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Imaging the Breakdown and Restoration of Topological Protection in Magnetic Topological Insulator MnBi2Te4

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1	Imaging the breakdown and restoration of topological protection in
2	magnetic topological insulator MnBi <sub>2</sub> Te <sub>4</sub>
3	
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21	Abstract –
22	Quantum anomalous Hall (QAH) insulators transport charge without resistance along
23	topologically protected chiral one-dimensional edge states. Yet, in magnetic topological
24	insulators (MTI) to date, topological protection is far from robust, with zero-magnetic field QAH
25	effect only realised at temperatures an order of magnitude below the Néel temperature $T_{\rm N}$ ,
26	though small magnetic fields can stabilize QAH effect. Understanding why topological
27	protection breaks down is therefore essential to realising QAH effect at higher temperatures.
28	Here we use a scanning tunnelling microscope to directly map the size of exchange gap $(E_{g,ex})$
29	and its spatial fluctuation in the QAH insulator 5-layer MnBi <sub>2</sub> Te <sub>4</sub> . We observe long-range
30	fluctuations of $E_{g,ex}$ with values ranging between 0 (gapless) and 70 meV, appearing to be

uncorrelated to individual surface point defects. We directly image the breakdown of topological protection, showing that the gapless edge state, the hallmark signature of a QAH insulator, hybridizes with extended gapless regions in the bulk. Finally, we unambiguously demonstrate that the gapless regions originate from magnetic disorder, by demonstrating that a small magnetic field restores E g,exin these regions, explaining the recovery of topological protection in magnetic fields. Our results indicate that overcoming magnetic disorder is key to exploiting the unique properties of QAH insulators.

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#### 40 Main Text-

Topological protection has become a crucial concept in the recent development of condensed matter 41 physics<sup>1, 2, 3, 4</sup>. In the quantized versions of the Hall effect (QHE), spin Hall effect (QSHE) and 42 43 anomalous Hall effect (QAHE), topological protection manifests as one-dimensional electronic edge states where scattering due to local perturbations is prohibited<sup>5</sup>. This opens the way towards high-44 temperature lossless electronic transport applications<sup>6,7</sup> as well as new approaches to topologically-45 protected fault-tolerant quantum computing<sup>8, 9</sup>. These technologies require robust topologically 46 47 protected edge channels, but in electronic devices this protection is often observed to break down. Breakdown of the QHE due to disorder, temperature, and current has been understood within scaling 48 theory<sup>10</sup>, and was a fundamental development in the understanding of continuous quantum phase 49 transitions<sup>11</sup>. The microscopic origins of disorder-induced QHE breakdown are still a vibrant area of 50 investigation, with new developments in graphene showing unique aspects of backscattering in the 51 presence of both electron- and hole-like edge channels<sup>12, 13</sup>. The QSHE is robust to non-magnetic 52 disorder at zero temperature<sup>14</sup>, but magnetic disorder can cause scattering at finite temperatures<sup>15</sup>, with 53 a quantum phase transition from helical liquid to insulator under strong interactions<sup>14</sup>. In contrast, the 54 chiral quantum anomalous Hall (QAH) edge channel supports unidirectional flow of electrical current 55

that should be robust to any potential perturbations smaller than the exchange energy gap  $E_{g,ex}$  which is opened in the surface-state of a thin film 3D topological insulator (TI) via long-range magnetic order. <sup>16, 17</sup> Despite this, breakdown of topological protection is ubiquitously observed at far lower temperatures than  $E_{g,ex}/k_B$  or  $T_N$ , where  $k_B$  is the Boltzmann constant. In dilute magnetic doped topological insulators, this is thought to be due to magnetic disorder, leading to non-uniform magnetization and a fragile QAHE that is only observable at extremely low temperatures (<1 K).<sup>18, 19</sup>

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Intrinsic stoichiometric magnetic topological insulators (MTIs) which possess both non-trivial 63 topology and intrinsic magnetism, for example MnBi<sub>2</sub>Te<sub>4</sub><sup>20, 21</sup> should in principle circumvent the issues 64 associated with dilute magnetic doping. Promisingly, odd-layers of MnBi<sub>2</sub>Te<sub>4</sub> in the 2D limit host 65 quantum anomalous Hall states <sup>22</sup>, leading to observation of the QAHE at 1.4 K, with the temperature 66 increasing to 6.5 K under application of an external magnetic field<sup>23</sup>. Yet, this value is still substantially 67 lower than  $T_{\rm N} = 25$  K and the activation energy extracted from transport measurements of  $\Delta E = 0.64$ 68 meV is two orders of magnitude smaller than the predicted  $E_{g,ex} = 70$  meV (800K). Furthermore, 69 QAHE is not routinely observed in ultra-thin odd-layer MnBi<sub>2</sub>Te<sub>4</sub> samples or quantization is only 70 observable in a large perpendicular magnetic field<sup>24</sup>. These results hint at the presence of various types 71 of surface disorder that act to suppress the  $E_{g,ex}^{25}$  and destroy QAHE. 72

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To understand the mechanism of topological breakdown requires direct measurement of the interplay between surface disorder, local fluctuations in  $E_{g,ex}$ , and the chiral edge state with atomic-scale precision using low-temperature scanning tunneling microscopy and spectroscopy (STM/STS). A technique previously used to probe band gap fluctuations and edge states in other 2D materials<sup>2,18,28-<sup>31</sup>. To date, most STM/STS measurements on MnBi<sub>2</sub>Te<sub>4</sub> have been performed on bulk crystals and have focused on point defects<sup>26, 27, 28</sup>. Little attention has been paid to ultra-thin films of MnBi<sub>2</sub>Te<sub>4</sub> and the mechanisms of topological protection breakdown and suppression of QAHE. A recent report</sup> suggests connection between local magnetic  $Mn_{Bi}$ ,  $Bi_{Mn}$  anti-site defects (notation  $X_Y$  means a X ion replaces a Y ion in the lattice) and collapse of the Dirac mass gap in high defect regions, but did not measure bandgap fluctuations over large areas to understand possible short-range behavior from magnetic disorder, how disorder interacts with the chiral edge state <sup>29</sup> or how the disorder effects respond to a magnetic field. Thus, the mechanism by which topological protection is destroyed, as well as how it recovers in *B* field, are still not understood.

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In this work, we utilize magnetic field STM/STS to study the origin of QAHE suppression in 5 septuple layer (SL) MnBi<sub>2</sub>Te<sub>4</sub>. We directly measure spatial fluctuation of  $E_{g,ex}$  and importantly observe the electronic overlap of the gapless edge state with gapless metallic bulk states, providing the route to breakdown of the QAHE. Finally, we demonstrate that by applying a magnetic field well below the spin-flop transition, we are able to restore the magnetic gap in the gapless regions, explaining the recovery of QAHE in small magnetic fields.

