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



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The utilization of terahertz (THz) emission spectroscopy in femtosecond photoexcited spintronic heterostructures has emerged as a versatile tool for investigating ultrafast spin-transport in a non-contact and non-invasive manner. However, the investigation of ultrafast orbital-transport is still in the primitive stage. Here, we experimentally demonstrate the orbital-to-charge current conversion in Co/Zr/Al₂O₃ heterostructures. Our experimental results indicate a photoinduced orbital current (J_L) from Co propagating through Zr layer with a long-distance ballistic transport and a velocity of $\sim 0.27 \pm 0.02$ nm fs⁻¹. On the one hand, we demonstrate a critical pump fluence required to overcome the collisions in orbital transport, enabling a swifter flow of J_L . On the other hand, a critical temperature is observed, below which the orbital transport is impeded. Finally, we observe a nearly 2.95-fold enhancement in the THz emission due to an additional conversion of the spin-to-orbital current conversion from a 1 nm thick W-insertion layer between Co and Zr layers. Our results pave the way for designing promising opto-spin-orbitronic devices and THz emitters.

The spin-to-charge-current conversion (SCC) has been extensively explored in the field of spintronics, through the spin Hall effect (SHE) and the spin Rashba-Edelstein effect (SREE)^{1–5}. The inverse spin Hall effect (ISHE) and inverse spin Rashba-Edelstein effect (ISREE) are used for the detection of spin transport^{6–9}. The spin source can be one from either spin pumping¹⁰, spin Seebeck current¹¹ or optical excitation^{12,13}, etc. The ISHE and ISREE have paved the way for potential technological applications, including spin-orbit torque devices^{14–16} and spintronics broadband terahertz (THz) emitters^{17–22}. The conversion between spin and charge works better in heavy metals that have strong spin-orbit coupling (SOC), such as tantalum (Ta), tungsten (W), platinum (Pt), and so on^{17,18}.

Actually, electrons carry two distinct angular momenta, the spin angular momentum (S) and orbital angular momentum (L)^{23,24}. Manipulating the magnetic moment produced by the L instead of S can bypass the

requirement for materials possessing robust SOC. The research field of encoding data through orbitals has been referred to orbitronics, which has the potential to develop advanced electronic devices²⁵. The L -induced orbital Hall effect (OHE) can convert the charge current (J_C) into orbital current (J_L) in the transverse direction with an applied external electric field²⁶. Compared with the SHE, the OHE has several technological advantages. Firstly, the conversion between J_L and J_C does not require materials with the strong SOC, thereby expanding the material range to include light metals with weak SOC. Secondly, the theoretical calculations have shown that the orbital Hall conductivity (OHC) is considerably larger than the spin Hall conductivity (SHC) in numerous materials, as shown in Table 1^{27–41}. Therefore, the orbital torque can be efficiently induced by OHE^{42–45}. Thirdly, in many nonmagnetic materials, the orbital diffusion length surpasses the spin diffusion length. The Onsager reciprocity allows the inverse orbital Hall

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Table 1 | Comparison of spin conductivity (σ_{SH}), orbital conductivity (σ_{OH}), spin diffusion length (λ_{sd}) and orbital diffusion length (λ_{od}) in different materials

Materials	$\sigma_{SH}(\Omega^{-1} \cdot \text{cm}^{-1})$	$\sigma_{OH}(\Omega^{-1} \cdot \text{cm}^{-1})$	$\lambda_{sd}(\text{nm})$	$\lambda_{od}(\text{nm})$
W	-324 ²⁷ -83 ²⁸	+3293 ²⁷ +4490 ²⁸	4 ± 1 ³³	\
Pt	+2152 ²⁹ +1739 ²⁸ +815 ²⁷	+2919 ²⁹ +4330 ²⁸ +1918 ²⁷	1.4 ± 0.2 ³⁴ 3.4 ± 0.4 ³⁵ 8.0 ± 0.5 ³⁶	\
Ta	-274 ²⁹ -286 ²⁷ -7 ²⁸	+6803 ²⁹ +3820 ²⁷	4 ± 1 ³³	\
Ti	-40 ²³	+3800 ²³	\	74 ± 24 ²³
Cr	-130 ³⁰ -70 ³¹	+8200 ³⁰ +7000 ³¹	1.8 ± 0.6 ³⁷	6.1 ± 1.7 ³⁷ 6.6 ± 0.6 ³¹
Cu	+30 ³⁰	+1200 ³⁰	\	2.7 ⁴¹
Nb	-48 ²⁸ -117 ²⁷	+5930 ²⁸ +3641 ²⁷	3.1 ³⁸	\
Zr	-170 ³²	+5300 ³²	3.1 ± 1.3 ³² 3.68 ± 1.17 ³⁹	14.2 ± 0.8 ³⁹
Ni/Zr	\	\	\	9.66 ± 2.32 ³⁹
Ni/W/SiO₂	\	\	\	80 ⁴⁰
Ni/Cu/MgO	\	\	\	9 ⁴⁷
Co/Zr/Al₂O₃	\	\	\	7.0–15.6 (this work)

