

Supplementary Information for

Antiferromagnet-topological insulator heterostructure for polarization-controllable terahertz generation

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Supplementary Note 1: Characteristics of MnSe/(Bi, Sb)₂Te₃ thin film

The in-situ reflective high energy diffraction (RHEED) patterns from MnSe(111)/(Bi, Sb)₂Te₃ film are shown in Fig. S1a. When the deposition process was switched from (Bi, Sb)₂Te₃ to MnSe, a rapid change in the RHEED pattern could be observed. As shown in Fig. S1a, the streak spacing of MnSe is about 10% wider than that of the (Bi, Sb)₂Te₃ layer, which is consistent with the lattice constants in the literature. Figure S1b displays a transmission electron microscopy cross-sectional image, in which the (Bi, Sb)₂Te₃ and MnSe layers are separated by a clear interface.

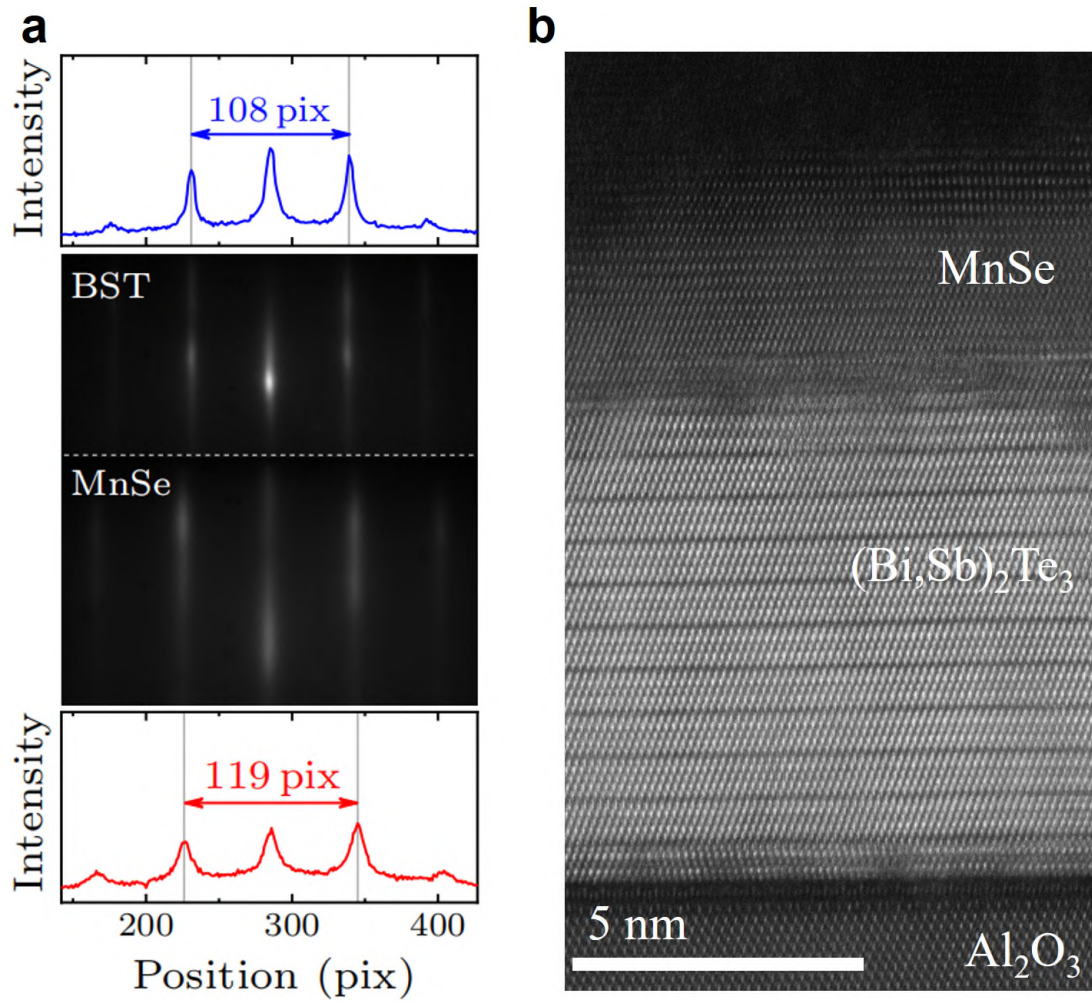


Fig. S1 | Characterization of MnSe/(Bi, Sb)₂Te₃ heterostructure. **a**, RHEED patterns of (Bi,Sb)₂Te₃ (BST) and MnSe. The curves represent the corresponding intensity. **b**, Cross-sectional dark field TEM image of MnSe/BST heterostructure.

