

Supplementary Information

**Generation and manipulation of light-induced orbital
transport in Co/Zr/Al₂O₃ heterostructure probed with
ultrafast terahertz emission**

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Supplementary Note 1. Structural characterization by XRD patterns.

Figure S1 shows the room temperature X-ray diffraction (XRD) pattern obtained by grazing incidence. The substrate Al_2O_3 exhibits prominent (006) characteristic peaks. The investigated Zr layer displays distinct (002) and (100) characteristic peaks, indicating a high degree of crystallinity.

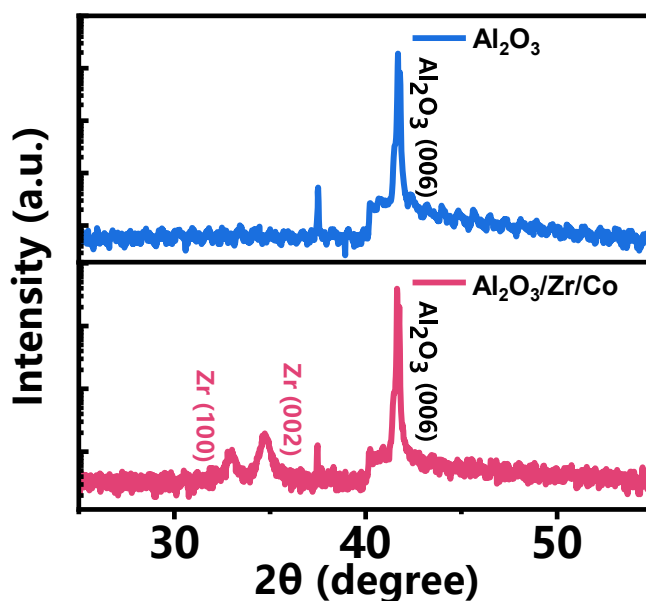


Figure S1 XRD pattern of a Al_2O_3 substrate (blue) and $\text{Co}/\text{Zr}/\text{Al}_2\text{O}_3$ heterostructure (red) with Zr thickness of 20.0 nm.

Supplementary Note 2. THz emission with both-side excitation for samples with different thicknesses.

Figure S2 (a) and (b) show the THz time-domain waveforms generated under front-side (red) and back-side (blue) excitation for Co/Zr/Al₂O₃ and Co/W (1)/Zr/Al₂O₃ heterostructures, respectively, with different d_{Zr} . Compared to back-side excitation, THz pulses generated by front-side excitation exhibit opposite polarities [1]. The time delays between THz pulses with front and back excitations are due to the group velocity mismatch of 800 nm and THz signals in Al₂O₃ single crystal substrates.

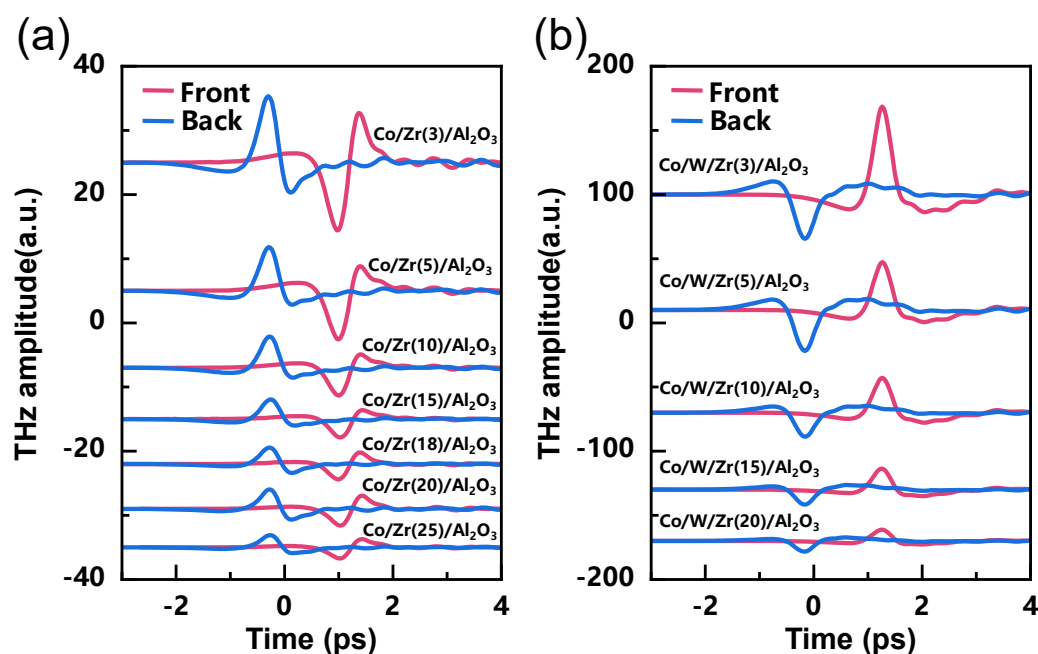


Figure S2 (a) THz signals generated by Co/Zr (3–25)/Al₂O₃ bilayer heterostructures under optical pumping from the front side (red) and the back side (blue). The measurements were performed under an external magnetic field strength of $H=100\text{mT}$, and the THz signals for different d_{Zr} values were vertically offset for clarity. **(b)** THz signals generated by Co/W(1)/Zr (3–20)/Al₂O₃ trilayer heterostructures under optical pumping from the front side (red) and the back side (blue). Similar to (a), the measurements were conducted under an external magnetic field strength of $H=100\text{mT}$, and the THz signals for different d_{Zr} values were vertically offset for clarity.