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95 Fig. 1(a)-(b) presents the crystal structure of one septuple layer (SL) MnBi<sub>2</sub>Te<sub>4</sub>. Lattice constants, magnetic moments and possible lattice defects are labelled. Within each SL, intra-layer Mn<sup>2+</sup> ions are 96 coupled through ferromagnetic interaction. Between two adjacent SLs, two Mn<sup>2+</sup> atomic layers are 97 coupled through anti-ferromagnetic (AFM) interaction, resulting in thickness-dependent magnetic 98 99 properties. We grow high-quality epitaxial ultra-thin MnBi<sub>2</sub>Te<sub>4</sub> using molecular beam epitaxy (MBE) 100 on Si(111)–7×7. Due to interfacial charge transfer as a result of the different work functions of p- and 101 *n*-type silicon (111) the doping level in  $MnBi_2Te_4$  films can be tuned. This allows for STM/STS measurements performed on MnBi<sub>2</sub>Te<sub>4</sub> on *p*-type Si(111) with a Fermi energy that sits in the Dirac 102 103 gap, whilst for ARPES measurements performed on  $MnBi_2Te_4$  on *n*-type Si(111) the films are electron-104 doped allowing the Dirac electron band to be observed. See Methods for growth details, whilst 105 structural characterization as well as the role of substrate and doping are found in Supplementary 106 Information S1 and S2. Supplementary Fig. S1(b) shows a typical large-area STM topography scan 107 with coexisting regions of 4 and 5 SL MnBi<sub>2</sub>Te<sub>4</sub> islands that are atomically flat, along with small 108 pinholes of bare substrate. Fig. 1(c) shows a 20×20 nm atomic resolution STM image revealing the expected 1×1 atomic surface structure with lattice constant 4.3 Å. Several different defects are present; 109 110 bright spots on the surface correspond to negatively charged Bi<sub>Te</sub> point defects whilst the dark triangles are Mn<sub>Bi</sub> defects, similar defects have been observed in Cr, Mn-doped 3D TIs<sup>19, 30, 31</sup>. The third defect 111 112 type - Bi<sub>Mn</sub> (located in the middle of each SL) is not directly visible in (c) but presented in 113 Supplementary Fig. S3.

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**Fig. 1(d)** shows an ARPES spectrum of 5 SL MnBi<sub>2</sub>Te<sub>4</sub> thin film grown on *n*-type Si(111) taken at 8 K along ΓM direction where a Dirac cone is clearly visible near the Fermi level. The strong spectral weight near Γ in the Dirac point region could be due to Te-orbital-related matrix elements<sup>32, 33</sup> or be the result of bandgap fluctuations as the spectral signal is averaged over the beam spot size (100×100 µm). To demonstrate this possibility we fit the ARPES spectrum in **Fig. 1(d)** with three possible scenarios (red curve: full band gap  $E_{g,ex} = 70$  meV extracted from energy distribution curve analysis<sup>32</sup>, green curve: reduced  $E_{g,ex} = 45$  meV, blue curve: gapless  $E_{g,ex} = 0$  meV limit).

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123 To properly understand the  $E_{g,ex}$  distribution, we use STS to measure the dI/dV spectrum (the 124 differential conductance dI/dV as a function of sample bias V) which is proportional to the local density 125 of states (LDOS) at energy  $E_F + eV$ , where e is the elementary charge. Fig. 1(e) shows three typical STS from the same SL terrace. The size of  $E_{g,ex}$  changes drastically with location: the red STS curve 126 127 corresponds to  $E_{g,ex} = 70$  meV, the green curve shows a reduced band gap  $E_{g,ex} = 45$  meV, and the blue 128 curve is consistent with a gapless ( $E_{g,ex} = 0 \text{ meV}$ ) spectrum (see Fig. S4 and Fig. S5 for details on 129 extracting bandgap and the minimal tip-induced band bending). All spectra in Fig. 1(e) were taken 130 more than 5 nm away from step edges in order to exclude effects from edge states.

131 Before turning to the origin of the band gap variations, we probe the step edge between 4 and 5 SL 132 MnBi<sub>2</sub>Te<sub>4</sub>, to verify the presence of the conductive edge state, a consequence of topological protection and signature of a QAHI. In principle, the edge state exists at the edge of the sample where the height 133 134 profile on the sample drops from 5SL thickness to the underlying substrate. However, due to a variety 135 of growth and measurement practicalities, measuring the edge state on the sample edge with STM is extremely challenging. Therefore, instead we probe the edge state on the 5SL (Chern number of  $\pm 1$ ) 136 137 to 4SL (Chern number of 0) step edge enabled by the unique thickness-dependent topological property of MnBi<sub>2</sub>Te<sub>4</sub>.<sup>22</sup> Since the edge state appears on the boundary between two phases with different 138 139 topological invariant, the physics of the edge state on the terrace edge is equivalent to that of the edge 140 state on the sample edge.

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142 Figure 2(a) and (b) show STM topography and dI/dV maps taken across two different 4 to 5 SL step edges on two separately grown MnBi2Te 4thin films. Since the chiral edge state exists within the 143 Dirac band gap, the sample bias was tuned into the Dirac band gap to image the edge state. The lower 144 panel of Fig. 2(a) shows a dI/dV map taken at +25 mV: a pronounced increase in dI/dV signal is 145 observed that is localized at the step edge (which is marked by a red dashed line), indicating a 146 conductive edge state. This conductive edge state is mostly continuous, but has strong hybridization 147 between the edge state and disordered bulk states and not spatially isolated. The second edge 148 presented in Fig. 2(b) shows another step edge consisting a 5SL edge (orange arrow) and sub-step 149 150 edge (excluded in the figure). The sub-step edge structure has also been observed in previous STM study on MnBi<sub>2</sub>Te  $_{4}$ thin films<sup>29</sup>. The middle and lower panel of Fig. 2(b) show dI/dV maps taken 151 at 0 and -15 mV: that also show a pronounced increase in dI/dV signal that is localized at the step 152 edge. At bias outside the Dirac gap (-15 mV), this edge state is weakly coupled to bulk state with 153 finite spectral weight between the edge channel and the gapless bulk states. As the bias moves into the 154 155 Dirac gap (0 mV), the edge states become spatially isolated unlike the edge state presented in Fig. **2(a)**. We have included more dI/dV