effect (IOHE) and inverse orbital Rashba-Edelstein effect (IOREE). These orbit-to-charge current conversion (LCC) effects can be used to probe the J_L .

Previous work has presented a theoretical model, which describes the orbital angular momentum of mobile charge carriers interacts with both spin degrees of freedom and the lattice via the crystal field potential^{46–48},

$$\frac{\partial}{\partial t} \langle L \rangle = \langle F^L \rangle + \langle \eta_{L-S} S \times L \rangle + \frac{1}{i\hbar} \langle [L, V_{CF}] \rangle \quad (1)$$

where the first term $\langle F^L \rangle$ is the orbital flux term, the second term $\langle \eta_{L-S} S \times L \rangle$ expresses a transfer of angular momentum between spin and orbital degrees of freedom, η_{L-S} is the spin-orbit correlation factor. The third term $\langle [L, V_{CF}] \rangle$ describes the angular momentum transfer between orbital and crystal lattice via the crystal field potential V_{CF} .

When the ferromagnetic (FM)/non-magnetic (NM) heterostructures are subjected to ultrafast photoexcitation, the ultrafast demagnetization occurs, resulting in a coupling between the local magnetic moment and the spin of nonlocalized conduction electrons via exchange interactions, and then generating spin current (J_S) in the FM layer^{17–19,49–51}. When CoFeB or Ni₈₁Fe₁₉ (Py) are chosen as the FM layer, because of small η_{L-S} , only the dominated J_S is converted into the transient transverse J_C after injection into the NM layer and radiates a THz pulse, as shown in Fig. 1a. The NM layer is typically chosen to be a heavy metal with large SOC (such as Pt, Au), $J_C \sim \theta_{SCC} J_S$, where θ_{SCC} describes the efficiency of SCC, including bulk ISHE and interfacial ISREE.

The orbital current can be generated by magnetization dynamics, the process can be referred to as orbital pumping and/or spin fluctuations^{24,52–54}. Orbital pumping relies on the presence of SOC in the microscopic mechanism. When we choose FM layer with strong SOC, such as Ni ($\eta_{L-S} = +4.55$), Co ($\eta_{L-S} = +2.91$), CoFe ($\eta_{L-S} = +1.92$) or CoPt^{40,41,48,55–59}, the J_S in the FM layer can partially transform into the ultrafast J_L due to their relatively large η_{L-S} . Thus, both J_S and J_L are injected into the adjacent NM layer. For $\eta_{L-S} < 0$ the polarizations of spin and orbital momentum are antiparallel, while for $\eta_{L-S} > 0$ the polarizations of spin and orbital momentum are parallel. As shown in Fig. 1b, we only plot the dominate conversion from J_L to J_C in light metals (such as Ti or Cu), $J_C \sim \theta_{LCC} J_L$, where θ_{LCC} describes the LCC efficiency. LCC includes both IOHE in bulk and IOREE at the interface. The signs of θ_{SCC} and θ_{LCC} determine the direction of J_C , and consequently the polarity of THz emission.

Most recently, the orbital current converted to charge current in Ti and Mn layers has been investigated via IOHE⁵⁷. In addition, the insertion of a W layer with high spin-orbit conversion efficiency was used to generate larger orbital currents from spin currents. However, the influence of thickness of the W insert layer on the whole spin-orbitronic THz emitters has not been investigated. As described by Eq. (1), one of the greatest challenges is to understand how the orbital angular momentum of mobile charge carriers interacts with spins, photons, and phonons in different material platform, and how these interactions can be used in devices and technologies.