Supplementary Note 3. THz emission under film-side excitation with positive and negative magnetic fields.

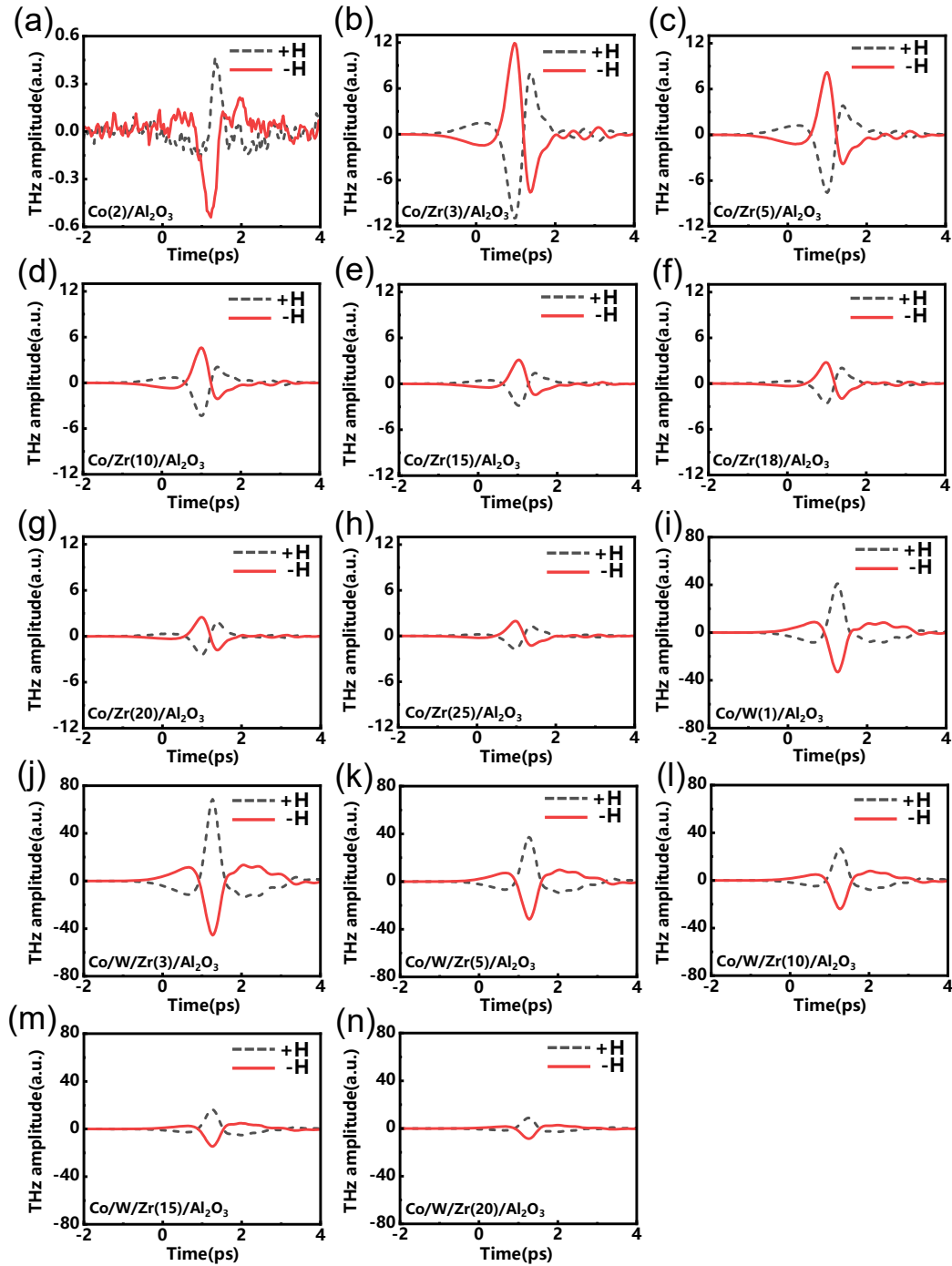


Figure S3 (a) and (i) THz time-domain waveforms generated by Co(2)/Al₂O₃ and Co/W(1)/Al₂O₃, respectively, under the magnetic field conditions of +H (black dashed line) and -H (red solid line). These waveforms serve as reference signals. (b)-(h) THz

emission waveforms of Co/Zr(3-25)/Al₂O₃ bilayer heterostructures under film-side excitation with +H/-H. (j)-(n) THz emission waveforms of Co/W (1)/Zr(3-20)/Al₂O₃ trilayer heterostructures under film-side excitation with +H/-H.

Figure S3 (a) shows that the single-layer Co generates a very weak THz signal, and its polarity reverses upon inverting the magnetic field. This observation is consistent with the THz radiation generated by ultrafast demagnetization, as analyzed in the text.