156 maps at other voltage bias, see Fig.S6 to demonstrate the decoupled edge channel in the Dirac gap. 157 We must point out the spectral intensity along the edge in Fig. 2(b) seems to be disconnected, which has been also observed in previous reports on topological edge states of MnBi<sub>2</sub>Te<sub>4</sub><sup>34</sup>, its related 158 heterostructure with Bi<sub>2</sub>Te<sub>3</sub><sup>35</sup>, as well as the 2D topological insulator WTe 2<sup>36</sup> and may arise due to 159 local roughness and structural disorder along the edge. Furthermore, we have also performed a similar 160 161 dI/dV map on a 4SL to 3SL edge, and no spectral intensity is observed (Fig.S7), indicating the 1D edge state could be only present on odd-layer SL MnBi<sub>2</sub>Te 4that is buried underneath the step edge and 162 163 is very difficult to probe from the surface. For a perfect QAHI there should be a well-defined 164 suppression of the bulk LDOS within the bandgap, but we observe bulk regions well away from the edge that also show strong LDOS at the same energy, indicating the coupling between edge state and 165 166 disordered bulk metallic regions. To confirm that some disordered bulk regions are indeed metallic, in 167 Fig. 2(c) we measure dI/dV spectra corresponding to the edge state (red curve) at point A, normal bulk 168 states (purple curve) at point B, and disordered bulk states (black curve) at point C, D in Fig. 2(b). The normal bulk region right next to the edge shows the expected insulating behavior with E 169  $g_{ex} = 40 \text{ meV},$ 170 but the dI/dV spectra at both the edge and within these disordered bulk state regions are quite different, with states filling the entire bulk gap. The disordered bulk states have much stronger spectral intensity 171 above zero bias in the conduction band range and the STS curves resembles the DOS of a gapless 172 173 Dirac cone<sup>37</sup>. This indicates a continuous metallic percolative path for electron transport from the edges 174 through the bulk. Figure 2(d), (e) shows spatial dI/dV profiles as a function of distance away from the 175 edge measured along the two green lines in Fig. 2(b) (labelled as Cut 1 and Cut 2) that demonstrate the extended nature of the edge state feature along the step edge located at 5 nm, as marked by the red 176 arrow. The other spectral intensity between 0 to 2 nm is clearly separated from the edge state, and we 177 assign it to the disordered bulk states, marked by a white arrow. Figure 2(f) shows the summed dI/dV178 179 intensity (red curve extracted from Fig. 2(d); green curve extracted from Fig. 2(e)) within the bulk Dirac gap. Moving away from the edge shows the expected exponential decay for a 1D topologically 180

 $^{2,38}$ The extracted exponential decay lengths are  $0.82 \pm 0.38$  nm and  $1.22 \pm 0.41$ 181 non-trivial edge state. nm. Although the observed edge state appears to be isolated from the metallic bulk states, the small 182 183 spatial separation (less than 3 nm) implies fragile edge state conduction. The observation of a 1D edge state on two different step edges (Fig. 2 and additional data presented in Fig. S8), suggests the 1D 184 edge state exists on the 5SL to 4SL edge, independent of the exact edge structure and evidence of a 185 186 QAH gapless edge state. However, the presence of both disordered bulk state and edge state on the 187 5SL terrace suggests that the edge state hybridizes with the metallic regions formed by these disordered 188 bulk states, and these metallic regions represent continuous conductive pathways that guide the edge 189 state into the bulk, leading to the conductive breakdown of QAHE through dissipative bulk conduction 190 and resulting in non-perfect quantization of Hall conductance and non-zero longitudinal resistance.

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192 To investigate the origin of these metallic regions formed by disordered bulk states, we perform atomic resolution topography and STS maps around the magnetic defects MnBi and BiMn, which allow us to 193 extract maps of the spatial variation of  $E_{g,ex}$ , and the gap center energy  $E_c$  to determine the influence 194 each defect has on the electronic structure.  $E_c$  is equivalent to extrapolating the massive Dirac bands 195 196 linearly into the gap to obtain the Dirac point in the gapless limit. At locations where the Dirac bands 197 are gapped,  $E_c$  is a good measure of local doping shifts associated to the magnetic order. Bi<sub>Te</sub> defects, are non-magnetic, thus, are unlikely to result in fluctuations in the exchange energy gap  $E_{g,ex}$ . Figure 198 199 3(a) illustrates the position of a Mn<sub>Bi</sub> defect in the crystal lattice, and Fig. 3(b) shows an atomic 200 resolution image of 10×5 nm area (-500 mV, 3 nA) with MnBi defects marked in black triangles. The 201 Mn<sub>Bi</sub> defect density can be estimated to be around 6%, which is very similar to the defect concentration reported in previous study on MBE grown MnBi<sub>2</sub>Te<sub>4</sub> film<sup>29</sup> but is almost double the concentration 202 observed on the surface of a cleaved bulk MnBi<sub>2</sub>Te<sub>4</sub> crystal <sup>27</sup>. The substitution of Bi<sup>3+</sup> by Mn<sup>2+</sup> causes 203 contraction on the three neighboring surface Te atoms<sup>27</sup>. Fig. 3(c)-(d) are maps of  $E_{g,ex}$  and  $E_c$  extracted 204 205 from the dI/dV spectra on the same area in (b) (details found in Supplementary Fig. S4).

206 Representative dI/dV curves from different locations, marked with E generative generative different locations, marked with E generative different locations are plotted in 207 **Supplementary Fig. S9.** The Mn Bidefects on the surface (green triangles) are barely visible in the  $g_{exand} E_{cin}$  (c) and (d) respectively, with only small local decrease in E 208 maps of E gexand slight 209 increase in *n*-type doping (due to the negative charge of the Mn Bidefects) observed. Instead, the  $g_{ex}$  and E cappear to be correlated, and to be spatially coherent over length scales of at 210 fluctuations in *E* 211 least a few nanometers, i.e. larger than a single defect which is about 0.5 nm. Gap size histograms of Mn 212 Biregions and Mn Biexcluded regions are shown in Fig. 3(e). In regions without Mn Bidefects, the E 213 g.exhistogram is skewed towards larger gap size when compared to regions with Mn Bidefects, which can 214 be explained by effectively reduced exchange coupling between surface state and magnetic moments 215 due to the AFM interaction between the Mn Bidefects and Mn<sup>2+</sup>ions. Overall, the patterns of gapped and 216 gapless regions formed over several nanometers are very different from the band gap fluctuations in 217 dilute magnetic doped TIs<sup>19,31</sup>. In dilute magnetic doped TIs, the random distribution of antisites formed by 218 magnetic 3d transition metal ions is directly responsible for the fluctuation. But in the current case of 5SL 219 MnBi<sub>2</sub>Te 4thin film, the observed pattern in the E g,exmap appears to be not correlated to distribution of Mn 220 Bidefects, suggesting additional major contribution from magnetic disorder to the gap fluctuations. The 221 origin of the magnetic disorder can be complicated but is very likely to be defect driven. The local 222 deficiency of Bi *p*-states on Mn Bidefects could indirectly modulate the weakened intralayer ferromagnetic 223 interaction among Mn  $^{2+}$ magnetic moments through non- negligible *p*-*d* interactions, as reported by 224 recent magneto-optics and inelastic neutron scattering studies<sup>39,40</sup>. In Fig. 3(f), a Bi <sub>Mn</sub>defect in the 225 226 middle atomic layer is depicted, where the substitution results in the absence of magnetic moment. Such 227 defects manifest as large bright triangles and are only visible at positive bias as marked by purple triangles 228 in (g). The three bright dots in each triangle are due to Te p-orbitals on the surface responding to Bi 229 Mn defects<sup>27</sup>. d*I*/d*V* mapping was performed in the area marked by the yellow box in (g), with the band gap 230 and gap center maps shown in Fig. 3(h)-(i). The band gap map in (h) and the histogram in (j) show that the substitution of magnetic Mn <sup>2+</sup>ions at