Zr is a 4d light metal for orbital transport due to its 4d²5s² orbital configuration, the unfilled *d* cell facilitates the efficient transportation of L ³². Although Zr has been used to generate the orbital torque³⁹, and further realizing the efficient reversal of the magnetic moment with perpendicular magnetic anisotropy⁶⁰. Both the spin-to-charge and orbit-to-charge conversions in Zr layer on ultrafast time scales are still unexplored. In addition, the active control of ultrafast J_L is not fully understood.

In this work, we provide experimental evidences for the conversion of a light-induced J_L from Co into the J_C by LCC within Co/Zr/Al₂O₃ and Co/W/Zr/Al₂O₃ heterostructures using THz emission spectroscopy. The Zr thickness dependency of THz emission indicates a long-distance ballistic transport of J_L within the Zr layer. In addition, it can be analyzed to extract information regarding the velocity and propagation characteristics of the ultrafast J_L . Note that an inset W layer can be used to manipulate the J_L from J_S , as illustrated in Fig. 1c. We demonstrate that an additional J_L channel from a 1 nm thick W insertion layer enhances the THz emission from Co(2)/W(1)/Zr(3)/Al₂O₃, which is 6.24 times larger than that from Co(2)/Zr(3)/Al₂O₃ with opposite polarity. The numbers in parentheses represent the thickness in nm. Furthermore, we demonstrate that both optical pumping fluence and temperature play crucial roles in actively controlling the ultrafast J_L in Co/Zr/Al₂O₃. Our work highlights the controllable and long-distance ballistic J_L transport in Co/Zr/Al₂O₃ heterostructures, which open up the possibilities for developing orbital-based information transfer in the THz frequency region and spin-orbitronic THz emission devices.

Results and Discussion

Samples and experiment

We fabricated the FM/NM heterostructures with the stack of Co(2)/Zr(3–25), Co(2)/W(1)/Zr(3–20), and Co(2)/W(1–5)/Zr(3) on the *c*-Al₂O₃ (0001) single crystal substrates and protected with a 5.0-nm MgO capping layer, by using