Figure S3 (b)-(h) and (j)-(n) show the THz emission signals under excitation from the front-side for Co/Zr/Al₂O₃ and Co/W (1)/Zr/Al₂O₃ samples with varying d_{Zr} , respectively. Upon reversal of the magnetic field, the polarity of the THz signal reverses, while the amplitude exhibits mostly symmetry [1]. Under the same magnetic field, the polarities of the THz signals from Co/Zr/Al₂O₃ and Co/W (1)/Zr/Al₂O₃ samples are opposite.

Figure S3 (i) shows the THz signal generated by the Co/W (1)/Al₂O₃ bilayer.

Supplementary Note 4. Influences of the polarization of the pump laser

We pump the Co/Zr(3)/Al₂O₃ with femtosecond laser for σ^+ , σ^- helicities, and linear polarization. **Figure S4** show time traces of THz emission for σ^+ , σ^- helicities and linear polarization pumping. We observe that the THz waveforms remain the same, regardless of the laser pulses' helicities.

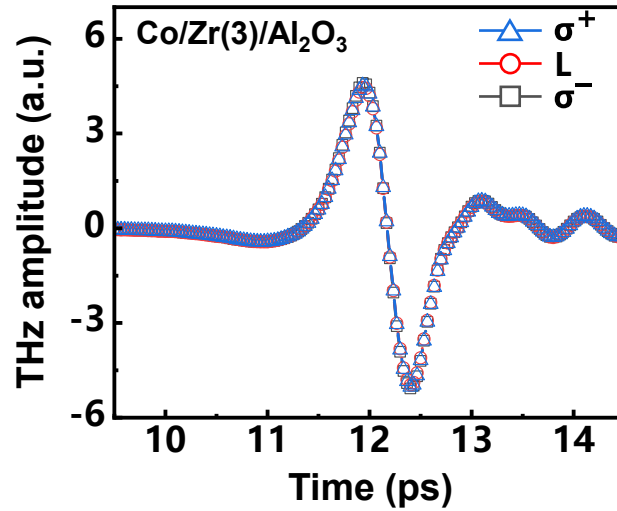


Figure S4 The THz emission induced by pump lasers with σ^+ , σ^- helicities, and linear polarization.

Supplementary Note 5. Fast Fourier transform of the THz waveforms from Co/Zr/Al₂O₃ and Co/W/Zr/Al₂O₃ heterostructures.

We performed Fourier transform on the THz time-domain waveforms of Co/Zr/Al₂O₃ and Co/W(1)/Zr/Al₂O₃ heterostructures with varying thicknesses of Zr, as show in **Figure S5** (red for Co/Zr/Al₂O₃ and blue for Co/W (1)/Zr/Al₂O₃). The peak of THz spectra of Co/Zr/Al₂O₃ heterostructures show a frequency shift when Zr thickness is increasased, corresponding to the the time-delay observed in the time-domain[2].

The dashed curves in (a) and (b) are the spectra for Co/Zr(25)/Al₂O₃ and Co/W/Zr(20)/Al₂O₃ with maximum normalzied to unity.

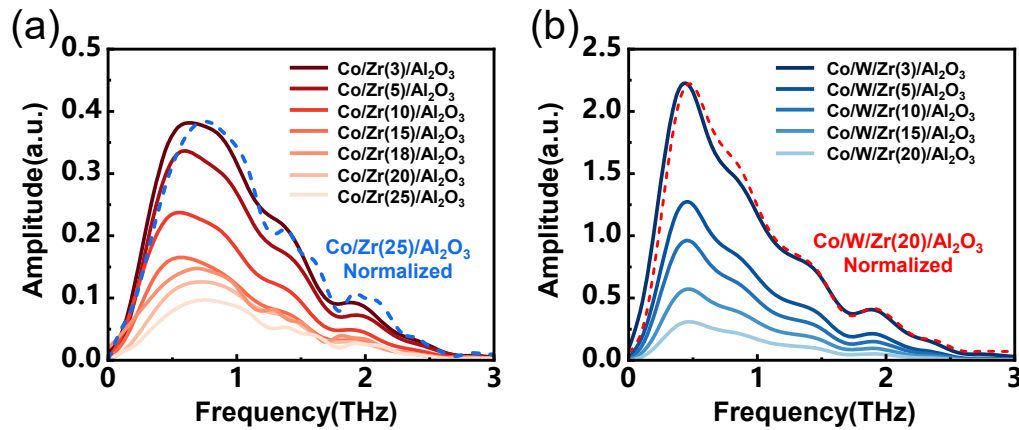


Figure S5 (a) The solid lines (red) represent the Fourier spectra of the emitted THz waveforms for Co/Zr/Al₂O₃ heterostructures with varying d_{Zr}. The dashed line (blue) corresponds to the normalized Fourier spectrum of the THz waveform for Co/Zr(25)/Al₂O₃. (b) The solid lines (blue) represent the Fourier spectra of the emitted THz waveforms for Co/W (1)/Zr/Al₂O₃ heterostructures with varying d_{Zr}, while the dashed line (red) corresponds to the normalized Fourier spectrum of the THz waveform for Co/W (1)/Zr(20)/Al₂O₃.

Supplementary Note 6. THz-TDS transmission measurement.

