dependent anomalous Hall effect for five samples at V\(_g\) = 0 V.](image)

**Supplementary Figure 1** | Temperature-dependent anomalous Hall effect for five samples at \(V_g = 0\) V. The topology driven magnetic quantum phase transition is observed by varying the Se/Te ratio. The data for \(x = 0\) sample is divided by 3 for clarity.

**C. Angle resolved photoemission spectroscopy measurements**

The *in situ* angle resolved photoemission spectroscopy measurements are carried out at \(T = 120\) K. The energy resolution of the Scienta R2002 electron energy analyzer is set at 15 meV. Photon source is Helium discharge lamp with photon energy \(h\nu = 21.218\) eV. The experimental setup of angle resolved photoemission spectroscopy measurements can also be found in our previous report\(^4\). Before angle resolved photoemission spectroscopy measurements, 300 nm thick Ti is evaporated on the edges of the substrate to get rid of charging effect induced by the insulating SrTiO\(_3\) substrate. All the spectra in the paper are taken along the K-\(\Gamma\)-K direction.

Figure S2 shows that the \(x = 0\) and \(x = 0.52\) samples have well-defined Dirac surface states originated from the inverted bulk band structure. As Se concentration is increased to \(x = 0.86\), the Dirac surface states disappear and a trivial gap is opened at the \(\Gamma\) point, indicating a topological phase transition from topologically nontrivial phase to topologically trivial phase. The gap size is further enhanced by increasing Se content to \(x = 1\). The Fermi level \(E_F\) cuts deep into the bulk conduction band for Se content \(x \geq 0.52\), which is consistent with the large carrier density observed in these samples. We note that in the \(x = 0\) sample, only surface states are observed. According to first principle calculations\(^5\), the bulk states of pure Bi\(_2\)Te\(_3\) are mainly located at the \(\Gamma\)-M direction,
whereas the angle resolved photoemission spectroscopy measurements here are taken along the Γ-K direction.

Supplementary Figure 2 | Angle resolved photoemission spectroscopy measurements on eight-quintuple-layer $(Bi_{0.89}Cr_{0.11})_2(Se_{x}Te_{1-x})_3$ films with $x = 0, 0.52, 0.67, 0.86$ and 1. Well-defined topological surface states are clearly shown in the $x = 0$ and $x = 0.52$ sample. As Se concentration is increased to $x = 0.86$, the Dirac surface states disappear and a trivial gap is opened at the Γ point, indicating a topological phase transition from topologically nontrivial phase to topologically trivial phase. The gap size is further enhanced by increasing Se content to $x = 1$. The Fermi level $E_F$ cuts deep into the bulk conduction band for Se content $x \geq 0.52$, which is consistent with the large carrier density observed in these samples.

**D. Gate-tuned carrier density**

The normal Hall coefficient $R_N$ can be obtained from the slope of the anomalous Hall curve and the nominal carrier density can be calculated as $n_H = 1/R_N$. As shown in Fig. S3, at $T = 1.5$ K the Hall slopes between 0.5 T and 1 T are almost parallel at different gate voltages for the $x = 0.67$ sample, indicating the gate injected carrier density is negligible compared to the large bulk carriers. The carrier density is estimated by linear fit to the Hall curve between 0.5 T and 1 T. For $x = 0.67$ sample, the carrier density is $1.02 \times 10^{14} cm^{-2}$ at $V_g = -210$ V and $1.26 \times 10^{14} cm^{-2}$ at $V_g = +210$ V.

For the sample in ferromagnetic phase ($x = 0.52$), the carrier density changes from $1.25 \times 10^{14} cm^{-2}$ to $1.42 \times 10^{14} cm^{-2}$ when gate voltage is swept from $V_g = -210$ V to $V_g = +210$ V. Similarly, the carrier density of paramagnetic sample ($x = 0.86$) is $1.61 \times 10^{14} cm^{-2}$ and $1.85 \times 10^{14} cm^{-2}$ at two limiting gate voltages. Therefore, for all the three samples, the carrier density
variation between $V_g = \pm 210 \text{ V}$ is around $2 \times 10^{13} \text{ cm}^{-2}$, which is consistent with a previous report\textsuperscript{6}. However, here the magnetic phase transition is only observed for the $x = 0.67$ sample near topological quantum critical point.

\textbf{Supplementary Figure 3 | Gate dependence of anomalous Hall effect for three samples at $T = 1.5 \text{ K}$}. The Hall slopes are almost parallel in different gate voltages in all three samples, indicating the gate injected carrier density is negligible compared to the large bulk carriers.