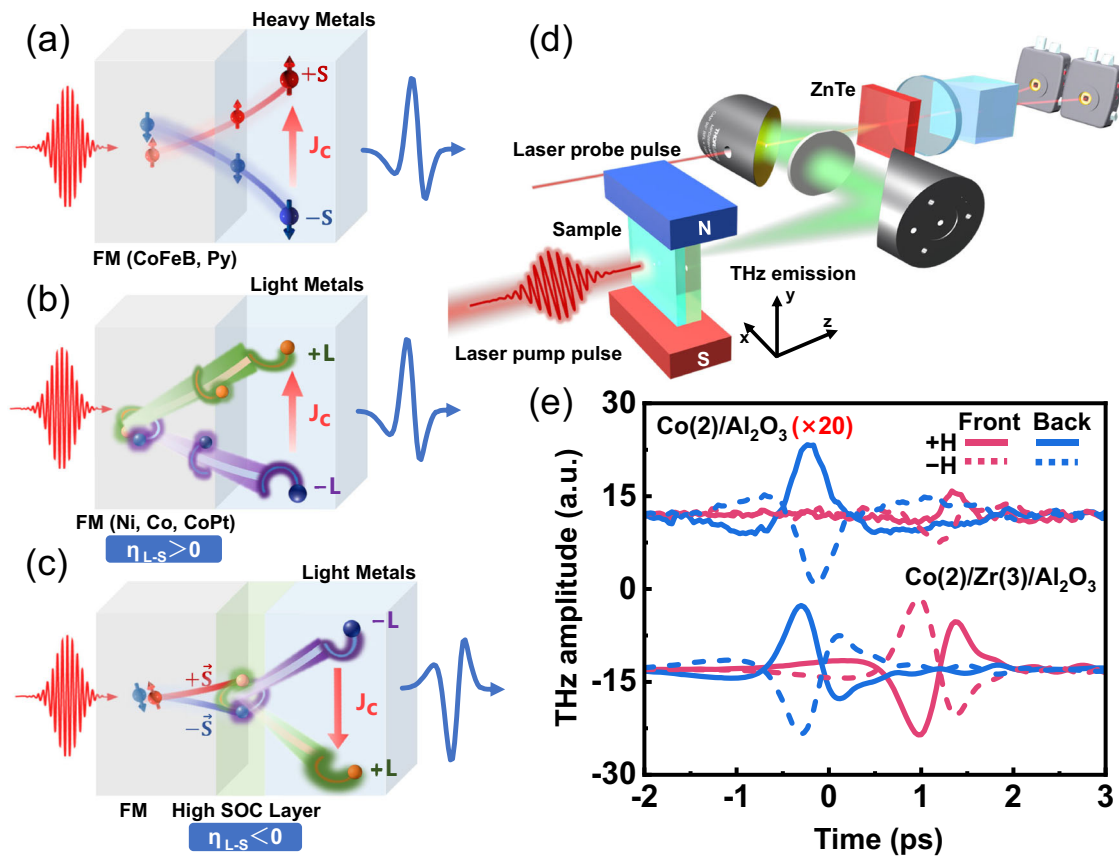


Fig. 1 | Ultrafast THz emission from Co(2)/Al₂O₃ and Co(2)/Zr(3)/Al₂O₃ heterostructure. **a** Schematic illustration of the spin-to-charge-current conversion (SCC) in ferromagnetic (FM) (such as CoFeB, Py)/heavy metal heterostructures. **b** Schematic illustration of the orbit-to-charge current conversion (LCC) in FM (such as Ni, Co, CoPt)/light metal heterostructures. **S** and **L** label the spin angular momentum and the orbital angular momentum, respectively. η_{L-S} is the spin-orbit correlation factor. **c** After the insertion of a high spin-orbit coupling layer, a synergistic effect of SCC and LCC modulates

the charge current J_c and THz emission. **d** Experimental schematics of THz emission spectroscopy and detection setup. Ultrafast laser pulses traveling along z -axis are incident on the sample, leading to the emission of broadband THz pulses, which are focused by two off-axis parabolic mirrors and detected by a 1.0-mm ZnTe (100) crystal. **e** THz emission signals from Co(2)/Zr(3)/Al₂O₃ and Co(2)/Al₂O₃ single-layer measured under front (red line)/back (blue line) excitations applied with external magnetic fields [+H (solid line) and -H (dash line)].

ultrahigh vacuum magnetron sputtering (see Methods for samples' details). The crystallinity of the heterostructures was confirmed by X-ray diffraction pattern (Supplementary Note 1). To investigate the generation and manipulation of ultrafast J_L in Co/Zr/Al₂O₃ and Co/W/Zr/Al₂O₃ heterostructures, we conducted the THz emission spectroscopy measurements, as shown in Fig. 1d. In brief, a train of linearly polarized ultrashort laser pulses (duration 35 fs, center photo-energy of 1.55 eV, repetition rate of 1 kHz) from a Ti:sapphire laser amplifier was used to excite Co/Zr/Al₂O₃ and Co/W/Zr/Al₂O₃ heterostructures. An in-plane magnetic field of $H = \pm 100$ mT was applied to keep the samples in the saturated magnetization state. The emitted THz pulse propagates in the dry-air atmosphere and is detected as an electrooptical sampling (EOS) signal by a 1.0-mm ZnTe (100) crystal (see Methods for THz emission-setup details).

THz emission from Co(2)/Zr(3)/Al₂O₃ and Co(2)/Al₂O₃

Figure 1e shows the THz emission signals from both Co(2)/Zr(3)/Al₂O₃ and Co(2)/Al₂O₃ single-layer. We note that the polarity of THz signal remains unchanged when the Co(2)/Al₂O₃ single-layer is flipped, which is consistent with the ultrafast demagnetization⁴⁹ and rule out the anomalous Hall effect^{61,62} and the anomalous Nernst effect⁶³ as the dominated THz emission mechanism. In contrast, when the Co(2)/Zr(3)/Al₂O₃ sample is flipped from front-side to back-side excitation, the THz emission signals' polarities are reversed, under a fixed H (Supplementary Note 2). In addition, the polarities of THz emissions are inverted by reversing the H , while keeping the optical excitation and detection geometry unchanged (Supplementary