Figure S6 (a) and (c) show the THz-TDS transmitted through Co/Zr and Co/W (1)/Zr samples with different d_{Zr} , respectively, measured by a THz transmission spectroscopy system. It is observed that as d_{Zr} increases, the THz amplitude transmitted through the samples gradually decreases. This indicates that samples with larger d_{Zr} exhibit larger absorption of THz pulses. For quantitative analysis, the time-domain signals transmitted through the samples are Fourier transformed into frequency-domain spectra, $E_{sam}(\omega)$. The spectrum of THz transmission through the Al_2O_3 substrate serves as a reference, $E_{sub}(\omega)$.

The THz transmission spectra $\frac{E_{sam}(\omega)}{E_{sub}(\omega)}$ in the frequency range of 0.1 to 2.0 THz are obtained, as shown in **Figure S6** (b) and (d). The transmittance at 1 THz frequency as a function of d_{Zr} are shown in the insets of **Figure S6** (a) and (c). With increasing d_{Zr} , transmission @ 1 THz exhibits linearly decreases. Specifically, the transmission @ 1 THz of Co/Zr decreases from 96% to 51%, when the thickness increases from 3.0 nm to 20.0 nm. The transmission @ 1 THz of Co/W (1)/Zr heterostructures ranges from 88% to 39%, when the thickness increases from 3.0 nm to 20.0 nm.

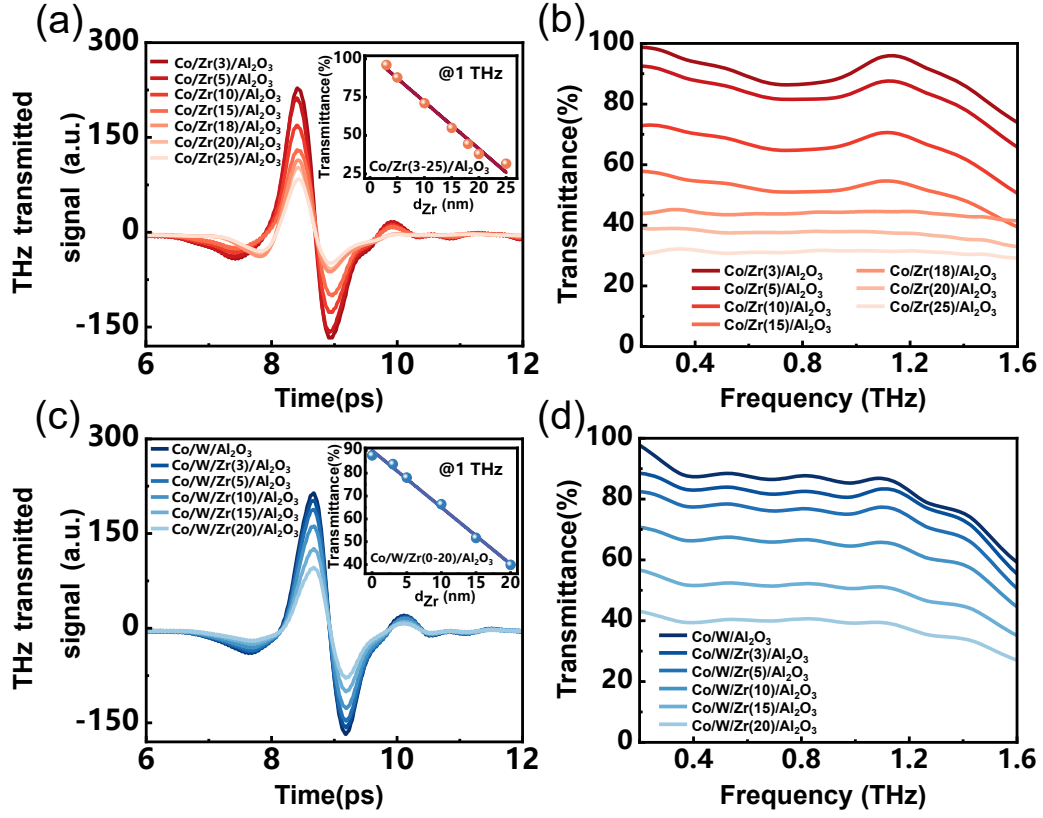


Figure S6 (a) and (c) The THz transmitted time-domain waveforms through Co/Zr/Al₂O₃ and Co/W (1)/ZrAl₂O₃ heterostructures with different d_{Zr} , respectively. (b) and (d) The THz transmission coefficients of Co/Zr/Al₂O₃ bilayer and Co/W (1)/Zr/Al₂O₃ trilayer heterostructures with different d_{Zr} in the frequency range between 0.2 to 1.6 THz.

We employed the THz transmission spectroscopy to measure the electrical conductivity of the heterostructures. We measured the THz transient signal of the bare substrate and the substrate coated with Co/Zr/Al₂O₃ and Co/W(1)/Zr/Al₂O₃ heterostructures. By measurement the THz transmission through the sample relative to that through the bare substrate, the impedance of the thin film is given by using the thin-film approximation,

$$Z_{sam}(\omega) = \frac{Z_0}{n_1(\omega) + n_2(\omega)} \frac{E_{sam}(\omega)}{E_{sub}(\omega)} \quad (S1)$$

where Z_0 is the vacuum impedance, $n_1(\omega)$ and $n_2(\omega)=1$ are refractive indices of the substrate and the air, respectively. The THz refractive index $n_1(\omega)$ of Al₂O₃ substrate was also measured by the THz-TDS system, as shown in **Figure S7**.