\textbf{E. Carrier-dependent ferromagnetism}

In a magnetic topological insulator, both surface\textsuperscript{7} and bulk charge carriers\textsuperscript{8-10} can mediate ferromagnetism via the Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism. Surface Dirac fermion mediated ferromagnetism is proposed by Liu \textit{et al.} (ref. 11), which states that ferromagnetic order becomes stronger when the Fermi level $E_F$ is moved towards the Dirac point due to the increase of Fermi wave length $\lambda_F \sim 1/2k_F$, where $k_F$ is Fermi wave vector. This novel ferromagnetism is verified experimentally in gated Mn-doped Bi$_2$Te$_3$$_y$Se$_y$ devices\textsuperscript{7}. It was found that the ferromagnetic order is weakened with increasing surface carrier density, and vanishes completely when the Fermi level $E_F$ is tuned into the bulk conduction band at a critical carrier density around $4.3 \times 10^{13} \text{ cm}^{-2}$. This trend is consistent with the theoretical proposal of ferromagnetic order mediated by surface Dirac fermion. The eight-quintuple-layer (Bi$_{0.89}$Cr$_{0.11}$)$_2$(Se$_x$Te$_{1-x}$)$_3$ films studied are in the heavily electron doped regime with carrier density on the order of $1 \times 10^{14} \text{ cm}^{-2}$. In this system, Fermi level $E_F$ lies deep in to bulk conduction band (Fig. S2) and the transport is dominated by the bulk states. Based on the experimental data and
theoretical paper published previously, in this regime the surface Dirac fermion mediated ferromagnetism becomes irrelevant. Moreover, for the sample at the verge of topological phase transition, the penetration depth of Dirac surface states increases and diverges at the critical point\textsuperscript{12}. Therefore, the topological surface states are severely weakened near the topological phase transition and thus the Dirac surface state mediated ferromagnetism cannot explain the electrically tuned magnetic phase transition in the $x = 0.67$ sample.

The second carrier mediated mechanism is the bulk RKKY effect, as has been demonstrated in Cr doped Sb$_2$Te$_3$ (ref. 13) and Cr doped (Bi,Sb)$_2$Te$_3$ (refs 9,10). In this picture long-range ferromagnetic order is strengthened by increasing bulk carrier density. As shown in Fig. 3a and Fig. 3b in our main text, long-range ferromagnetic order in our sample is induced by negative gate voltage, which injects holes hence reduces the total electron-type carrier density (carrier density is $1.02 \times 10^{14} \text{cm}^{-2}$ at $V_g = -210$ V and $1.26 \times 10^{14} \text{cm}^{-2}$ at $V_g = +210$ V). Therefore, the gate-voltage dependence of the ferromagnetic order is totally opposite to that induced by bulk RKKY effect. Similarly, the observation that the $x = 0.67$ sample (with carrier density $1.02 \times 10^{14} \text{cm}^{-2}$ at $V_g = -210$ V) is ferromagnetic, whereas the $x = 0.86$ sample (with carrier density $1.61 \times 10^{14} \text{cm}^{-2}$ at $V_g = -210$ V) is paramagnetic again demonstrates that bulk RKKY is not essential for the ferromagnetic order in our system. Therefore, the bulk carrier density variation induced by gating of SrTiO$_3$ substrate is not the origin for the observed electrically tuned magnetic phase transition either.

F. Arrott plot of anomalous Hall effect for the $x = 0.67$ sample

The Arrott scaling analysis can accurately determines the Curie temperature $T_C$ of a ferromagnetic material\textsuperscript{14}, in which $M^2$ is plotted against the ratio of $\mu_0 H/M$ ($\mu_0 H$ is the external magnetic field, $M$ is the sample magnetization). In order to investigate the Stark effect, the films we studied are only eight quintuple layers so that direct magnetization measurements are impossible. However, the total two-dimensional Hall resistance in a ferromagnetic material can be expressed as $R_{yx} = R_s M + R_N H$, where the first term represents the anomalous Hall effect caused by the spontaneous magnetization and the second term is the normal Hall resistance\textsuperscript{3}. The common practice in experimental studies of magnetic thin films is to use the total Hall resistance in the low magnetic field range for the Arrott plot, in either metals\textsuperscript{15} or semiconductors\textsuperscript{16}, where the normal
Hall resistance is negligible small. Here the Arrott analysis is performed by using Hall resistance both before and after the normal Hall resistance is subtracted.

Figure S4 shows the Hall resistance before and after the subtraction of normal Hall resistance measured at $T = 1.5 \text{ K}$. The normal Hall coefficient $R_N$ is roughly estimated from Hall slope between 0.5 T and 1 T. The positive Hall hysteresis loop and negative Hall curvature at low magnetic field are nearly insensitive to the subtraction, indicating the normal Hall resistance is not important at low magnetic field. It should be noted that the subtracted Hall resistance at $V_g = -25 \text{ V}$ quickly saturates at $\mu_0H = 0.13 \text{ T}$ with a plateau resistance around $\sim 1.3 \Omega$. According to Arrott’s original theoretical paper, Arrott plot should be under the condition that $M \ll M_0$, so that the Taylor expansion can be performed\textsuperscript{14}, where $M_0$ is the spontaneous magnetization at zero temperature. Therefore, $\mu_0H$ should be significantly lower than 0.13 T when using subtracted Hall resistance to perform Arrott analysis.