Note 3). The findings demonstrate that the THz emission from Co/Zr/Al₂O₃ is different from the ultrafast magnetic dipole contribution. We also checked the measurements with linearly- and circularly-polarized pump pulses, which reveal a negligible impact on the THz emission (Supplementary Note 4). All the experimental results demonstrate that the electric dipole contribution is the dominate THz emission mechanism in Co/Zr/Al₂O₃ rather than the magnetic dipole contribution. As shown in Table 1, the calculated OHC $\sigma_{OH} \approx 5300$ ($\hbar \text{ e}^{-1}$) ($\Omega\text{-cm}$)⁻¹ is one order higher than SHC $\sigma_{SH} \approx -170$ ($\hbar \text{ e}^{-1}$) ($\Omega\text{-cm}$)^{-1 32,39}. Therefore, the THz generation from Co/Zr/Al₂O₃ should consists of two electric dipole contributions from both LCC and SCC mechanisms.

Generation of light-induced orbital transport in Co/Zr/Al₂O₃

To further identify the THz emission mechanism from Co/Zr/Al₂O₃ to be both LCC and SCC, as shown in Fig. 2a, we investigate the Zr thickness-dependent THz waveforms in the time-domain (frequency spectra are shown in Supplementary Note 5). Figure 2b plots the time delay τ_D in the peak of the emitted THz pulse as a function of the thickness of Zr, d_{Zr} . The observed τ_D is an experimental indication of J_L transport through the Zr layer rather than J_S transport⁶⁴. τ_D is related to the orbital carrier flying time through the Zr layer. When d_{Zr} increases from 3.0 nm to 15.0 nm, the τ_D shifts by approximately ~65 fs. In this thickness range, the τ_D nonlinearly increases with d_{Zr} associated with a concave shape. As the red curve fitting in Fig. 2b, $\tau_D \propto (d_{Zr})^\alpha$ with $\alpha \approx 0.495$, which rules out the diffusive regime ($\alpha = 2$), pure ballistic nature of long-distance ballistic transport ($\alpha = 1$)⁴⁰, and

Fig. 2 | Orbital-to-charge current (LCC) and spin-to-charge current (SCC) conversions-based THz emission from Co/Zr(d_{Zr})/Al₂O₃ heterostructures. **a** shows the THz emission from Co(2)/Zr(d_{Zr})/Al₂O₃ heterostructures with different Zr thicknesses. The marked small circles indicate a non-monotonic shift in THz peak as the thickness of Zr increased. **b** shows the time delay (τ_D) as a function of d_{Zr} measured at pump fluence of 1.87 mJ cm⁻². The dashed curves are calculated by Eq. (2), with an orbital diffusion velocity $v_0 = 0.27 \pm 0.02$ nm fs⁻¹ and different orbital flip times τ_{of} in the range of 100 fs to 500 fs. **c** shows the normalized peak-to-peak values of THz waveforms (ΔPK) by $Z^d A_F^d$ as a function of d_{Zr} . **d** shows the full width at half maxima (FWHM) of the THz pulses for Co/Zr/Al₂O₃ heterostructures with different d_{Zr} . Inset shows the measured FWHM of a THz waveform. The dashed purple curve is guides to the eye. **e** Conception diagram of the THz emission from different channels of conversions within Co/Zr/Al₂O₃: (1) the spin current J_S generated from the ferromagnetic (FM) layer is converted into charge current $J_{\text{C,SCC}}$ (2) the orbital current J_L is converted into charge current $J_{\text{C,LCC}}$. $J_L(0)$ is the charge current generated at the interface of Co and Zr. The error bars in **b**, **d** are the standard deviations calculated from multiple measurements and fits.