231 Bi<sub>Mn</sub> defects renders the local lattice site non-magnetic, thus, gapless regions. However, regions well 232 away from the Bi<sub>Mn</sub> defect still display band gap fluctuation with significant weight of gapless states, 233 suggesting that Bi<sub>Mn</sub> defects alone do not result in extended regions of suppressed Dirac band gap and 234 metallicity in the bulk. The results in Fig. 3 therefore demonstrate the band gap and gap center 235 fluctuations cannot be explained entirely by local gap suppression by any of the three types of isolated point defects discussed above and imply the possibility that longer-ranged collective behavior of 236 237 magnetic disorder is responsible for the extended Dirac band gap suppression on the surface of 238 MnBi<sub>2</sub>Te<sub>4</sub>.

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240 To understand the origin of the observed large-scale band gap fluctuations, we measure STS mapping with and without magnetic field to examine how the extended gapless structures respond to  $B_{i}$  field. 241 242 Figure 4(a) shows STM topography of the atomically flat scan region at  $B_{\perp}=0$  T. (See Fig.S10 for the 243 topography scan taken at 0T and 1T where the map region has been carefully aligned using the defects). 244 In Fig. 4(b) STS curves taken at different locations show three types of behavior: gapless regions 245 (blue), gapped regions with fluctuating Dirac band gap (green and red) and regions where the Dirac 246 electron band is suppressed and manifests as an anomalously large bulk gap (orange). Such conduction band (CB) suppression has been previously observed in bulk MnBi<sub>2</sub>Te<sub>4</sub><sup>27</sup>. The diminished CB intensity 247 248 prevents us from extracting accurate values of  $E_{g,ex}$ , thus, the CB suppressed regions are masked in 249 black in the following gap maps and excluded in subsequent analysis. The CB suppressed regions are 250 identified by summing the STS intensity above +20 mV bias at each point and comparing to a summed threshold value of  $1.28 \times 10^{-11}$  for 0 T map and  $1.85 \times 10^{-11}$  for 1T map that are estimated from STS 251 252 curves with weak CB intensity and optimized. Figure 4(d) plots STS curves taken at the same location 253 (green circle in (a)) that is initially gapless at  $B_{\perp}=0$  T (blue curve) and  $B_{\perp}=1$  T (red curve). It is 254 immediately clear that a 1 T field is sufficient to restore  $E_{g,ex}$  to 40 meV with enhanced exchange coupling. Having observed magnetic field induced band gap modulation, we now perform dI/dV255

mapping (-150 mV, 0.4 nA) on an 80×80 point-mesh on the same 30×30 nm area in (a) at  $B_{\perp}=0$  T 256 257 (Fig. 4(c)) and  $B_{\perp}=1$  T (Fig. 4(e)). Histograms of  $E_{g,ex}$  with and without B field are shown in Fig. 4(f). These maps reflect the spatial fluctuation of  $E_{g,ex}$  over larger scale and will be used to investigate its 258 259 origin beyond point defects. The histogram in the upper panel of Fig. 4(f), shows prominent weighting for  $E_{g,ex} < 10$  meV, corresponding to a skewed normal distribution (skewness 0.91) with mean of 26.3 260 meV and standard deviation of 25.8 meV. Upon applying  $B_{\perp}=1$  T, the histogram in the lower panel of 261 Fig. 4(f) shows a significant reduction in  $E_{g,ex} < 10$  meV regions, and a gap opening and 262 263 renormalization that results in a near-normal distribution (skewness 0.06) with an increased mean of 264 44.3 meV and smaller standard deviation of 20.2 meV. A statistical analysis of regions that possess 265 unsuppressed CB intensity at both 0T and 1T is presented in the Supplementary Information in Fig. 266 S11. This shows the average band gap increases by 20.6 meV to a value of 37.8 meV in the 1T magnetic field and more bimodal distribution. Additionally, to reveal the gradual change of the  $E_{g,ex}$ 267 in low to medium magnetic field, we have performed magnetic field dependent mapping between 0 268 269 and 0.8 T on a different sample, see Fig. S12 where the majority of gap renormalization occurs at 0.8 270 T.

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We now consider possible origins of the extended suppressed gap structures. As recently observed in 272 magnetic force microscopy measurements<sup>41</sup>, whilst the bulk of MnBi<sub>2</sub>Te 4thin film remains AFM 273 274 coupled, the surface exhibits magnetic spin flops which could be enhanced by Bi Mndefects or changes to the size of the inter-layer van der Waals gap near the surface<sup>42</sup>. In our thin film MnBi<sub>2</sub>Te<sub>4</sub>samples, 275 276 we are able to align the magnetic moments at  $B_{\perp}=1$  T, much lower than required for inducing surface (2-3.5 T) and bulk spin flop (7.7 T) in previous work<sup>41,43,44</sup>, which indicates that surface spin flop has 277 negligible contribution to the exchange gap fluctuation observed. This suggests that there is significant 278 magnetic disorder most likely in the first SL, a magnetic uncompensated layer, that causes band gap 279 fluctuation on the nanometer scale<sup>25</sup>. Such magnetic disorder occurs in the Mn <sup>2+</sup>layer located at the 280

center of the top SLs, which is very hard to probe using STM. However, the exchange interaction of the Dirac states with Mn <sup>2+</sup>ions enables indirect mapping of such magnetic disorder based on the suppression of  $E_{g,ex}$  as seen in **Fig. 4(c)** and **(e)**.