Supplementary Note 2: THz emission spectroscopy and THz time-domain spectroscopy

The schematic experimental setup of the THz emission is shown in Fig. S2. A Ti:sapphire oscillator with central wavelength of 800 nm, pulse duration of 100 fs and repetition rate of 80 MHz is used as the laser source. The output from the oscillator is split into two beams: one is used for optical pumping (about 90 mW after the chopper), and the other (about 20 mW) is introduced into a THz detector for detecting the THz electric field. Two wire-grid THz polarizers are used to analyze the polarization state of the emitted THz pulses. Low-temperature grown GaAs photoconductive antenna is used as the THz detector. The whole setup except the laser system is enclosed in a transparent box purged with dry nitrogen gas to minimize water vapor absorption.

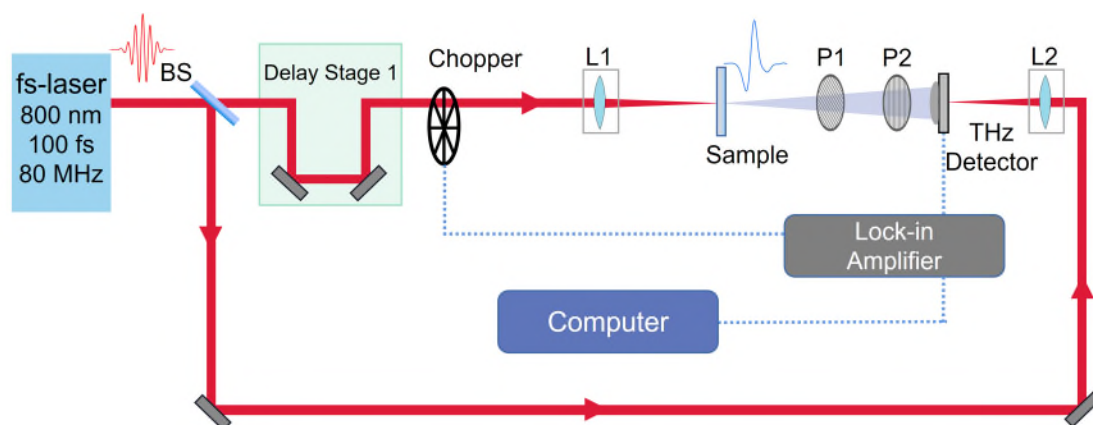


Fig. S2 | Schematic diagram of the THz emission spectroscopy. BS: beam splitter; L1 and L2: lens for focusing the 800 nm laser; P1 and P2: wire-grid polarizer; THz detector: low-temperature grown GaAs antenna integrated with silicon lens.

Supplementary Note 3: Two-mechanism THz radiation process

The flowchart of the overall THz emission process is shown in Fig. S3. The femtosecond laser pulse firstly generates a transient magnetism $\mathbf{M}(t)$ in the AFM layer. This abrupt change of magnetic moment leads to a MD radiation, as depicted by the red arrow branch. Meanwhile, the impulsive $\mathbf{M}(t)$ generates a spin current injecting from the AFM to TI layer (blue arrow branch). The spin current is converted into a transverse charge current at the TI surface through the inverse Rashba-Edelstein effect, which radiates THz waves via the electric dipole radiation. Consequently, the total THz radiation is determined by the two mechanisms.

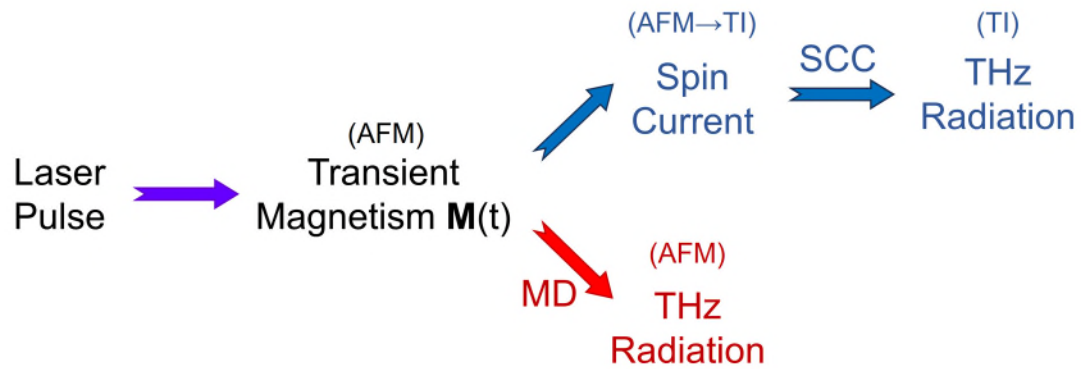


Fig. S3 | The schematic of the two-mechanisms THz radiation process.

