Supplementary Information

Spectroscopic evidence of intra-unit-cell charge redistribution in a charge-neutral magnetic topological insulator

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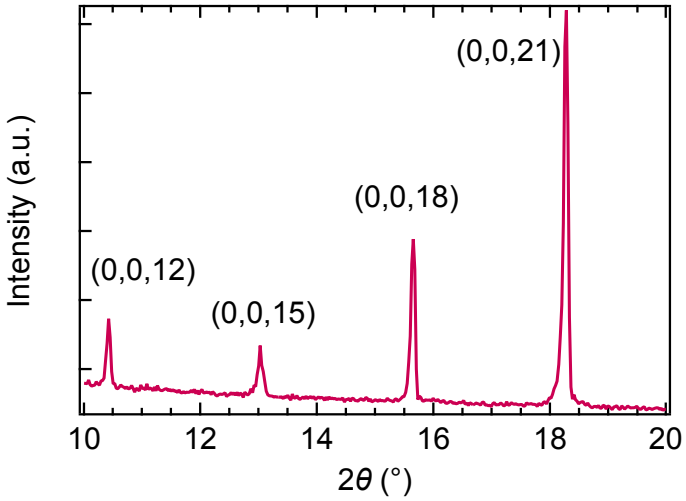

Fig. S1. X-ray diffraction pattern of $Mn(Bi_{1-x}Sb_x)_6Te_{10}$ crystal, with $x = 0.18$. The four peaks are at almost exactly the same locations as in undoped $MnBi_6Te_{10}$, indicating the same crystal structure. The wavelength of the X-ray source is 1.54059Å.

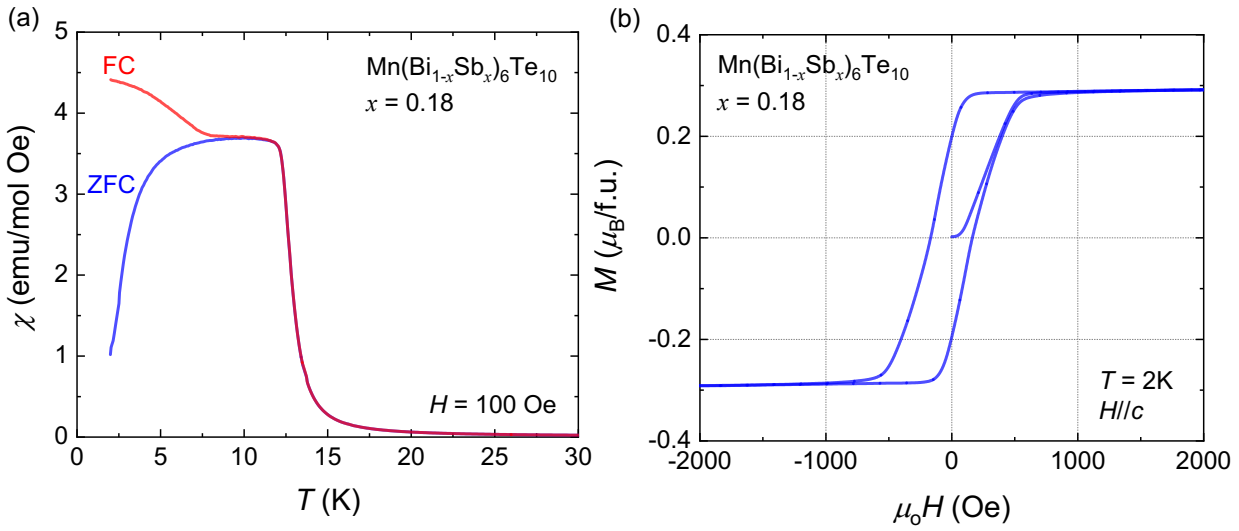

Fig. S2. Ferromagnetic ground state of $Mn(Bi_{1-x}Sb_x)_6Te_{10}$ crystal, with x = 0.18. (a) Temperature dependent zero-field-cooled (ZFC) and field-cooled (FC) magnetic susceptibilities indicate $T_C \sim 13$ K. (b) Isothermal magnetization curves with the magnetic field applied along the *c*-axis taken at 2 K.

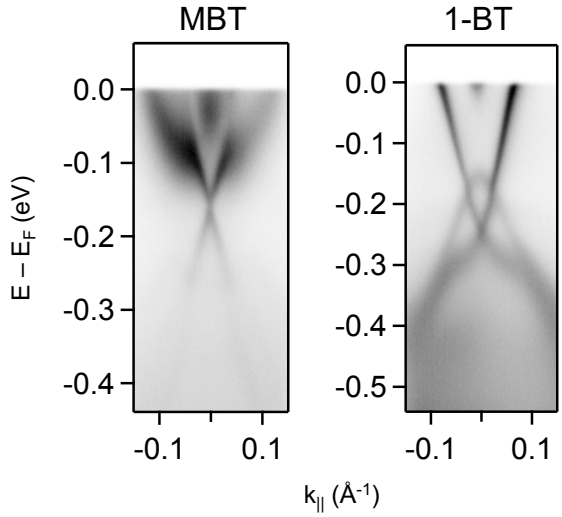

Fig. S3. Band structure of the MBT and 1-BT termination with wider ranges of binding energy. Both terminations show that the spectral features become diffuse at more than 0.15 eV below the Dirac point. This characteristic leads to the consequence that if the 2-BT termination is heavily hole-doped, the corresponding spectrum will show diffusive features, which cannot be distinguished from spectra originating from poorly cleaved areas.