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Fig. 4(g) illustrates the situation schematically. Magnetic disorder causes local suppression of the 285 exchange gap (blue gapless Dirac spectrum) in extended regions due to long-range exchange 286 287 interactions among local moments, while some regions retain partial (green band structure) or full (red 288 band structure) gaps. Application of  $B_{\perp}=1T$  (bottom) aligns the moments of magnetically disordered 289 regions, increasing the gap (red band structure). Interestingly, the B field also decreases the area of 290 suppressed CB regions, suggesting CB suppression is also related to disordered magnetic moments 291 beyond the influence of deficient Bi orbitals due to Mn<sub>Bi</sub> defects<sup>27</sup>. Finally, we propose an explanation 292 to the origin of the magnetic disorder. The prevalent band gap fluctuation observed implies weakened 293 inter-layer and intra-layer exchange interaction in 5 SL MnBi<sub>2</sub>Te<sub>4</sub>. Its low magnetic anisotropy energy 294 makes MnBi<sub>2</sub>Te<sub>4</sub> similar to a 2D Heisenberg magnet that does not sustain long-range ferromagnetic order<sup>40, 44, 45</sup>. Such weakened magnetic anisotropy makes the magnetic ordering more vulnerable to 295 magnetic defects, especially Bi<sub>Mn</sub>. With Mn<sup>2+</sup> ions replaced by non-magnetic Bi<sup>3+</sup> ions, exchange 296 coupling between intra-layer  $Mn^{2+}$  ions is weakened, depending on the concentration of such defects. 297 298 Although, the direct exchange coupling between MnBi and surface states is much weaker as discussed 299 in Fig. 3, Mn<sub>Bi</sub> can still cause local deficiency of Bi *p*-orbitals and indirectly influence the magnetic 300 moments in the middle of SL by p-d interaction. Therefore, the ferromagnetic configuration of Mn<sup>2+</sup> 301 ions could be canted and disordered in the presence of a large amount of anti-sites defects, resulting in 302 reduced magnetization and gapless spectra over extended areas. Similar mechanism has also been reported recently<sup>29</sup>. Lastly, we present an  $E_{g,ex}$  map of a region with larger amount of Bi<sub>Mn</sub> defects in 303 304 0 T and 1 T magnetic field in Fig. S13. This region is indeed mostly gapless. Upon applying the 1T 305 field, a significant reduction of gapless regions occurs (further details in Supplementary Fig. S13).

306 CB suppression is almost absent in this region, which can be explained by the large amount of  $Bi_{Mn}$ 307 defects that offers sufficient *p*-states to form Dirac bands.

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309 Using magnetic field STM/STS measurements, we have demonstrated that the gapless edge state in QAHI 5 SL MnBi<sub>2</sub>Te 4is directly coupled to extended percolating bulk metallic regions arising from 310 band gap fluctuations caused by magnetic surface disorder. By applying a magnetic field, the band gap 311 fluctuations can be greatly reduced, and the average exchange gap increased to 44 meV, close to 312 predicted values for 5 SL MnBi<sub>2</sub>Te  $4^{21,22,32}$ . These results provide insight on the mechanism of 313 topological breakdown and how it can be restored in a magnetic field<sup>23,24</sup>. Minimizing magnetic 314 disorder will be the key to realizing QAHE in not only odd-layer MnBi<sub>2</sub>Te 4ultra-thin films but also 315 other MTIs at elevated temperature in the future. The weak interlayer interaction and intralayer 316 317 ferromagnetic ground state close to instability limit in MnBi<sub>2</sub>Te 4makes it difficult to sustain longrange magnetic order especially with significant amount of anti-site defects, and improved MnBi<sub>2</sub>Te<sub>4</sub> 318 crystal or film growth alone may not be sufficient to fully mitigate magnetic disorder. Therefore, other 319 320 strategies such as heterostructure engineering MnBi<sub>2</sub>Te 4with other robust, highly anisotropic 2D ferromagnets <sup>46</sup>or ferromagnetic/topological insulators sandwich heterostructures <sup>47, 48, 49</sup> may be 321 required to achieve the robust topological protection required for next-generation lossless electronics 322 and topological quantum computing<sup>6,7,8,9</sup>. 323

324

#### 325 Methods

#### **326** Growth of Ultra-thin MnBi<sub>2</sub>Te<sub>4</sub> on Si(111)

Ultra-thin MnBi<sub>2</sub>Te<sub>4</sub> thin films were grown in a Scienta Omicron Lab 10 molecular beam epitaxy 327 (MBE) growth chamber. The Si(111) substrate was flash-annealed at 1180°C with direct current 328 329 heating to achieve an atomically flat  $(7 \times 7)$  surface reconstruction. Effusion cells were used to evaporate elemental Mn (99.9%), Bi (99.999%) and Te (99.95%). A quartz crystal microbalance was 330 used to calibrate rates before growth and reflection high-energy electron diffraction (RHEED) was 331 332 used to monitor the crystal growth in-situ. Each SL of MnBi<sub>2</sub>Te<sub>4</sub> was grown by first growing 1 333 quintuple-layer Bi<sub>2</sub>Te<sub>3</sub> followed by growing a bilayer MnTe in overflux of Te at 230 °C. 1 SL MnBi<sub>2</sub>Te<sub>4</sub> forms spontaneously by re-arranging MnTe layer into the middle of 1QL Bi<sub>2</sub>Te<sub>3</sub> similar to 334 MnBi<sub>2</sub>Se<sub>4</sub><sup>50</sup>. The growth time for each 1QL Bi<sub>2</sub>Te<sub>3</sub> and MnTe was calibrated from the oscillation of 335 336 the RHEED pattern. Then the process was repeated five times to reach the desired thickness and finished with a post-annealing process in Te flux for 10 min to improve crystallinity. The films were
subsequently capped with 10nm amorphous Te, to allow transfer in air to the STM chamber.

### 340 Scanning Tunneling Microscopy/Spectroscopy (STM/STS) Measurements

The capped films grown on boron doped silicon (111) (resistivity 0.1-0.2  $\Omega \cdot cm$ ) were transferred in 341 air to a Createc LT-STM chamber and were annealed in UHV at 290°C for 2.5 hours to remove the Te 342 343 capping before performing STM measurements at 4.3 K. A PtIr tip was prepared and calibrated using an Au (111) single crystal, confirming the presence of the Shockley surface state at -0.5 V and flat 344 345 LDOS near the Fermi level before all measurements. The STM differential conductance measurements 346 (dI/dV) were performed using standard lock-in method with 5 mV AC excitation voltage at 797Hz for Dirac gap mapping and 2mV AC excitation voltage at 797Hz for edge state mapping. Differential 347 348 conductance measurements were made under open feedback conditions with the tip in a fixed position 349 above the surface. For the magnetic field dependent STM/STS measurements, a magnetic field up to 350 351 1T was applied perpendicular to the sample.