Supplementary Note 4: Effect of the laser polarization on MnSe film

As a comparison to the MnSe/BST/substrate, we measure the dependence of the emitted THz electric field of the a MnSe/substrate sample on the pump polarization angle θ . The results are shown in Fig. S4a. The THz radiation waveform hardly changes with θ , and its THz E -field intensity only changes up to 8.5%. The polarity of the E -field never reverses. We explicitly present the THz E -field amplitude as a function of θ (Fig. S4b). The curve exhibits a two-fold pattern, as shown in Fig. S4b. These behaviors are significantly different from those in MnSe/BST/substrate samples, indicating that the AFM/TI interface affects the THz radiation via the SCC process.

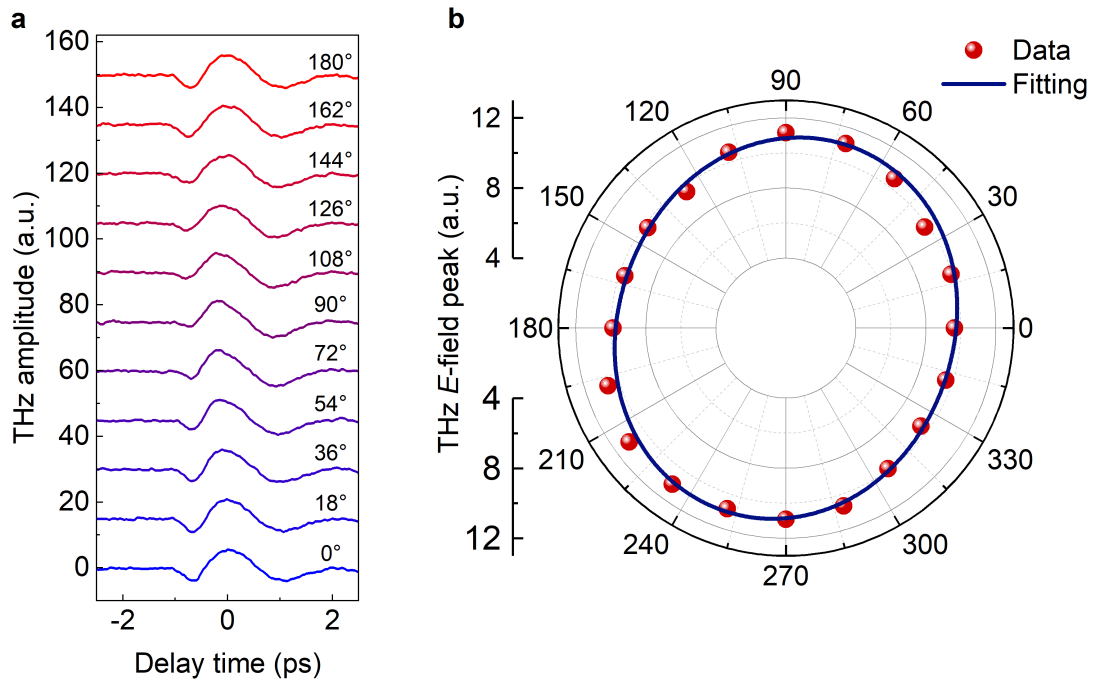


Fig. S4 | THz signals from a MnSe/substrate sample under various pump polarization directions. **a**, THz waveforms under different pump polarization θ . **b**, Amplitude of the THz emission as a function of θ . Solid curve: a cosine fit of the data.

Supplementary Note 5: Extracted MD and SCC emission components

According to the polarization-dependent THz waves in Fig. 2a, we extract the time trace of the MD and SCC channels and present them in Fig. S5. Given in line 209-218, the total THz signal is $S_{\text{TOTAL}} = A_{\text{SCC}}\cos(2\theta + \varphi) + A_{\text{MD}}$, which is illustrated in Fig. S5a. In Fig. S5b, the time trace at 30° (S_{max}) and 120° (S_{min}) are presented. Their sum ($S_{\text{max}} + S_{\text{min}}$) (red curve) and difference ($S_{\text{max}} - S_{\text{min}}$) (blue curve) are also plotted. In Fig. S5c, we shown the SCC and MD time traces. According to Fig. 3d, the MD channel THz radiation is only slightly θ -independent, and the SCC channel radiation oscillates as $S_{\text{THz}} = A_1\cos(2\theta + \varphi)$. Thus, the SCC channel corresponds to the $(S_{\text{max}} + S_{\text{min}})/2$ term (red curve in Fig. S5c) and the MD channel corresponds to the $(S_{\text{max}} - S_{\text{min}})/2$ term (blue curves in Fig. S5c). Figure S5d is the corresponding FFT spectra.