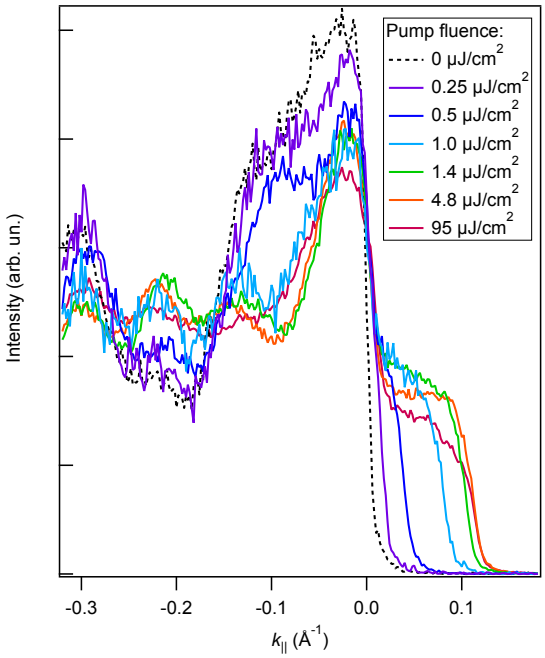

Fig. S4. Fluence-dependent surface photovoltage in the MBT termination. The figure shows energy distribution curves (EDCs) taken around the Γ point at -1 ps for different pump fluences.

A series of time-resolved ARPES (tr-ARPES) experiments were conducted on the MBT termination, as shown in Fig. 2 of the main text. The crystal was excited using 1.5 eV light with incident fluences varying from 0.25 to 95 μJ/cm². As illustrated in Fig. S4, the SPV effect, indicated by an energy shift, saturates at approximately 100 meV with a pump fluence of 4.8 μJ/cm². Notably, even when the fluence was significantly increased to 95 μJ/cm², no further shift was observed. This saturation behavior can only be attributed to the SPV effect in the "flat-band condition" rather than any conventional charging effect induced by photoexcitation.

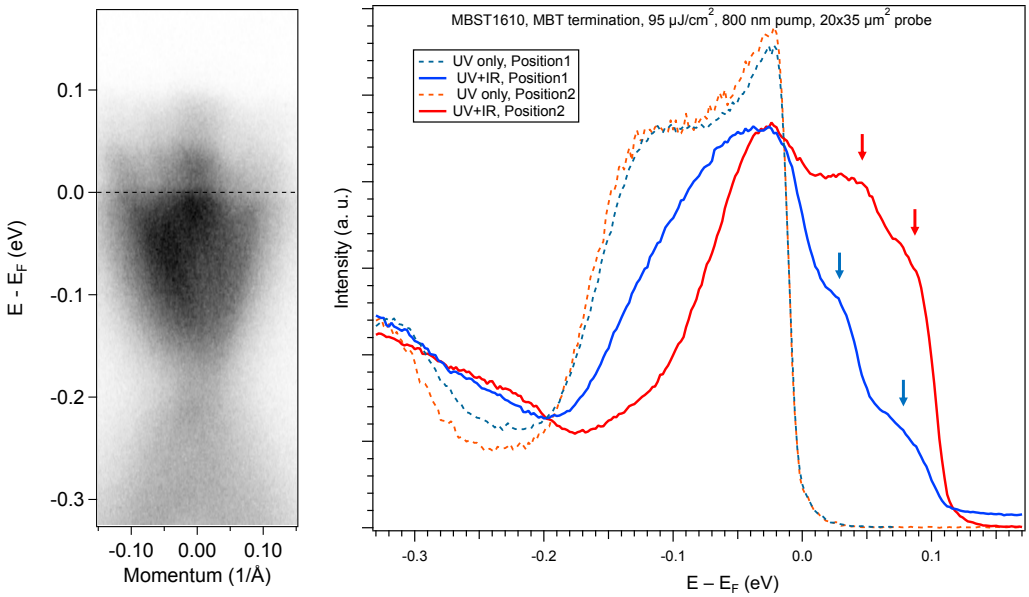

Fig. S5: Triple SPV effect on a region of the MBT termination. Left) The trARPES spectrum taken at –1 ps shows three replicas of the band structure. Right) EDCs taken around the Γ point at –1 ps for two positions where the triple SPV effect was observed. The experiment was done with an incident pump fluence of 95 µJ/cm^2.

On a different cleaved surface with an MBT-like termination, a triple SPV effect is observed, as evidenced by the splitting of the band structure into three nearly equidistant copies in energy, as shown in Fig. S5 (left). Energy distribution curves (EDCs) taken around the Γ point provide further quantitative insight into this SPV effect, as depicted in Fig. S5 (right). The energy spacing between closest band replicas is ~50 meV. This observation is consistent with the assumption that one-unit-cell offset in the *c*-direction between adjacent crystal domains induces a fixed potential difference across these surfaces.