Supplementary Figure 4 | Hall resistance before and after the subtraction of normal Hall resistance for the $x = 0.67$ sample at varied gate voltages at $T = 1.5 \text{ K}$. The positive Hall hysteresis loop and negative Hall curvature at low magnetic field are insensitive to the subtraction, indicating the normal Hall resistance is not important at low magnetic field.
Here for the purpose of direct comparison, the Arrott plot is presented in the form: \( R_{yx}^2 = a + bH/R_{yx} \) (red solid square) and \( (R_{yx} - R_0H)^2 = a + bH(R_{yx} - R_0H) \) (blue solid square) in Fig. S5. The Hall resistance is using the data with magnetic field \( \mu_0H \) below 0.1 T. The linear fit in Arrott plot between 0.05 and 0.1 T gives the intercept \( a \). The spontaneous Hall resistance \( R_{yx S} \), which characterizes the spontaneous magnetization \( M_S \) is estimated by the square root of the intercept \( a \). In a ferromagnetic (paramagnetic) state, the intercept \( a \) is positive (negative)\(^{14-16} \). As shown in Fig. S5, the intercept at \( V_g \leq -100 \) V is always positive regardless of whether the normal Hall resistance is subtracted, indicating ferromagnetic ground state. On the other hand, the negative intercept at \( V_g \geq -50 \) V is also independent of the subtraction of normal Hall resistance, indicating paramagnetic ground state. The robust sign reversal of Arrott intercept clearly demonstrates the magnetic phase transition observed here, again indicating the normal Hall resistance is not important at low magnetic field.

Supplementary Figure S5 | The Arrott plots with and without the subtraction of normal Hall resistance for the \( x = 0.67 \) sample at varied gate voltages at \( T = 1.5 \) K. The intercept at \( V_g \leq -100 \) V is always positive regardless of the subtraction, indicating a ferromagnetic ground state. On the other hand, the negative intercept at \( V_g \geq -50 \) V is also independent of the subtraction of normal Hall resistance, indicating a paramagnetic ground state. The robust sign reversal of Arrott intercept clearly demonstrates the magnetic phase transition tuned by external gate voltage.
Figure S6a summarizes $R_{xy}^S$ for the $x = 0.67$ sample as a function of $V_g$ measured at $T = 1.5$ K. $R_{xy}^S$ shows positive values for $V_g \leq -100$ V before and after the subtraction of the normal Hall resistance and becomes zero for $V_g \geq -50$ V. The value $V_g = -50$ V lies in the vicinity of the magnetic phase transition, and the zero field Hall resistance $R_{xy}^0$ takes still a small positive value when the magnetic field is reduced from positive to zero. However, the Arrott intercept at $V_g = -50$ V (Fig. S5) is already negative and thus we take the spontaneous Hall resistance $R_{xy}^S$ as zero. Figure S6b displays the coloured contour plot of the anomalous Hall resistance measured at $T = 1.5$ K, which separates the $H-V_g$ phase diagram into two distinct regimes. The positive anomalous Hall effect is always associated with the ferromagnetic state while the negative anomalous Hall effect only exists in the paramagnetic regime.

Supplementary Figure 6 | Magnetic phase transition revealed by spontaneous Hall resistance at varied gate voltages in the $x = 0.67$ sample. a, Spontaneous Hall resistance before and after subtraction of the estimated normal Hall resistance clearly demonstrates the gate-tuned magnetic phase transition at $T = 1.5$ K. b, Contour plot of the total anomalous Hall resistance as a function of gate voltage and magnetic field at $T = 1.5$ K. Positive anomalous Hall effect is associated with ferromagnetic order while negative anomalous Hall resistance exists in the paramagnetic regime.

Similarly, when the ferromagnetic state is approached from above Curie temperature $T_C$, the intercept $a$ changes sign from negative to positive. Figure S7a displays the temperature dependence
of Arrott plots for the anomalous Hall curves measured at $V_g = -210$ V using raw data. At $T = 1.5$ K and 3 K, the linear fit to the Arrott plot yields positive $a$, consistent with the behaviour of ferromagnetic state. The negative intercept at 5 K indicates the paramagnetic ground state. The spontaneous Hall resistance $R_{yx}^S$ decreases to zero at 5 K, indicating the Curie temperature $T_C$ at $V_g = -210$ V is between 3 K and 5 K. We noted that the dielectric constant of SrTiO$_3$ decreases with increasing temperature, which affects the electric field across the sample.