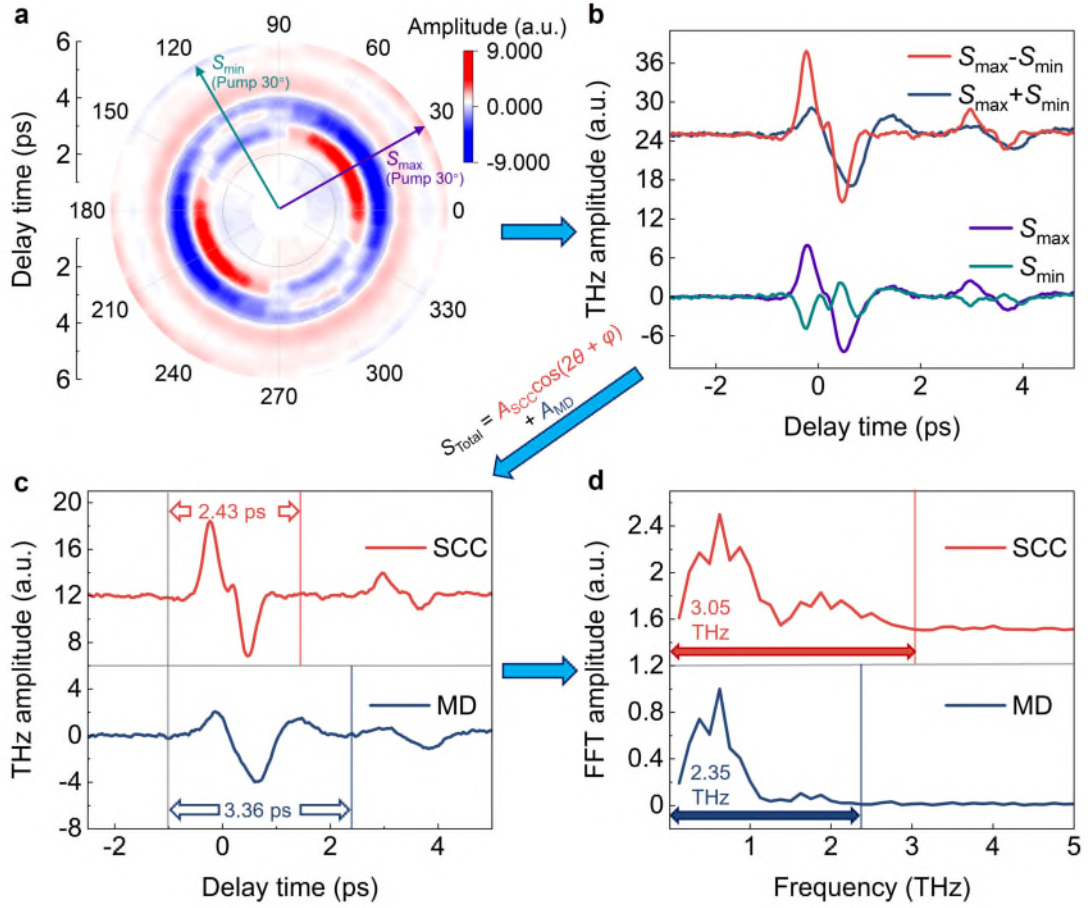


Fig. S5 | Extracted SCC and MD channel time- and frequency-domain data. **a**, Replotted Fig. 2a with the maximum (30°) and minimum (120°) amplitudes marked. **b**, The time traces of the sum and difference of the two components. **c**, Explicit extracted SCC and MD time traces. **d**, FFT of the time traces in **c** for the SCC and MD channels, respectively.

Supplementary Note 6: Theoretical outline of the laser-induced $\mathbf{M}(t)$

Under an ultrafast optical pumping, ac magnetization $\mathbf{M}^{(2)}(\Omega)$ can be parametrically generated through a stimulated Raman-type difference-frequency generation (DFG) process^{1,2}

$$M_i^{(2)}(\Omega) = \chi_{ijk}^{(2)}(\Omega; \Omega - \omega, \omega) E_j^*(\Omega - \omega) E_k(\omega), \quad (\text{S1})$$

where $\chi_{ijk}^{(2)}$ is a third-rank axial tensor in the coordinate frame of spins. The $\mathbf{M}^{(2)}(t)$ obeys the selection rule based on the conversion law of angular momentum. The changes of angular momentum from the electromagnetic field ($\mathbf{J}^{\text{EM}}$) itself, the electronic excitations in solids ($\mathbf{J}^{\text{ex}}$), and the crystalline structure ($\mathbf{J}^{\text{c}}$) are summed to be zero, which can be expressed as $\Delta\mathbf{J}^{\text{EM}} + \Delta\mathbf{J}^{\text{ex}} + \Delta\mathbf{J}^{\text{c}} = 0$. It has been derived that, owing to this angular momentum conservation law, only a material with three-fold rotational symmetry can sustain such a laser-induced $\mathbf{M}(t)$ (see Equation S1 and Fig. S6). For details, see Supplementary Reference 3. For explicit expression of $M_y(t)$ in a specific material preserving three-fold rotational symmetry, see Supplementary Note 7.