### 352 Angle-resolved Photoemission Spectroscopy (ARPES) Measurements

ARPES measurements were performed at Beamline 10.0.1 at Advanced Light Source (ALS) in Lawrence Berkeley National Laboratory, USA. A 5 SL MnBi<sub>2</sub>Te<sub>4</sub> sample was grown on antimony doped silicon (111) substrate (resistivity 0.1-0.2  $\Omega \cdot$  cm) following the same growth procedure in an MBE system integrated with the beamline endstation and transferred into the measurement chamber after growth. Data was taken using a Scienta R4000 analyser at 8K and photon energy of 50 eV was selected to optimize the signal. The combined energy resolution is 15-20 meV and the angular resolution is 0.2°, or equivalent to 0.01Å<sup>-1</sup> momentum resolution for the photon energy used.

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### 371 Author contributions

M. T. E and Q. L. devised the STM experiments. Q. L. performed the MBE growth and STM/STS
measurements at Monash University. I. D. B., B. L., L. W. and T.H.Y.V assisted the scanning probe
measurements. J. H., S.-K. M. and C. X. T assisted the ARPES measurements. Q. L. performed data
analysis with assistance from M. T. E, M. S. F, J. M and D. M. Q. Li, and M. T. E. composed the
manuscript. All authors read and contributed feedback to the manuscript.

### 378 Data availability

All raw and derived data used to support the findings of this work are available from the authors on reasonable request. 381 FIGURES

#### 

#### FIGURE 1





Figure 1 | Characterization of epitaxial ultra-thin MnBi<sub>2</sub>Te<sub>4</sub> and overall electronic structure from ARPES. a, b Crystal structures of a septuple layer of MnBi<sub>2</sub>Te<sub>4</sub>. (a) side view of the lattice with lattice constants, atom species, and defects labelled. The magnetic moments on Mn<sup>2+</sup> ions are marked with blue arrows. (b) Top view of the lattice. (c) Atomic resolution image (-2 V, 180 pA) of a flat 20×20 nm area where MnBi (dark triangles) and BiTe (bright dots) defects are clearly visible. The insert shows the fast Fourier transformed image of the same area. (Note the spots corresponding to  $1 \times 1$ surface atomic structure). (d) Angle-resolved photoemission spectrum of five-layer MnBi<sub>2</sub>Te<sub>4</sub> along  $\Gamma$ -M where the fully gapped ( $E_{g,ex} = 70 \text{ meV}$ ) band dispersion is marked by red curve. Green and blue are illustrations of possible reduced gap and gapless dispersions ( $E_{g,ex} = 35$  meV and 0 meV respectively). (e) dI/dV spectra taken at different locations on the same terrace of 5 SL MnBi<sub>2</sub>Te<sub>4</sub> (-0.2 V, 400 pA) showing gapless (blue curve), reduced gap (green curve) and fully gapped dI/dV curves from different regions on the same terrace.





Figure 2 | Visualizing the gapless edge state and its coupling to bulk metallic states. (a)-(b) STM topography taken at -1V (upper panels) and dI/dV maps (lower panels) taken across two different 4 to 5 SL MnBi<sub>2</sub>Te 4step edges. (a) dI/dV map at +25 mV bias (40 pA) shows a pronounced increase in intensity at the edge state and its strong coupling to bulk metallic states. The location of the edge state is marked by a red dashed line. (b) dI/dV maps (-0.15V, 400pA) at 0 mV and -15 mV bias show the spatial distribution of the edge state taken across a step edge and how the edge state is isolated from the bulk metallic states. The positions of the 5SL edge is marked with orange arrows. (c) dI/dVspectra (-0.15V, 400pA) taken from edge state region A (red circle), disordered bulk region C, D (black circle) and normal bulk region B (purple circle) as marked in (b). (d),(e) dI/dV spectra (-0.15V, 400pA) and height profile taken across the edge from paths marked by green lines in (b). The horizontal axis of the spectra is aligned with the height profile. The edge state is marked by red arrow and other in-gap peaks are attributed to disordered bulk states which is marked by white arrows. The white horizontal dashed line shows the range of Dirac gap which is the same as the shaded region in (c). (f) dI/dV intensity averaged in the Dirac gap and plotted as a function of distance in y direction showing exponential decay. The height profiles of the two cuts are shown below the dI/dV profiles respectively where the intensity maximum of the edge state is marked. 



453 Figure 3 | Local response of the exchange gap and doping to point defects. (a) Illustration of a 454 Mn<sub>Bi</sub> defect in the lattice. (b) Topography of a 10×5 nm area (-150 mV, 3 nA) with Mn<sub>Bi</sub> defects 455 manifesting as dark triangles (marked in black triangles). (c) An exchange gap,  $E_{g,ex}$ , map extracted from dI/dV spectra (-100 mV, 0.8 nA) on a 40×80 mesh for visualizing band gap fluctuation and (d) 456 gap center from the same region as (c). (e) Histograms of the  $E_{g,ex}$  extracted from regions with and 457 458 without  $Mn_{Bi}$  defects respectively. (f) Illustration of a  $Bi_{Mn}$  defect in the lattice. (g) Topography of a 459 40×40 nm area (+1.7 eV, 80 pA) with Bi<sub>Mn</sub> defects which manifest as bigger bright triangles (marked as purple triangles). Insert: a 5×5 nm region where dI/dV spectra (-100 mV, 0.91 nA) on a 50×50 460 mesh was taken to show its effect on  $E_{g,ex}$  (h) and doping level which is reflected on gap center (i). (j) 461 462 Histograms of  $E_{g,ex}$  extracted from the defect region and region excluding the defect.



