

iScience, Volume 25

Supplemental information

**Terahertz wave emission
from the trigonal layered PtBi₂**

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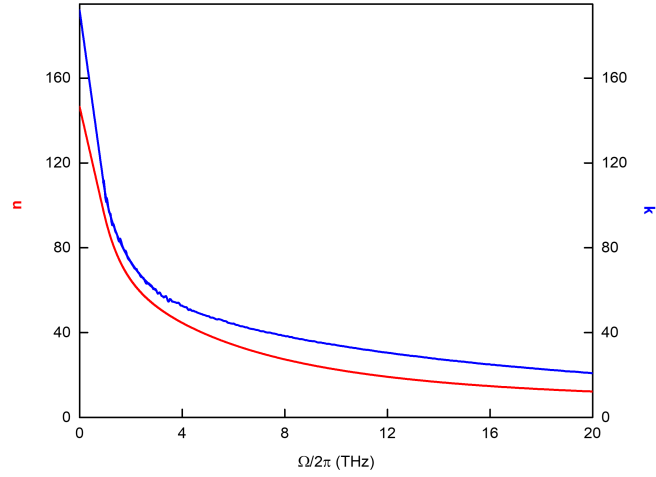


Figure S1: The complex refractive index. This figure is related to “Ultrafast photocurrent calculation and refractive index data” in the STAR methods section. The complex refractive index ($\tilde{N} = n + ik$) of PtBi₂ in THz regime, the red curve represents n , the blue curve represents k .

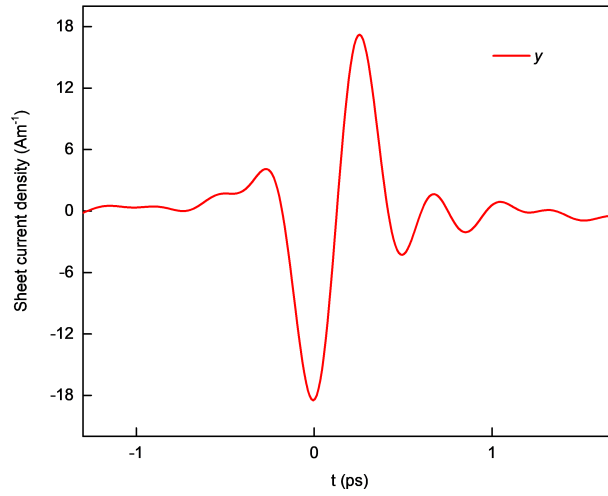


Figure S2: The ultrafast sheet current density. This figure is related to “Ultrafast photocurrent calculation and refractive index data” in the STAR methods section. The ultrafast sheet current density $J(t)$ from trigonal layered PtBi₂ with y -polarized pump light.

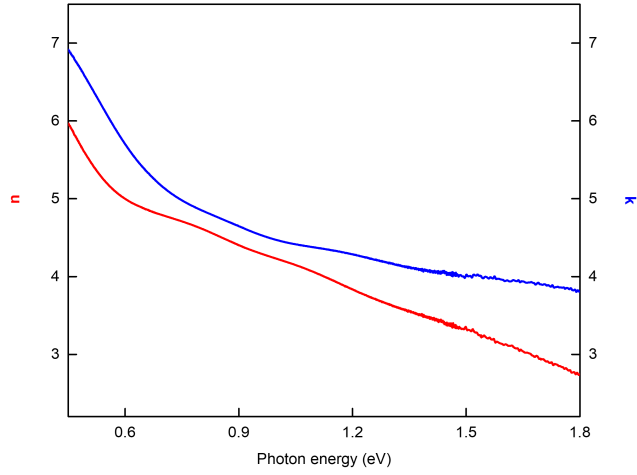


Figure S3: The complex refractive index. This figure is related to “Ultrafast photocurrent calculation and refractive index data” in the STAR methods section. The complex refractive index ($\tilde{N} = n + ik$) of PtBi₂ as the function of photon energy, the red curve represents n , the blue curve represents k .