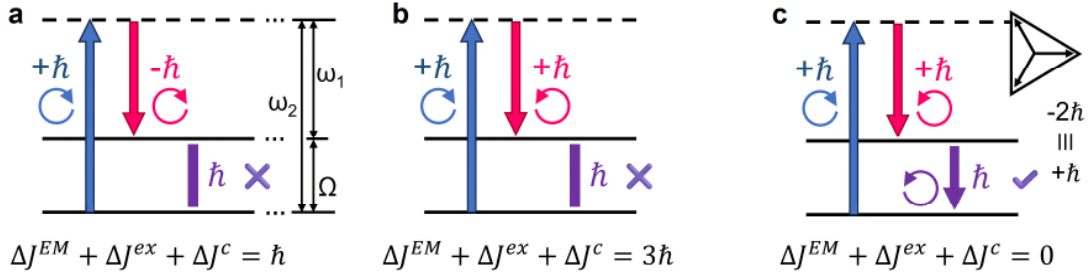


Fig. S6 | Selection rules for the DFG mechanism of laser-induced magnetization [adapted from Supplementary References 3]. **a**, **b**, Angular momentum changes for photons in the DFG processes with identical helicity (**a**) and opposite helicity (**b**). Blue and pink arrows: annihilation and creation of photons. Purple arrow: generation of $\mathbf{M}(t)$. **c**, Three-fold rotational symmetry crystalline structure allows the light-induced $\mathbf{M}(t)$ by providing an extra angular momentum of integer number times of $3\hbar$.

Supplementary Note 7: The three-fold periodic oscillation of radiation derived from $\mathbf{M}(\mathbf{t})$: in both MD and SCC mechanisms

In both the MD and SCC mechanisms, a prominent three-fold periodic oscillation characteristics has been observed (Fig. 3b). Given that $\mathbf{E}_{\text{THz}} \propto \partial^2 / \partial t^2 (\mathbf{e} \times \mathbf{M}(t))$ for the MD mechanism and $\mathbf{E}_{\text{THz}} \propto \mathbf{J}_c \propto \alpha_{\text{SH}} (\mathbf{J}_s \times \boldsymbol{\sigma}(t))$ and $\mathbf{J}_s \propto \mathbf{M}(t)$ for the SCC mechanism, the output THz electric field's three-fold periodic oscillation behavior must be due to a three-fold periodic oscillation in the $\mathbf{M}(\mathbf{t})$. We show why this is true as follows.

The details of the derivation has been reported in Refs. S3 and S4. There are twelve spin domains (named as $T_a S_b$, a and b denotes different spin domains with $a = 1, 2, 3, 4$ and $b = 1, 2, 3$) in the type-II AFM with rocksalt structure. In a single spin domain $T_l S_l$ for the [111]-oriented MnSe, the nonzero components of $\chi_{ijk}^{(2)}$ in (111) plane are only χ_{iii} and χ_{ijj} . When rotating the sample around [111]-axis by an angle of α , the transformation matrices for the spin coordinate frame (ijk) to the laboratory frame (xyz) can be written as

$$R = \begin{pmatrix} \cos\alpha & -\sin\alpha & 0 \\ \sin\alpha & \cos\alpha & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad (\text{S2})$$

and the transformation of second-order tensor is described by

$$\chi_{xyz}^{(2)T_1 S_1} = R_{xi} R_{yj} R_{zk} \chi_{ijk}. \quad (\text{S3})$$

Thus, the tensor components contributing to $M_y^{(2)}(\Omega)$ in the $T_l S_l$ spin domain can be obtained⁴ as

$$\chi_{yyy}^{(2)T_1 S_1} = \frac{1}{4} (\sin 3\alpha + \sin \alpha) \chi_{iii}^{(2)} + \frac{1}{4} (3\sin \alpha - \sin 3\alpha) \chi_{ijj}^{(2)}, \quad (\text{S4})$$

$$\text{and } \chi_{yxx}^{(2)T_1 S_1} = -\frac{1}{4} (\sin 3\alpha - 3\sin \alpha) \chi_{iii}^{(2)} + \frac{1}{4} (\sin 3\alpha + \sin \alpha) \chi_{ijj}^{(2)}. \quad (\text{S5})$$