Figure 4 | Magnetic field-induced modulation of the exchange gap. (a) Topography scan (-0.5 V, 100 pA) of a  $30 \times 30$  nm area where magnetic field dependent STS measurements were conducted. (b) Representative dI/dV spectra taken at different locations, blue: gapless regions, green: reduced-gap regions, red: large-gap regions and orange: regions where Dirac electron band is suppressed which prevents us from extracting the band gap. (c) Band gap map  $(80 \times 80 \text{ points}, -150 \text{ mV}, 400 \text{ pA})$  of the region in (a) at magnetic field B = 0 T. (d) dI/dV spectrum taken at the position marked by purple circle in (a) at B = 0 T (blue) and B = 1 T (red). A magnetic Dirac gap of 40.5 meV is opened with 1 T field in a gapless region at 0 T. (e) Band gap map (80×80 points, -150 mV, 400 pA) of the same region in (a) at B = 1 T. Black regions in (c) and (e) correspond to the suppressed Dirac electron band regions which prevent accurate determination of the exchange gap and are excluded from the maps and subsequent histograms in (f). (f) Histograms showing gap size at B = 0 T (upper panel) and B = 1T (lower panel). A clear renormalization of bandgap is observed with magnetic field. (g) Illustration of the band gap spatial fluctuation caused by surface magnetic disorder which can be reduced significantly by applying a perpendicular magnetic field. The blue, green and red Dirac cones represent gapless, partially gapped and fully gapped regions. Their representative dI/dV curves can be found in Figure 1(f). Upon applying a perpendicular magnetic field, the exchange gap in the Dirac cones increases until it reaches saturation. 

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682	Supporting Information				
683	Imaging the breakdown and restoration of topological protection in				
684 685	magnetic topological insulator MnBi <sub>2</sub> Te <sub>4</sub>				
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### 1. Structure and surface characterization of 5 SL MnBi<sub>2</sub>Te<sub>4</sub> epitaxial film



Figure S1 | Structure and surface characterization of ultra-thin MnBi<sub>2</sub>Te<sub>4</sub> epitaxial film. (a) Reflection high energy electron diffraction of the thin film showing strong sharp streaks which indicates high crystallinity. (b) STM topography scan on a 500×500 nm area (-2 V, 20 pA). (c) Histogram of number of SLs from (b) which shows majority of 4 SL terrace (green) with regions of 5 SL terrace (yellow). The thickness is determined from the depth of pinholes (dark blue and purple regions) which represent the Si substrate. (d) A line profile extracted from one of the 4 SL-5 SL step edges as marked by the black line in (b) and fitting to an edge function, which yields a step edge of 1.39 nm. (e) A line profile extracted across a pin hole to show the thickness of terraces as marked by the black line in (**b**).



Figure S2 | ARPES spectra and representative scanning tunnelling spectroscopy (STS) spectra of 5SL MnBi<sub>2</sub>Te<sub>4</sub> grown on (a) p-type and (b) n-type silicon substrates respectively using He-II light source (hv = 21.2eV). The Dirac cone is not visible in the ARPES spectra because of the low photon flux of the helium lamp. The energy distribution curve is taken at the wave vector marked by the red dashed lines and are plotted on the right of each spectrum. The edge of valence bands is determined from ARPES and the Dirac point energy is determined from the minima in the STS spectra, which are used for estimating the doping level of the samples. A doping level difference between the two types of Si (111) substrates is ~180 meV. 



Figure S3 | Schematics and determination of Bi<sub>Mn</sub> defects. (a) Top view of the crystal structure with primitive cell (green), Mn<sub>Bi</sub> defect (blue) and Bi<sub>Mn</sub> defect (red) marked. (b) Side view of the Bi<sub>Mn</sub> defect in a SL. The Te atoms on the very top appear as bright triangles at positive bias because of DOS propagated along Te  $p_z$  orbitals centered around Bi<sub>Mn</sub> defect. The three Te atoms appear to be more positively charged due to the extra charge from a  $Bi^{3+}$  ion replacing  $Mn^{2+}$  ion. (c) Topography scan on a 40×40 nm area (+1.7 V, 80 pA) showing Bi<sub>Mn</sub> defects on an atomically flat terrace. (d) A zoom-in image of the selected region marked by red box in (c) for extracting dimensions of defects. The length of the triangle's edge is around 1.35 nm which is close to the theoretical value of 1.31 nm. The same orientation of Mn<sub>Bi</sub> dark triangles and Bi<sub>Mn</sub> bright triangles also matches the schematics (a).

### 4. Determination of Dirac band gap and gap center from dI/dV spectrum



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**Figure S4** | Determination of Dirac band gap and gap center from dI/dV spectrum. Left, a dI/dVspectrum taken on a 5 SL MnBi<sub>2</sub>Te<sub>4</sub> terrace showing a gap in the Dirac states. **Right**, a schematic of the Dirac cone corresponding to the dI/dV spectrum on the left. The Dirac cone in 5 SL MnBi<sub>2</sub>Te<sub>4</sub> is lifted out of VB and resides between bulk conduction band (CB) and bulk valence band (VB). The Dirac gap is extracted based on the noise floor of the dI/dV spectrum (black dashed line). The width of the region between valence and conduction band edges corresponds to the size of Dirac gap  $\Delta$  as illustrated on the right figure.

To extract the Dirac gap from STS spectra, the edges of valence band and conduction band are determined from the spectrum as the onset of dI/dV intensity above the noise floor (black dashed line in **Figure S4** left). A logarithmic scale is chosen for dI/dV intensity axis to better account for the sudden change of intensity near the band edge. The local doping level can be estimated from the center of Dirac gap which can be calculated using

$$E_c = \frac{\int f(\mathbf{r}, E) E dx}{\int f(\mathbf{r}, E) dE}$$

where f(r, E)=1 if STS curve g(r, E) < noise floor and f(r, E)=0 otherwise<sup>19</sup>. This is essentially averaging the position of center of gap and is equivalent to extrapolating the gapped Dirac bands as shown in **Figure S4** right. Since the Dirac cone is now gapped by magnetic order and Dirac point no longer exists, the gap center (purple dot) is a good measure of the local doping shift by defects.

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#### 5. Bias dependent and set point current dependent STS measurements



BIAS (V) Figure S5 | Bias dependent and set point current dependent STS measurements. (a) STS spectra taken at different bias, the tip-sample tunneling junction decreases as the bias voltage decreases bias, resulting in a stronger signal which is necessary to observe the Dirac bands in dI/dV. (b) STS spectra taken at various set point current and fixed parking bias of -200 mV showing a Dirac gap. The band edge position shows minimal shift at different set point current and bias, indicating minimal tip induced band bending.

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853 As shown in **Figure S5(a)**, the STS spectrum on the terrace only shows a large bulk gap at parking bias of -1 V. As the tip is parked closer to the surface by decreasing bias, more features in the spectrum, 854 including surface states and Dirac band gap, can be resolved within the bulk gap. Therefore, we choose 855 856 parking bias of typical value -0.2 V and 300 pA for our STS maps. In Figure S5(b), STS spectra taken 857 on the same location and fixed bias but with varying set point current are plotted and offset manually. 858 From set point current of 300 pA to 1.5 nA, there is no significant shift of band edges or increase of band gap. In the case of tip induced band bending, increasing the set point current will increase the 859 860 band bending and lead to increase of band gap. Apparently, the most noticeable change in the Figure S5(b) is reduced noise in the spectra while the overall shape and position of the band edge stay the 861 862 unchanged. Therefore, we can rule out contribution from tip induced band bending to the band gap.