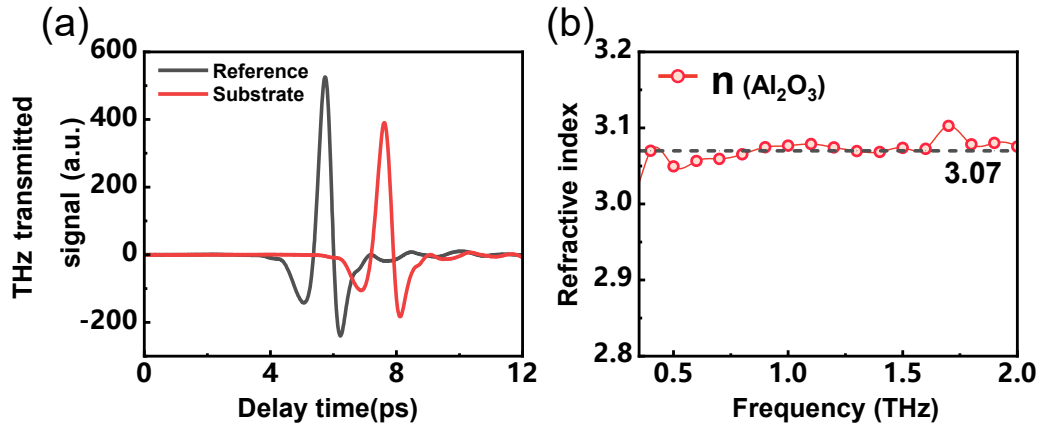


Figure S7 (a) THz time-domain spectra of the reference (black line) and the substrate signal (red line). (b) Refractive index of sapphire substrate in a frequency range of 0.2 - 2.0 THz.

Figure S7 (a) shows the THz time-domain signals of the reference and the substrate. The calculation result is shown in **Figure S7** (b). The average refractive index of the sapphire substrate is about 3.070 ± 0.003 .

Figure S8 (a) shows the measured impedance of the sample Co/Zr (3-20)/Al₂O₃ heterostructures. **Figure S8** (b) shows the measured Z @ 1 THz as a function of the thickness of Zr layer. The mean values of $Z(\omega/2\pi)$ for different thickness of Zr are used in the normalization of THz emission amplitude.

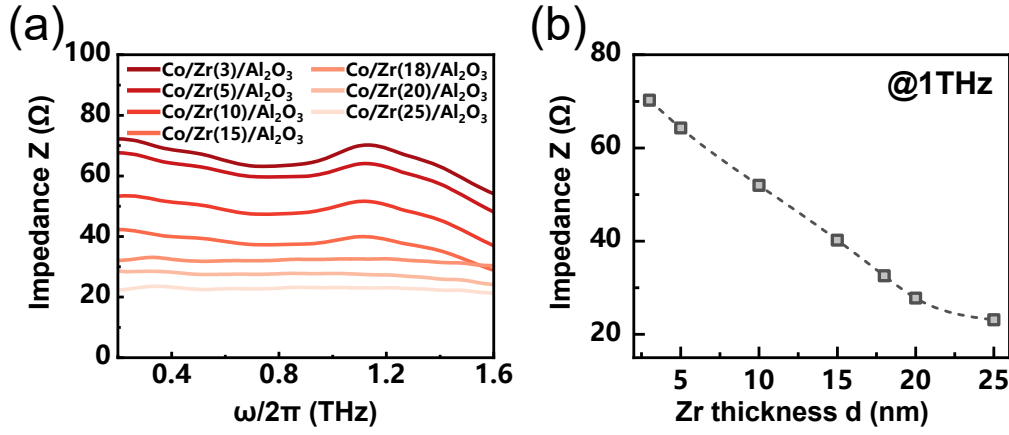


Figure S8 (a) Measured frequency dependent THz impedance $Z(\omega/2\pi)$ for Co/Zr (3-25)/ Al_2O_3 heterostructures. (b) The dependence of THz impedance Z on d_{Zr} at a frequency of 1 THz.

The conductivity of the heterostructures at THz frequencies using Tinkham's formulas,

$$\frac{E_{\text{sam}}(\omega)}{E_{\text{sub}}(\omega)} = \frac{n_1(\omega) + n_2(\omega)}{n_1(\omega) + n_2(\omega) + Z_0 \sigma(\omega) d} \quad (\text{S2})$$