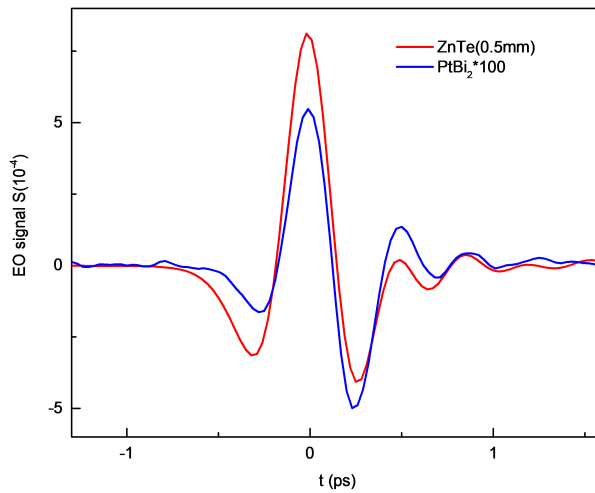


Figure S4: The comparison of THz wave emission between ZnTe and PtBi₂. This figure is related to the discussion of figure 4 in the main text. THz emission waveform from 0.5-mm-thick ZnTe and PtBi₂ under the same experimental conditions.

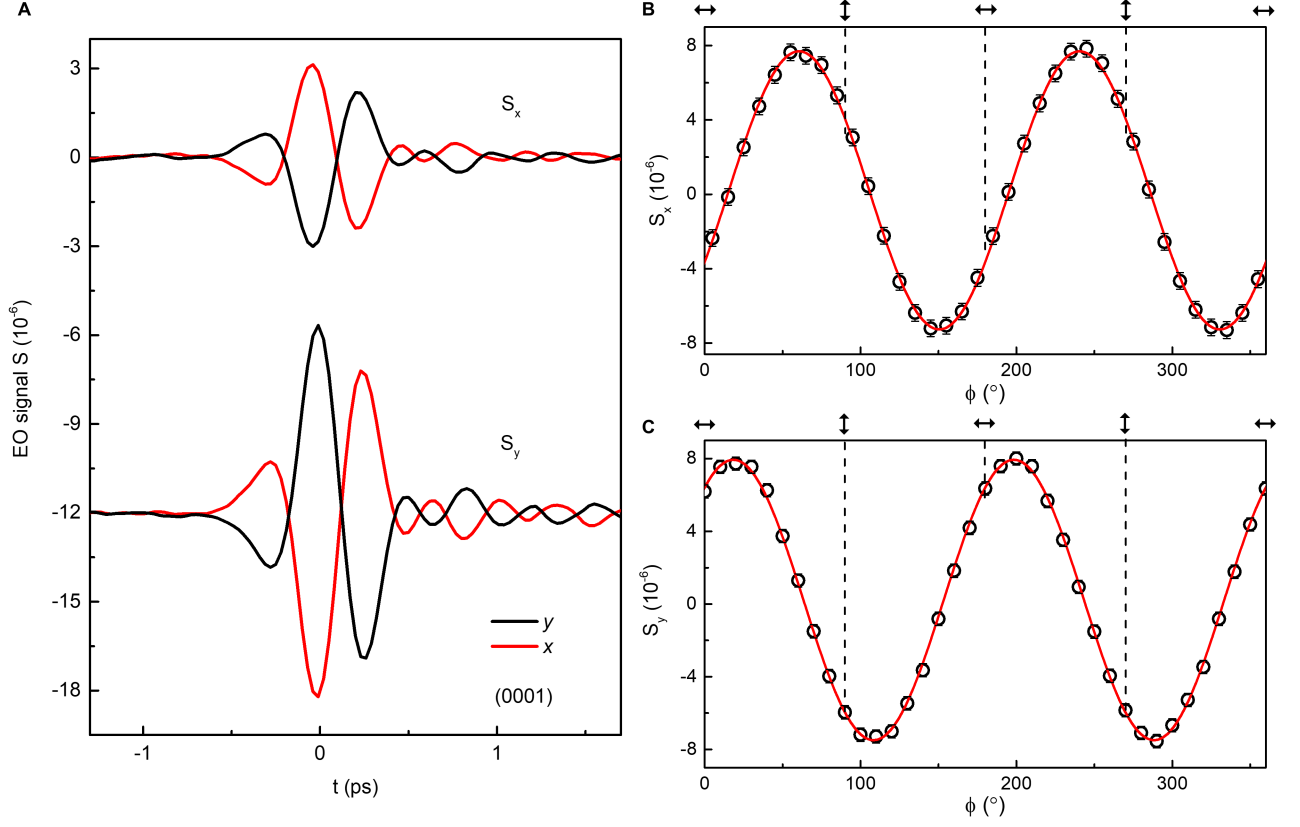


Figure S5: THz EO signal obtained by rotating the HWP angle with pump photon energy of 0.9 eV. This figure is related to figure 2 in the main text. (A) Typical THz EO-signal components $S_x(t)$ and $S_y(t)$ measured with x - and y polarization pump. (B) and (C) The peak value of S_x and S_y (at $t = 0$ ps) as functions of ϕ , respectively. The red solid curves represent for the fitting based on based on Equations (1) and (2) in the main text.