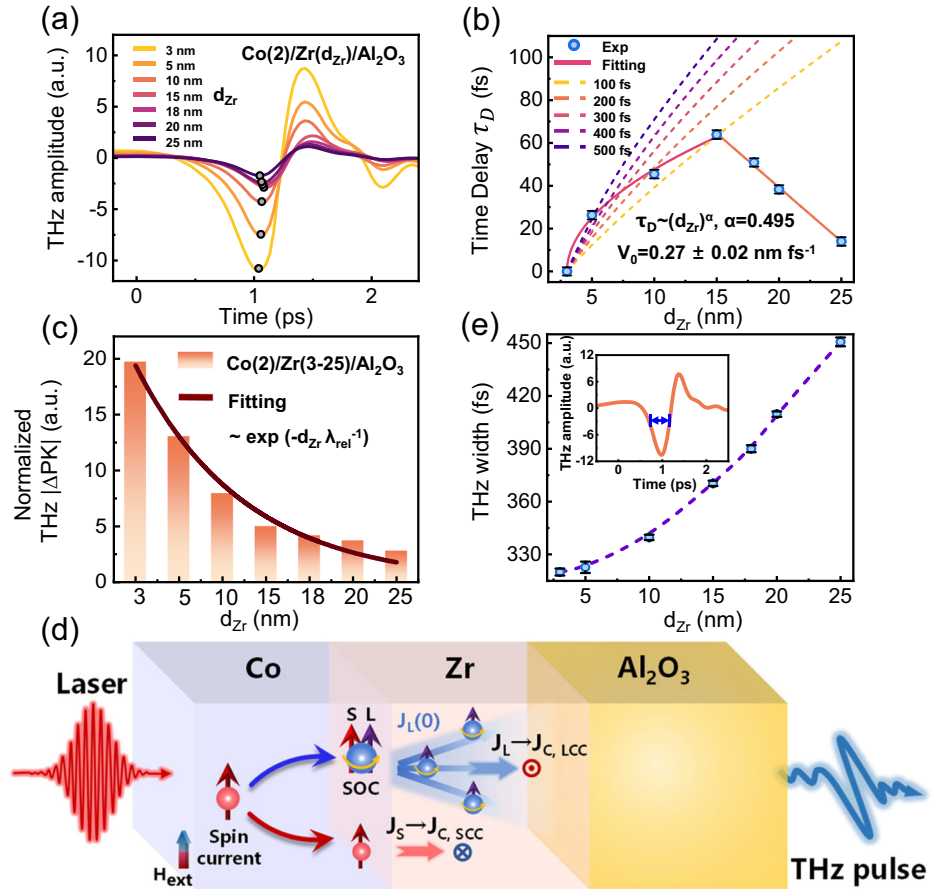


Table 2 | Comparison of orbital velocity and orbital diffusion length in different heterostructures

Heterostructures	Orbital velocity (nm fs ⁻¹)	Orbital diffusion length (nm)
SiO ₂ (substrate)/Ni/W/SiO ₂ (capping layer)	0.14 ⁴⁰	80 ⁴⁰
SiO ₂ (substrate)/CoPt/Cu/MgO(capping layer)	0.15 ⁴¹	2.7 ⁴¹
SiO ₂ (substrate)/Ni/Pt	0.14 ~ 0.18 ⁴⁸	18 ⁴⁸
MgO/Cu/Ni/MgO/Ta(capping layer)	0.26 ± 0.08 ⁵⁵	9.0 ⁵⁵
Al ₂ O ₃ (substrate)/Zr/Co/MgO(capping layer)	0.27 ± 0.02	7.0 ~ 15.6

super-diffusive regime ($1 < \alpha < 2$). Therefore, $\alpha \approx 0.495$ indicates a ballistic transport regime accompanied by a long decoherence process (see Methods for Models of orbital carrier transport)^{39,55}.

To better quantitatively understand the concave shape of τ_D as a function of d_{Zr} , we used the formalism presented in⁵⁵, by introducing a probability of orbital flip or orbital decoherence, to express the τ_D :

$$\tau_D = \frac{\int_{t_0}^{\infty} dt \exp\left(-\frac{t}{\tau_{\text{of}}}\right)}{\int_{t_0}^{\infty} \frac{dt}{t} \exp\left(-\frac{t}{\tau_{\text{of}}}\right)} \quad (2)$$