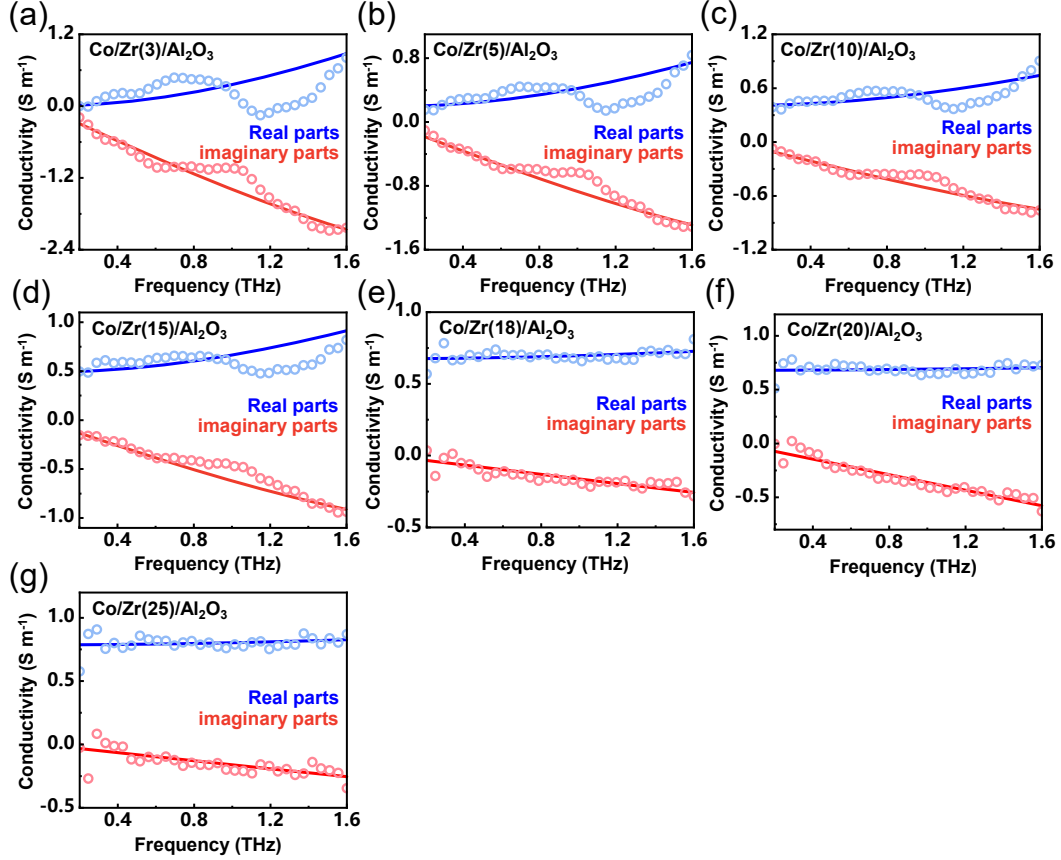


Figure S9 (a)–(g) The real (blue) and imaginary (red) parts of the conductivity for Co/Zr/Al₂O₃ heterostructures with varying d_{Zr} , as measured by THz-TDS. The circle symbols represent the experimental data, while the solid lines correspond to the fitted curves.

Figure S9 shows the real and imaginary parts of the optical conductivity for Co/Zr with different thickness. The real conductivity is positive, while its imaginary part is negative and decreases with frequency, that is attributed to the interplay of backscattering, quantum confinement, and carrier localization.

To quantitatively analyze the measured conductivity trends, we use the Drude-Smith model, expressed as follows:

$$\tilde{\sigma}_{\text{sheet}}(\omega) = \frac{\sigma_{\text{DC}}}{1 - i\omega\tau} \left[1 + \frac{c}{1 - i\omega\tau} \right] \quad (\text{S3})$$

where, σ_{DC} is the DC conductivity, τ is the momentum scattering time. Drude-Smith model is based on the dispersive free charge transport in a disordered medium. The carriers retain part of their initial velocity upon scattering. $c = -1$ and 0 represent the

carrier's momentum is completely backscattered and randomized, respectively. **Figure S9** show the experimental results can be fitted very well using the Drude-Smith model [3]. The parameters obtained from fitting using the Drude-Smith model are listed in Table S1.

Table S1 The fitting parameters of Drude-Smith model.

Sample	τ (fs)	σ_{dc} (S m ⁻¹)	c
Co/Zr(3)/Al ₂ O ₃	20	6.4×10^3	-0.99
Co/Zr(5)/Al ₂ O ₃	20.1	1.94×10^5	-0.97
Co/Zr(10)/Al ₂ O ₃	20.3	4.05×10^5	-0.92
Co/Zr(15)/Al ₂ O ₃	20.8	4.91×10^5	-0.92
Co/Zr(18)/Al ₂ O ₃	20.8	6.75×10^5	-0.83
Co/Zr(20)/Al ₂ O ₃	20.6	6.8×10^5	-0.84
Co/Zr(25)/Al ₂ O ₃	21.3	7.86×10^5	-0.85

Supplementary Note 7. Normalization processing of THz Pulses.

The normalized THz pulses in the article are obtained by dividing the original THz pulses by the absorbance of the sample to the laser (A_{FM}) and the THz pulse impedance (Z). Here, A_{FM} is defined by Equation S4:

$$A_{FM} = A_{total} \frac{t_{Co}}{t_{total}} \quad (S4)$$

Among them, A_{total} and t_{total} are respectively the total laser absorbance and total thickness of the bilayer or trilayer heterostructure, and t_{FM} is the thickness of the FM layer.

For the bilayer heterostructure, Z is represented by Equation S5; for the trilayer heterostructure, Z is represented by Equation S6:

$$Z = \frac{Z_0}{1 + n + Z_0(\sigma_{Co}t_{Co} + \sigma_{Zr}t_{Zr})} \quad (S5)$$

$$Z = \frac{Z_0}{1 + n + Z_0(\sigma_{Co}t_{Co} + \sigma_Wt_W + \sigma_{Zr}t_{Zr})} \quad (S6)$$

The Z is obtained by measuring the terahertz transmittance of the sample and the substrate respectively through the THz-TDS system.