**Supplementary Figure 7 | Estimation of the Curie temperature $T_C$ by Arrott plot.**

*a*, At $T = 1.5$ K and 3 K, the linear fit to the Arrott plot yields positive $a$, consistent with the behaviour of ferromagnetic state. The negative intercept at 5 K indicates the paramagnetic ground state. 

**b**, The spontaneous Hall resistance $R_{yx}^S$ decreases to zero at 5 K, indicating the Curie temperature $T_C$ at $V_g = -210$ V is between 3 K and 5 K.

**G. Calculated electric potential energy distribution**

Angle resolved photoemission spectroscopy measurements shown in Fig. S2 on the $x = 0$ and $x = 0.52$ sample clearly show well-defined gapless Dirac surface states with linear dispersion. The topological surface states on the top and bottom surfaces for these two samples are totally decoupled at eight quintuple layers (ref. 17). In this condition, these samples are treated as three-dimensional systems. However, as the sample approaches the topological quantum critical point from the topologically nontrivial regime, the inverted band gap reduces and the penetration depth of topological surface states on both surfaces increases\textsuperscript{12}. Eventually, the penetration depth will exceed the thickness of the sample when the inverted band gap reduces. Furthermore, at the
topological quantum critical point where the inverted band gap is sufficiently small, the penetration depth of surface states will diverge. Under this circumstance, for the sample at the verge of topological phase transition, the top and bottom surface states will hybridize and open a surface gap. Effectively, these gapped surface states are no longer reside at the surfaces, but merge into the bulk. Therefore, this sample now can be considered as a two-dimensional system. In our theoretical calculations, we consider the sample near the quantum critical point as a quasi-two-dimensional system with two-dimensional subbands since our sample is only 8-nm thick. In this sample, the topological surface states will hybridize and open a gap at the Dirac point.

We employ a Poisson-Schrodinger equation to calculate self-consistently the electric potential energy distribution $V(z)$ where $0 \text{ nm} \leq z \leq 8 \text{ nm}$ is the out-of-plane direction coordinate inside the thin film. We first find all the electronic wave functions by diagonalizing the total Hamiltonian. The Model Hamiltonian is expressed as: $H = \sum_k c_k^\dagger h(k) c_k + V(z) c^\dagger(r)c(r)$ with kinetic energy part:

$$h(k) = \begin{pmatrix} \epsilon(k) + M(k) & A_2(k_x - ik_y) & 0 & A_1 k_z \\ A_2(k_x + ik_y) & \epsilon(k) - M(k) & -A_1 k_z & 0 \\ 0 & -A_1 k_z & \epsilon(k) + M(k) & A_2(k_x + ik_y) \\ A_1 k_z & 0 & A_2(k_x - ik_y) & \epsilon(k) - M(k) \end{pmatrix},$$

where $\epsilon(k) = D_0 + D_1 k_z^2 + D_2 (k_x^2 + k_y^2)$ and $M(k) = B_0 + B_1 k_z^2 + B_2 (k_x^2 + k_y^2)$. The parameters comes from interpolation of previous first principles calculations:\(^5\):

$D_0 = 0.130 eV, \quad D_1 = -0.105 eV \cdot nm^2, \quad D_2 = -0.0473 eV \cdot nm^2,$

$B_0 = -0.228 eV, \quad B_1 = 0.180 eV \cdot nm^2, \quad B_2 = 0.494 eV \cdot nm^2,$

$A_1 = 0.0786 eV \cdot nm, \quad A_2 = 0.335 eV \cdot nm$

For sample thickness $d$, the potential energy $V(z)$ is a function of $z \in [0, d]$ satisfying

$$\frac{dV(0)}{dz} = 0, \quad \frac{dV(d)}{dz} = q_e E,$$

where $q_e$ is the electron charge, $E$ is the electric field induced by the external gate voltage. $V(z)$ is calculated self-consistently via the Poisson equation.

Under a fixed Fermi level $E_F$, the charge density distribution $\rho(z)$ can then be calculated by adding up probabilities of occupied electron wave functions at $z$. Solving the Poisson equation
∇^2V = −ρ/ε_T ε_0 with this ρ(z) in turn gives the potential energy V(z). When the iteration procedure converges, we plot the potential energy V(z). By tuning the Fermi level E_F in the calculation, we are able to vary the electric field in a wide range. As shown in Fig. S8, the calculated electric energy potential distribution for eight-quintuple-layer topological insulator thin film along z-axis is non-parabolic, due to the existence of charge carriers.

Supplementary Figure 8 | Calculated potential energy distribution within the sample under three electric field. The distribution is non-parabolic due to the existence of charge carriers.

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