where the τ_{of} describes the orbital flip time. $t_0 = \frac{d_{\text{Zr}}}{v_0}$, v_0 is the group velocity of the orbital carriers. v_0 and τ_{of} can be obtained from the fitting of $\tau_D(d_{\text{Zr}})$, yielding $v_0 = 0.27 \pm 0.02$ nm fs⁻¹ and τ_{of} is in the range of 100 fs to 500 fs, as shown in Fig. 2b. In the ballistic transport regime of THz experiments, the orbital diffusion length can further be expressed in the usual diffusive picture of dc orbital currents:

$$l_{\text{of}} = v_0 \times \sqrt{\tau_{\text{of}} \times \frac{\tau}{3}} \quad (3)$$

where τ is the typical momentum relaxation time constant. We used THz time-domain spectroscopy (THz-TDS) to measure the electronic conductivity spectra of Co(2)/Zr(3-25)/Al₂O₃ (see Methods for THz time-domain spectroscopy). Using the Drude-Smith model, we extract the momentum relaxation time τ at room temperature is approximately 20 fs^{65,66} (Supplementary Note 6). Therefore, the orbital diffusion length of Zr can be estimated to be in the range of 7.0~15.6 nm, which agrees with the value of 14.2 ± 0.8 nm measured by spin torque ferromagnetic resonance (ST-FMR)³⁹. Co/Zr/Al₂O₃ exhibits comparable and high orbital velocity, compared with the values of 0.26 ± 0.08 nm fs⁻¹ and 0.14 nm fs⁻¹ for Ni/Cu/MgO and Ni/W/SiO₂, respectively, as summarized in Table 2. Note that, when d_{Zr} increases beyond 15.0 nm, which is larger than the orbital diffusion length, τ_D exhibits a rapid decrease. This experimental observation can be interpreted as a crossover from the LCC-dominant THz emission (with time-delay) to the SCC-dominant THz emission (without time-delay), which also agrees with the recent findings in the literature⁴¹.

To further understand the characterization of LCC, the THz peak amplitudes are normalized by $Z^d A_F^d$, where Z^d is the THz radiation impedance of the sample (Supplementary Note 7) and A_F^d is the incident pump pulse absorbance (Supplementary Note 8). The normalized THz signal is proportional to the efficiency of spin/orbit-charge conversion. As shown in

Fig. 3 | Manipulation of ultrafast ballistic orbital transport. **a** THz emission signals from Co(2)/Zr(3)/Al₂O₃ measured at pump fluences in the range of 0.19–2.24 mJ cm⁻². **b** The peak-to-peak amplitudes (ΔPK) of THz emission for Co(2)/Zr(3)/Al₂O₃ and Co(2)/Zr(5)/Al₂O₃ as functions of pump fluence. **c** The relative peak-to-peak durations for Co(2)/Zr(d_{Zr} = 3, 5, 10, 15, 18, 20, 25)/Al₂O₃ as functions of pump fluences. The peak-to-peak durations measured at pump fluence of 0.19 mJ cm⁻² were taken as the baselines. **d** The critical pump fluence with variation in d_{Zr} of Co(2)/Zr/Al₂O₃ and Co(2)/W(1)/Zr/Al₂O₃. **e** and **f** show ΔPK and relative peak-to-peak durations as functions of sample's temperature, respectively. The peak-to-peak durations measured at 20 K were taken as the baselines. The error bars in **e** are the standard deviations calculated from multiple measurements.