Finally, the normalized signal of the terahertz pulse is obtained by Equation S7:

$$E = \frac{E_{THz}}{A_{FM}Z} \quad (S7)$$

As shown in **Figure S10** (a) and (b), we provide the original raw data and the normalized data of the terahertz pulses emitted by the Co/W (1)/Zr (3)/Al₂O₃ bilayer heterostructure.

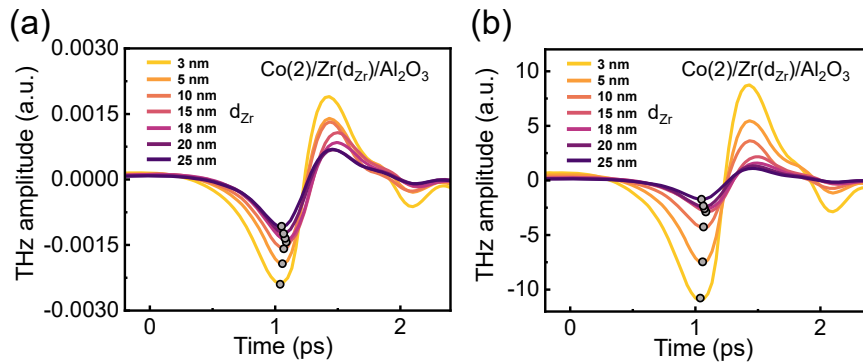


Figure S10 (a) The raw data of THz pulses emitted by the Co/W (1)/Zr(3)/Al₂O₃ heterostructure (b) the normalized data of the THz pulses

Supplementary Note 8. Laser absorption measurement.

We measured the transmitted/reflected/absorbed power of Co/Zr (d_{Zr})/Al₂O₃ heterostructure. Table S2 shows the experimental values of ratios of transmitted, reflected, and absorbed light power for different Zr thicknesses.

Table S2 The experimental values of ratios of transmitted/reflected/absorbed light power for different Zr thicknesses.

Zr thickness (nm)	Transmitted ratio (%)	Reflected ratio (%)	Absorption ratio (%)
3.0	50	16.7	33.3
5.0	42.7	21.3	36
10.0	28.5	30	41.5
15.0	18.7	37.3	44
18.0	11.6	50	38.4
20.0	9.3	52.5	38.2
25.0	5	59.2	35.8

Supplementary Note 9. The Dependence of THz emissions on Pump fluence.

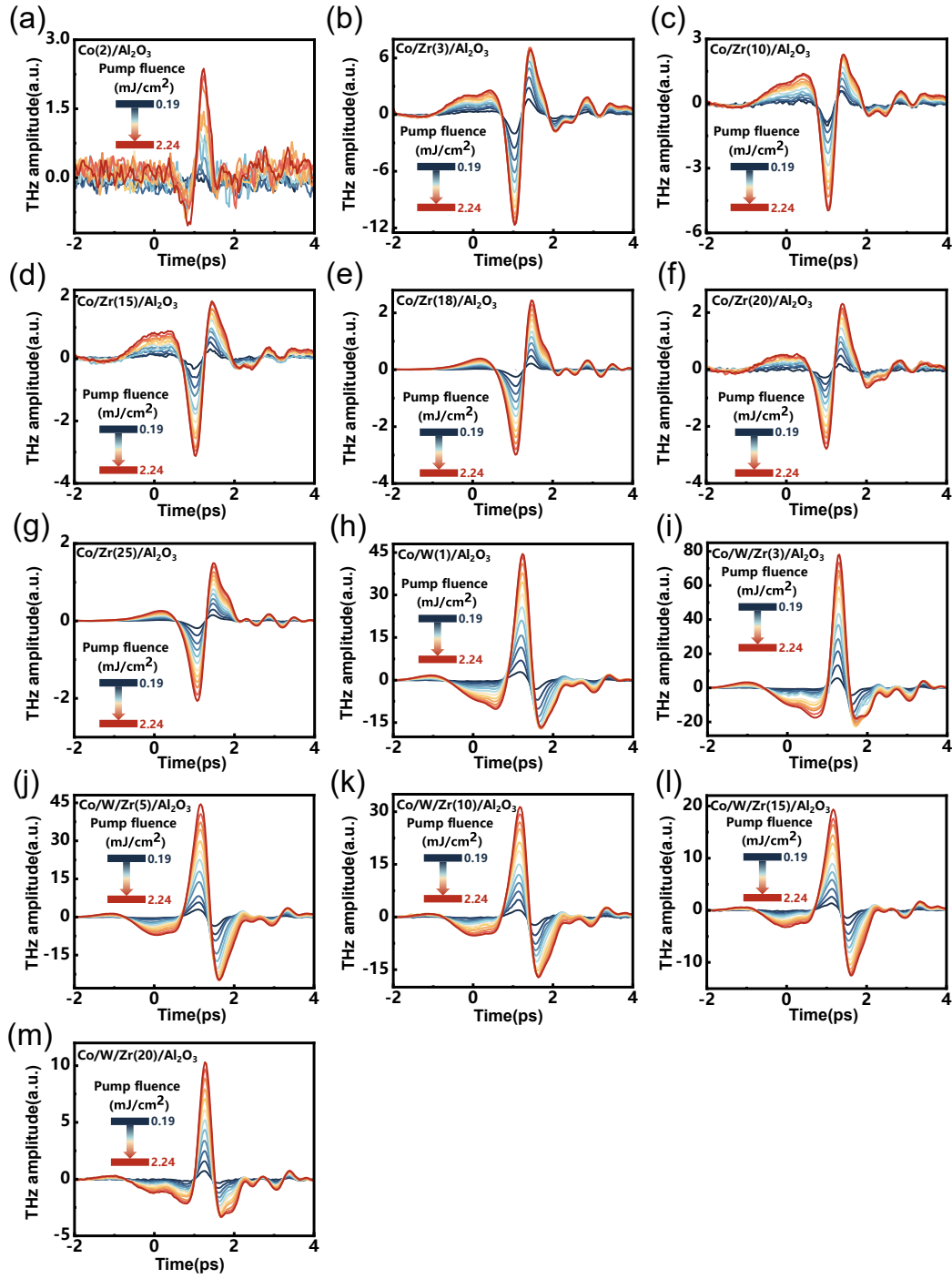
Figure S11 (a) shows the THz emission from single-layer Co (2) measured in the pump fluence range of 0.19-2.24 mJ cm⁻².