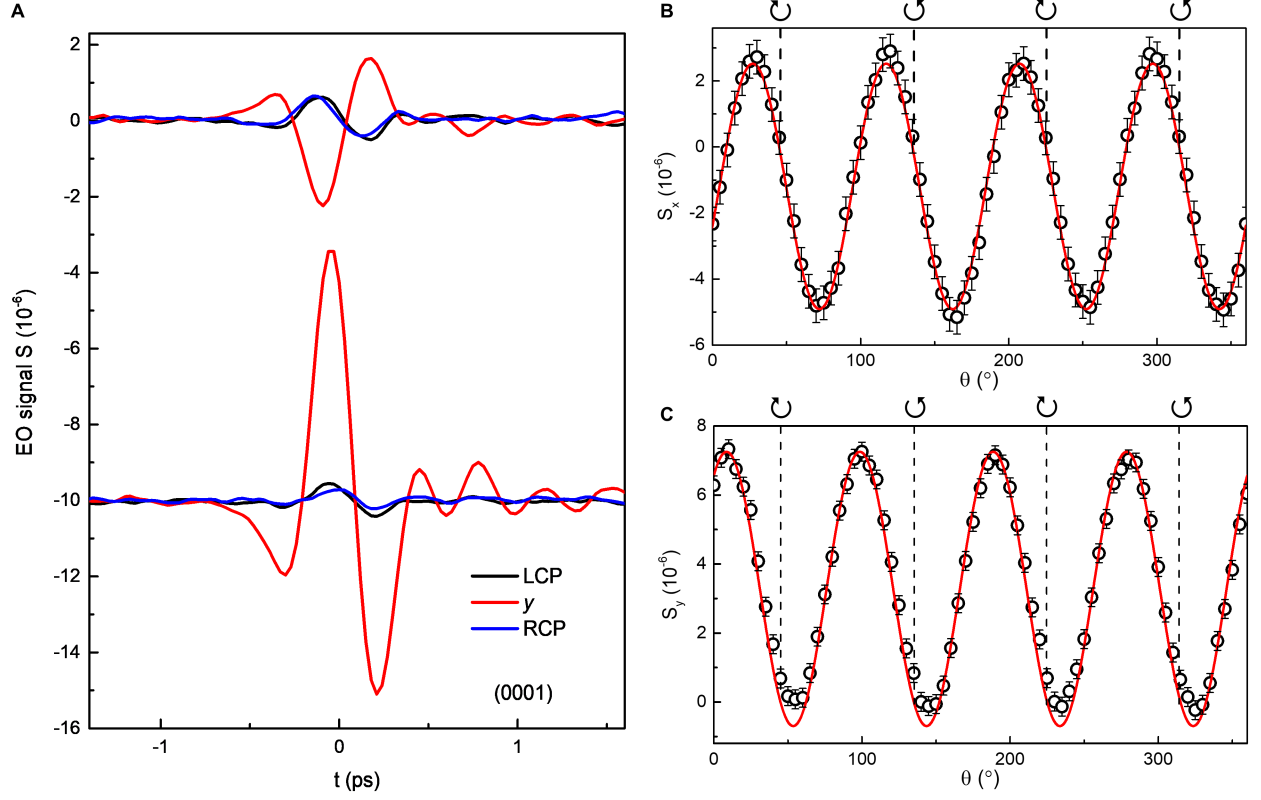


Figure S6: THz EO signal obtained by rotating the QWP angle with pump photon energy of 0.9 eV. This figure is related to figure 3 in the main text. (A) Typical EO signal $S_x(t)$ and $S_y(t)$ measured with different pump helicity (different QWP angle θ). Here, y ($\theta = 0^\circ$), LCP ($\theta = 45^\circ$), and RCP ($\theta = 135^\circ$) represent the y -polarized, left-hand, and right-hand circular polarized laser pump, respectively. (B) and (C) The peak value of S_x and S_y (at $t = 0$ ps) as functions of QWP angle θ . The red solid curves represent for the fitting based on Equations (3) and (4) in the main text.