For spin domains in MnSe other than the $T_l S_l$ domain, additional matrices are needed for transforming the coordinates of $T_2 S_l$, $T_3 S_l$, and $T_4 S_l$ spin domains into that of $T_l S_l$ spin domain, written as^{3,4}:

$$R^{T_1 \leftarrow T_2} = \begin{pmatrix} 2/3 & -\sqrt{3}/3 & \sqrt{2}/3 \\ \sqrt{3}/3 & 0 & -\sqrt{6}/3 \\ \sqrt{2}/3 & \sqrt{6}/3 & 1/3 \end{pmatrix},$$

$$R^{T_1 \leftarrow T_3} = R^{T_1 \leftarrow T_2} R^{2\pi/3},$$

$$\text{and } R^{T_1 \leftarrow T_4} = R^{T_1 \leftarrow T_2} R^{4\pi/3}.$$

Combining these contributions, the final tensor transformation is expressed as

$$\chi_{ymm}^{(2)} = \sum_{a,b} \eta(a,b) \chi_{ymm}^{T_a S_b} \quad (m = x, y), \quad (\text{S6})$$

where $\eta(a,b)$ is the ratio of contribution from different spin domains. Combining equations above, the transient $M_y(t)$ induced by the femtosecond laser pulses becomes

$$M_y(t) = \int \exp(i\Omega\tau) d\Omega \int \sum_j \chi_{ymm}^{(2)}(\Omega; \Omega - \omega, \omega) E_j^*(\Omega - \omega) E_j(\omega) d\omega. \quad (\text{S7})$$

Applying equation (S6) to equation (S7), $M_y^{(2)}(\tau)$ can be expressed in a simple form

$$M_y(t) = \left[A(\chi_{ymm}^{(2)\text{MEE}}) \cos(3\alpha + \phi) + B(\chi_{ymm}^{(2)\text{MEE}}) \cos(\alpha + \phi') \right] + C, \quad (\text{S8})$$

where coefficients A, B, and C are determined by $\chi_{ymm}^{(2)}$ and $\eta(a,b)$. Both the MD- and SCC-induced THz radiation have the same dependence on α . Equation S8 well explains the data in Fig. 3b.

Supplementary Note 8: Data fitting of the azimuth-dependent THz radiation from MnSe

Given the function of $E_{\text{THz}}=[A \cos(3\alpha+\phi)+B \cos(\alpha+\phi')]+C$ (Ref. 4), we present all fitting components in Fig. S7. The three-fold rotational symmetry is marked by the red curves in Fig. S7a). However, we find that the data fitting can be improved by including both a difference frequency $\cos 2\alpha$ and sum frequency $\cos 4\alpha$ component (Fig. S7c). The medium result with only extra $\cos 2\alpha$ component is also included as Fig. S7b. The data fitting is excellent in Fig. S7c, with the three-fold rotational symmetry clearly demonstrated (red oscillation curve). These $\cos 2\alpha$ and $\cos 4\alpha$ components might be due to some interacting terms between different spin domains in MnSe (Ref. S3 and S4).

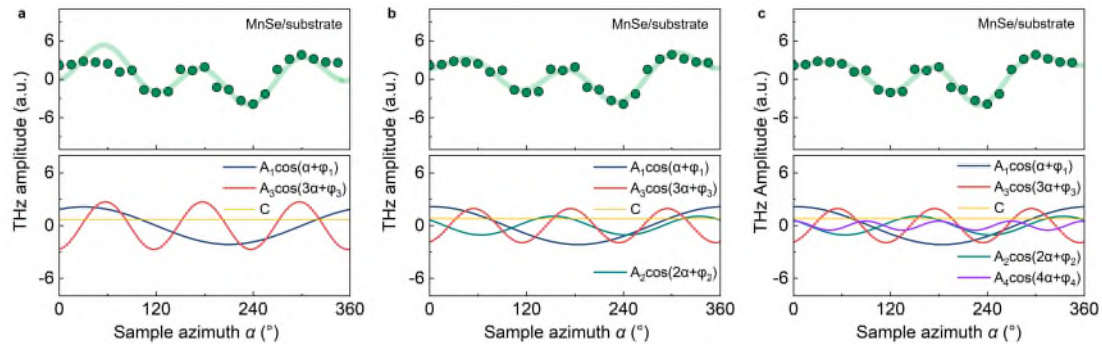


Fig. S7 | Dependence of MnSe/substrate THz radiation on the azimuth α . The data fitting is described by equations in Supplementary Note 7. **a-c**, Data fittings when 2, 3 and 4 cosine components are used in the fitting equations. Different fittings are marked by different colors. Green dots: experimental data.

Supplementary Note 9: Control knobs of THz radiation from $\text{MnSe}/(\text{Bi, Sb})_2\text{Te}_3$

The tunability of radiated THz pulses is crucial for real application. In the main text Fig. 4 and Fig. S8, the tunability of our device is explicitly demonstrated. By tuning the polarization of the pump beam, the polarization of the radiated THz beam can be easily controlled. The tuning knob is the polarization of the pump beam. The sample azimuth can also be an additional tuning knob, as a complement to the polarization control.