Figure S11 (b)-(g) show the THz emission from Co/Zr/Al₂O₃ heterostructures with varying d_{Zr} under different pump fluences.

Figure S11 (h) shows the THz emission from Co (2)/W (1) measured in the pump fluence range of 0.19-2.24 mJ cm⁻².

Figure S11 (i)-(m) show the THz emission from Co/W(1)/Zr/Al₂O₃ heterostructures with varying d_{Zr} under different pump fluences.

The THz amplitude emitted from all samples in **Figure S11** increases with increasing pump fluence in the pump fluence range (0.19-2.24 mJ cm⁻²).



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218 **Figure S11** The pump fluence dependent THz emission waveforms for (a) Co(2)/Al₂O₃,
 219 (b)-(g) Co/Zr(d_{Zr} = 3, 10, 15, 18, 20, 25 nm)/Al₂O₃ and (h) Co/W (1)/Al₂O₃, (i)-(m)
 220 Co/W(1)/Zr (d_{Zr} = 3, 5, 10, 15, 20 nm)/Al₂O₃ heterostructures. The pump fluence range
 221 between 0.19 and 2.24 mJ cm⁻².

Supplementary Note 10. The temperature dependence of the THz emission from Co (2)/Zr (3 and 5)/Al₂O₃ heterostructures.

Figure S12 (a) and (b) display the THz signals emitted by Co (2)/Zr (3 and 5)/Al₂O₃ heterostructures at various temperatures, ranging from 20 K to 300 K. As the sample temperature increases, the amplitude of the THz signals gradually decreases. Additionally, the time-domain signals exhibit a noticeable time delay.

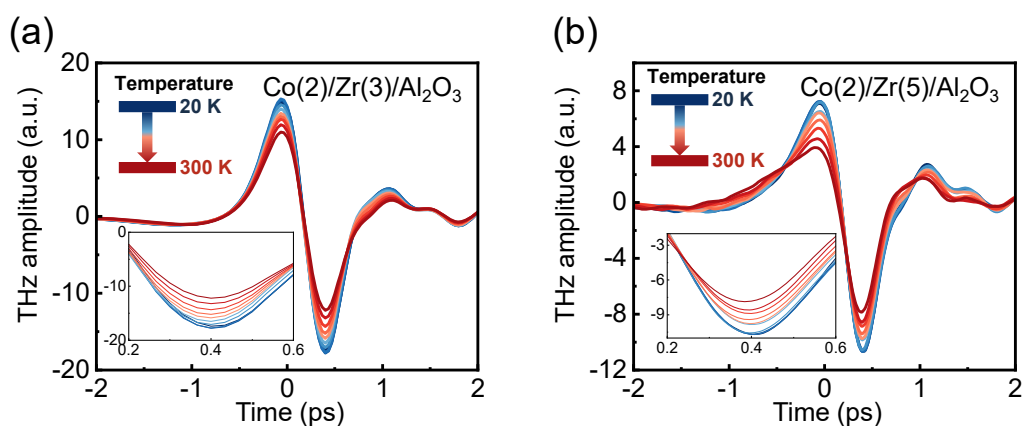


Figure S12 (a) and (b) THz time-domain signals emitted from Co (2)/Zr (3)/Al₂O₃ and Co(2)/Zr(5)/Al₂O₃ heterostructures, respectively, measured at different temperatures. The temperature range spans from 20 K to 300 K, with the waveforms' color transitioning from blue to red. Insets display magnified views of the peak signal.

234 **Supplementary References**

- 235 1. Huisman, T. & Mikhaylovskiy, R. et al. Femtosecond control of electric currents in
236 metallic ferromagnetic heterostructures. *Nature Nanotech* **11**, 455 – 458 (2016).
- 237 2. Jechumtál, J. et al. Accessing Ultrafast Spin-Transport Dynamics in Copper Using
238 Broadband Terahertz Spectroscopy. *Phys. Rev. Lett.* **132**, 226703243 (2024).
- 239 3. Smith, N. Classical generalization of the Drude formula for the optical conductivity.
240 *Phys. Rev. B* **64**, 155106 (2001).