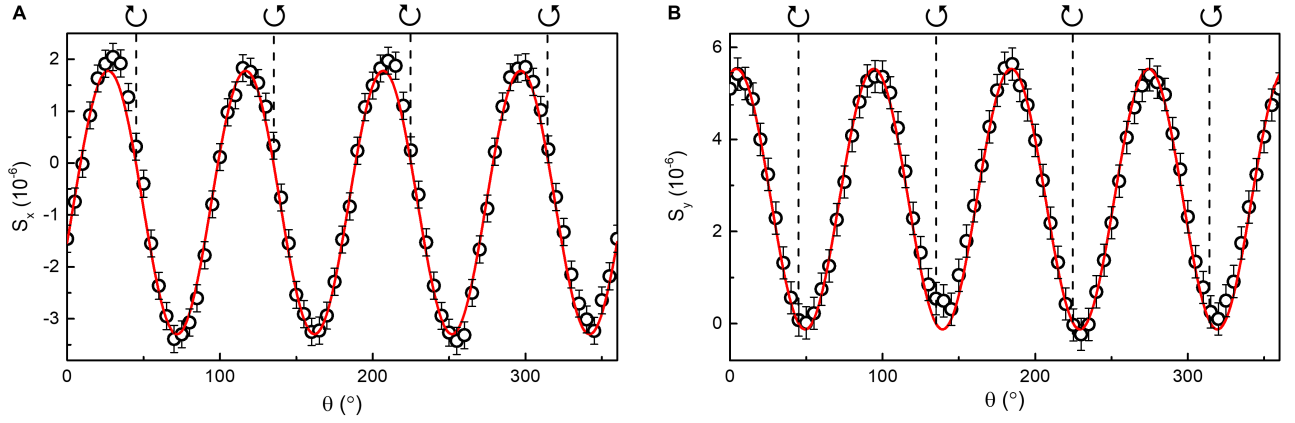


Figure S7: Helicity-dependent THz emission peak value with pump photon energy of 0.7 eV. This figure is related to figure 3 in the main text. (A) and (B) The peak value of $S_x(t)$ and $S_y(t)$ (at $t = 0$ ps) as functions of QWP angle θ . The red solid curves represent for the fitting based on Equations (3) and (4) in the main text.

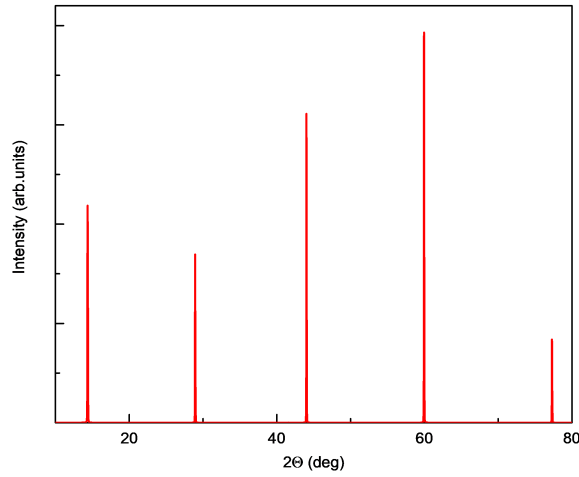


Figure S8: XRD data of PtBi₂. This figure is related to “Materials details” in the STAR methods section. X-ray diffraction pattern of our PtBi₂ sample.

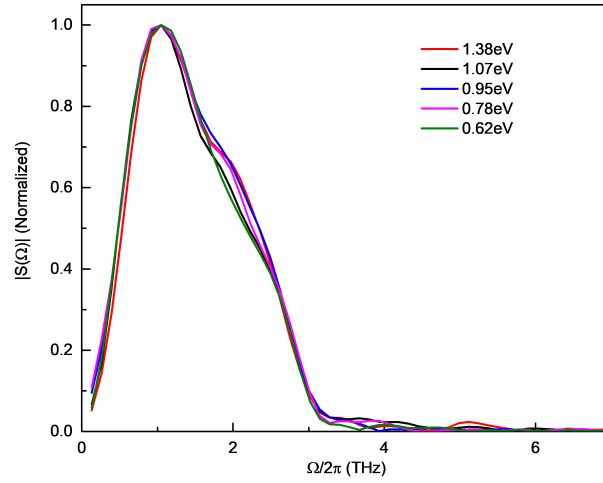


Figure S9: Fourier transform spectra at different pump photon energy. This figure is related to figure 4A in the main text. The Fourier transform spectra $|S(\Omega)|$ corresponding to the time-domain THz waveforms at different photon energy.