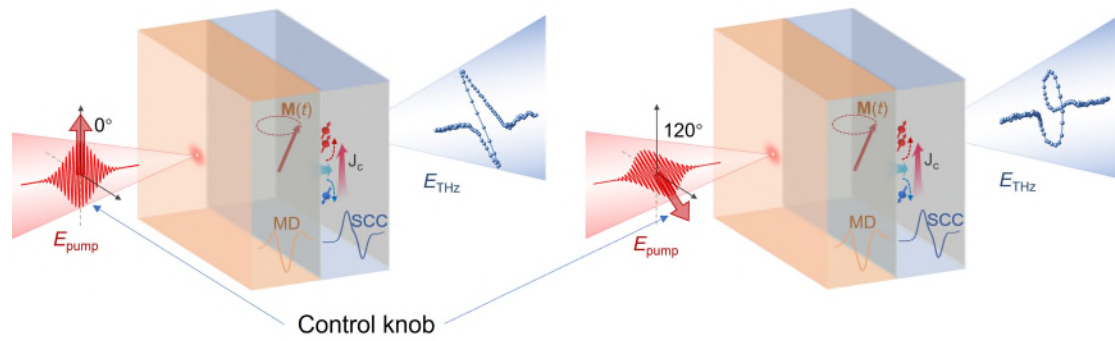


Fig. S8 | Schematic of the control knob. Controlling the polarization state of THz radiation using pump beam polarization as a convenient control knob (thick red arrow).

Supplementary Note 10: Electric field strength of the THz radiation

We calculate the THz field strength by comparing our THz radiation signal with that from ZnTe. First, we use a 0.5-mm thick ZnTe crystal to replace our sample, whose THz emission electric field is recorded by using another 2-mm thick ZnTe as the detection crystal. The detailed calculations are given in Ref. S5. At a pump beam power of 100 mW, the measured THz electric field for our 0.5-mm ZnTe is 503.5 V/m. Then, under the same condition (except for a pump beam power of 70 mW, to avoid sample damage), the THz radiation from our sample is also measured. By comparing the results with that from ZnTe, we measure the THz electric field from our MnSe/(Bi,Sb)₂Te₃ heterostructure to be 22.3 V/m (in Fig. S9).

To calculate the THz conversion efficiency, we estimate the single pulse energy of the 800 nm pump beam and THz radiation. The average power of the pump beam is 70 mW, and the single-pulse energy $\mathcal{E}_{800\text{ nm}}$ can be given by

$$\mathcal{E}_{800\text{ nm}} = \frac{P_{ave}}{f_{rep}} = \frac{70\text{ mW}}{80 \times 10^6\text{ Hz}} = 0.875 \times 10^{-6}\text{ mJ},$$

where f_{rep} is the repetition rate of the pump beam. The single pulse energy of THz radiation is calculated by the integration of the THz electric field in the time domain and area, expressed as

$$\mathcal{E}_{THz} = \epsilon_0 c \iint \vec{E}_{THz}^2 dA dt = \epsilon_0 c \cdot 2\pi \frac{(2.5\text{ mm})^2}{2} \times \int \vec{E}^2(0) dt ,$$

where $\vec{E}_{THz}(r) = \vec{E}(0) \cdot e^{-\frac{r^2}{(2.5\text{ mm})^2}}$, $\vec{E}(0) = 22.3\text{ V/m}$. The calculated single-pulse energy of THz radiation is $\mathcal{E}_{THz} = 1.78 \times 10^{-14}\text{ mJ}$. Thus, the THz conversion efficiency is $\eta = \mathcal{E}_{THz} / \mathcal{E}_{800\text{ nm}} = 2.03 \times 10^{-8}$, which is two orders of magnitude smaller than those from a typical THz nonlinear crystals (such as ZnTe and GaP). However, note that the thickness of our sample is only 12 nm. In an ideal case, when our sample was periodically stacked to a 1 mm equivalent thickness, the THz conversion efficiency can reach 0.17 %, which is significantly higher than most of the nonlinear THz crystals, such as ZnTe and GaP.

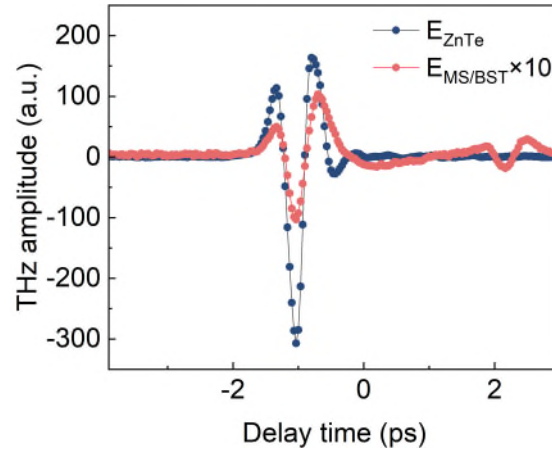


Fig. S9 | Comparison of THz radiation electric field strength from a 0.5-mm thick ZnTe sample and our MnSe/(Bi,Sb)₂Te₃ heterostructure.