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6. Topography image and dI/dV map of the 5SL to 4SL edge in Figure 2 at other bias values.



 $x^{(nm)}$   $x^{(nm)}$   $x^{(nm)}$ 872 Figure S6| (a) Topography image (-1 V, 30 pA) and (b)-(i) d*I*/d*V* maps (-0.15 V, 400 pA) of the 873 edge in Figure 2 at other bias values.



Figure S7 | dI/dV maps on an even-to-odd edge at several bias across the band gap. (a) topography scans (-2.5 V, 20 pA) of the edge, atomic-resolution scan (-2.5V, 20 pA) where maps are taken and (b) STS spectra (-0.15V, 400 pA) at locations marked. The terrace edge is marked by the red dash line. (c) to (h) dI/dV maps (-0.15V, 400 pA) at various bias with location of edge marked.

- 8. Visualizing the gapless edge state and its coupling to bulk metallic states.



Figure S8 | Visualizing the gapless edge state and its coupling to bulk metallic states. (a) Topography image (-1 V, 50 pA) taken across a step edge from 4 SL to 5 SL MnBi<sub>2</sub>Te<sub>4</sub>. (b) d*I*/dV map at +25 mV bias (40 pA) in the same region as (a) to show the spatial distribution of the edge state. (c),(d) dI/dV spectra (-0.15V, 350pA) and height profile taken across the edge from top of the area and bottom of the area respectively marked by red dashed lines. The horizontal axis of the spectra is aligned with the height profile. The edge state is marked by red arrow and other peaks are attributed to disordered bulk states which is marked by white arrows. The white horizontal dashed line shows the bias at which (b) is taken. 





**Figure S9** | **Representative** dI/dV **spectra from STS map around**  $Mn_{Bi}$  **defects. (a)** Band gap map extracted from dI/dV spectra (-0.1 V, 800 pA) with some locations where STS shown marked in green. (b) Gap center map extracted from dI/dV spectra. (c) Stack plot of dI/dV spectra taken from locations in (a)-(b), where band gap values are marked on each dI/dV spectrum, and gap center positions calculated using method discussed in Figure S4 are marked by blue points.

965 10.Topography scan of the area in Figure 4 to show the alignment of the scan region.



Figure S10 | Topography scans (both taken at -0.5 V, 100 pA) of the region for band gap mapping
in Figure 4 at 0T and 1T.

11.Figure 4 statistical analysis of regions with unsuppressed CB at both 0T and 1T



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Figure S11 | Statistical analysis of regions with unsuppressed CB at both 0T and 1T. (a)-(b),  $E_{g,ex}$ map. (c)-(d), their corresponding histogram, and (e)-(f), statistical information extracted from the histograms.

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In Figure S11, the histograms are taken from regions where the conduction band intensity is not suppressed at 0T or 1T. The analysis shows similar results as in Figure 4, with an increase in the average band gap size of 20.6meV. Interestingly the histogram in Fig. S11(d) appears to be more bimodal where the lower gap size mode corresponds to the weight shift from gapless to medium size gap and upper mode corresponds to shift from regions with medium size gap to fully gapped ones in a 1T magnetic field. Overall, regardless of the analysis method, this region shows significant increase of Dirac gap and reduction of skewness relative to a normal distribution in a 1T magnetic field.

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### 12.Magnetic field dependent Dirac gap map



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Figure S12 | Magnetic field dependent Dirac gap map and their corresponding histogram and statistical information. (a) Topography scan of the map area ( $40 \times 40$  nm, -1 V, 100 pA). (b) – (f) Magnetic field dependent band gap maps (-0.15 V, 400 pA, 75×75 pixels), their histograms and statistic information.

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- 1028 The evolution of the histogram is very small below 0.6T and there is a sudden increase of count of 1029 gapless state. We assign this anomaly to the realigning process of  $Mn^{2+}$  moments that are initially 1030 oriented opposite to magnetic field due to magnetic disorder. The reduction of the effective out-of-1031 plane magnetization in some regions results in the temporary increase of gapless state. At 0.8T, the 1032 count of gapless states is greatly reduced, and the bimodal feature emerges in the histogram, which is 1033 similar to the bimodal behavior in **Fig.S11(d)**.
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Figure S13 | Dirac band gap maps with/without magnetic field showing band gap fluctuation 1041 1042 from region with Bi<sub>Mn</sub> defects. (a) Topography scan of the same area as in Figure S3 (80 pA, -1 V). (b) Spatial dependent of Dirac band gap modulation at B=0 T (-0.17V, 400 pA). (c) Dirac band gap 1043 map taken at 1 T field (-0.17 V, 400 pA) where gapped regions now form pattern (blue). (d) dI/dV1044 1045 spectra taken from the green circle in (a) at B=0 T (blue) and 1T (red). An exchange gap is opened in 1 T field where exchange coupling is enhanced by the external field. (e) Band gap histogram extracted 1046 from (b) which shows significant counts from gapless regions (dark red in b). (f) Histogram extracted 1047 from (c) which shows that Dirac band gaps are increased with a drastic decrease of gapless regions. 1048 1049

1050 Figure S13 shows magnetic field dependent Dirac band gap maps from a region with a large concentration of Bi<sub>Mn</sub> defects. Clearly, upon applying a perpendicular magnetic field of 1 T, the band 1051 gap map shows up more regions with small band gap around 10 meV. In some regions the band gap 1052 1053 has increased from gapless to moderate value of 40 meV. As shown in (f), there is now visible counts from regions with gap larger than 30 meV. Statistical analysis on the maps in (b) and (c) shows an 1054 1055 increase of average band gap value from 4.6 meV to 14.8 meV. The emerging counts from moderate 1056 band gap region results in an increase of standard deviation from 12.5 meV to 18.3 meV and similar 1057 to the results in **Figure 4**, the restoration of Dirac band gap is accompanied by decreasing skewness 1058 from 4.5 to 1.5. The results in Figure S13 indicates that regions with large numbers of Bi<sub>Mn</sub> defects are typically much more gapless and the exchange gap can be restored partially with a perpendicular 1059 magnetic field of 1 T. Because Bi<sub>Mn</sub> defects are non-magnetic, the pattern emerged in Figure S13c 1060 reflects the change of magnetic disorder in the center Mn<sup>2+</sup> layer. The amount of Bi<sub>Mn</sub> defects (see 1061 Figure S3c) could be responsible for the magnetic disorder. 1062

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