Supplementary Note 11: Discussion on the nearly polarization-independent MD radiation

To avoid confusion, we first emphasize the difference between the effect of sample azimuth α and pump polarization θ on THz radiation. The pump polarization dependence and sample azimuth dependence are described by the same set of nonlinear coefficients. However, the difference comes from the fact that to detect the THz signal, the reference beam has a fixed polarization. Thus, rotating the pump polarization and sample azimuth are different. Usually, as shown in Fig. S10, the laser-induced magnetization has a direction that is uniquely determined by the sample crystal lattice. Although rotating the pump polarization and azimuth is equivalent from the point of view of coordinate transformation, the output is textured by the reference beam polarization at the detector. If the reference is fixed (which is apparently the usual case), then rotating the sample azimuth and the pump polarization will become different. Suppose the reference polarization and the detector is rotated together with the sample azimuth, then there will be no difference between rotating the pump polarization and sample azimuth.

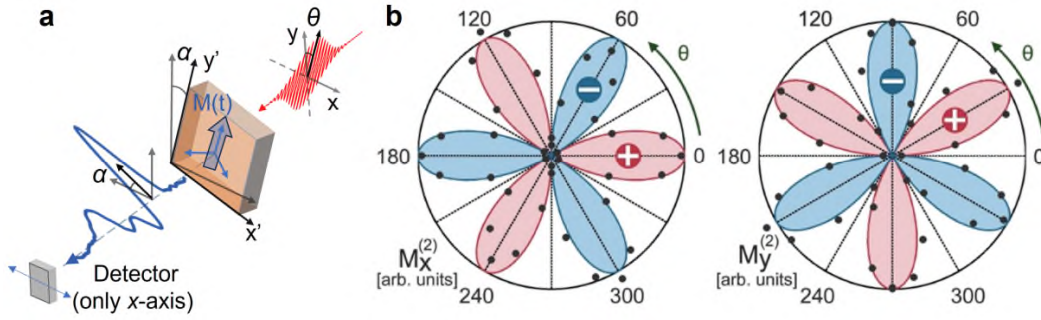


Fig. S10 | **a**, Schematic of the experimental setup. The direction of laser-induced magnetization (as well as the THz electric field) is rotated simultaneously with the sample azimuth. Rotating the pump polarization does not affect the direction of the THz electric field. Thick blue arrow: the direction of laser-induced magnetization. **b**, In plane components M_x and M_y of the laser-induced magnetization as functions of sample azimuth (adapted from Ref. S3).

Now we further discuss the dependence of MD radiation on pump polarization. From the theoretical point of view, based on the nonlinear susceptibility analysis in Refs. S3, S4), the transient magnetization induced by laser pulse is expressed as

$$M_y(\Omega) = \int \sum_j \chi_{ymm}^{(2)}(\Omega; \Omega - \omega, \omega) E_j^*(\Omega - \omega) E_j(\omega) d\omega.$$

When the direction of pump polarization is rotated by θ with respect to the y-axis, the electric field terms in the integral can be written as

$$E_j^*(\omega_1)E_j(\omega_2) = \begin{cases} E^2 \cos^2(\theta), & j = x \\ E^2 \sin^2(\theta), & j = y \end{cases}.$$

Hence, M_y can be expressed in a simple form as

$$M_y \propto \left[A(\chi_{ymm}^{(2)\text{MEE}}) \cos(3\alpha + \phi) + B(\chi_{ymm}^{(2)\text{MEE}}) \cos(\alpha + \phi') \right] [\cos(2\theta) + C],$$

where A, B and C are the coefficients determined by $\chi^{(2)\text{MEE}}$ and η (ratio of contribution from other spin domains). In main text line 200-201, we wrote “MD radiation in MnSe is nearly θ -independence”. The THz radiation from MnSe does have a $\cos 2\theta$ oscillation (Fig. 3d), with only 8.5% changes of the intensity. Hence, it is still consistent with the theoretical predictions of a $\cos(2\theta) + C$ form, except for a dominant much larger C term. The large C term’s origin is very possibly as: the MD is a function of time; the MD-induced THz radiation is actually not directly proportional to the laser-generated M, but its second order derivative relative to time, expressed as $E_{\text{MD}} \propto \partial^2 / \partial t^2 (\mathbf{e} \times \mathbf{M}(t))$, where $\mathbf{e}$ is the unit vector of THz radiation propagation direction. In contrast, the SCC-induced THz radiation is directly proportional to the induced magnetization M ($\mathbf{E}_{\text{SCC}} \propto \mathbf{J}_s \propto \mathbf{M}(t)$). This explains why the MD channel is not so sensitive to the pump polarization directly. The pump polarization direction θ can only affect the intensity of M, but not its changing rate.

Supplementary References

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