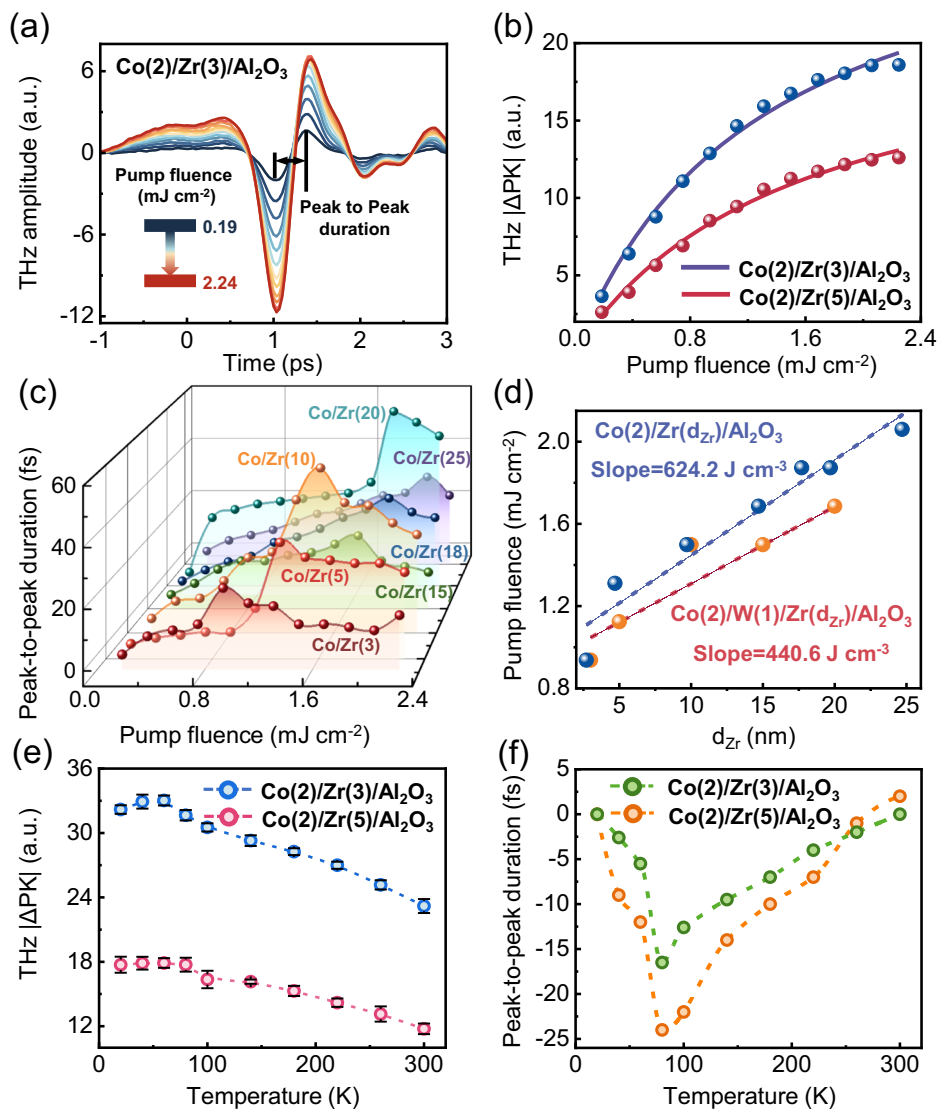


Fig. 2c, the peak of normalized ΔPK abruptly reaches its maximum as $d_{Zr} = 3.0$ nm and then single-exponentially decreases with a relaxation length constant of $\lambda_{rel} = 11.46 \pm 0.8$ nm. With increasing d_{Zr} , we do not see any progressive increase of active region for IOHE based THz emission, which indicates that the LCC cannot be attributed only to the IOHE. Figure 2d shows the FWHM of THz pulses, which broadens from 320 fs at $d_{Zr} = 3.0$ nm to 450 fs at $d_{Zr} = 25.0$ nm. A more pronounced angular dispersion of the J_L is observed in the thicker films. This visible chirping of THz pulse confirms the long-distance ballistic transport within the Zr layer⁶⁷. All these two observations indicate that both bulk IOHE and interfacial IOREE contribute to the LCC mechanism for THz emission in Co(2)/Zr(3–25)/Al₂O₃ heterostructures.

Modulation of light-induced orbital transport in Co/Zr/Al₂O₃

Based on Eq. (1), the orbital angular momentum can transfer between lattice and electrons⁴⁸, we make a further step to investigate the active modulation of orbital transport in the Zr layer by changing the pump fluence and temperature. Figure 3a shows the exemplary THz emission waveforms from Co(2)/Zr(3)/Al₂O₃ measured with different pump fluences. As shown in Fig. 3b, ΔPK values increase with increasing pump fluence, and start to saturate. The data are well fitted by the saturation formula (see Method for saturation model). The saturation fluence can be extracted $F_{sat} = 1.30$ mJ cm⁻² and 1.53 mJ cm⁻² for Co(2)/Zr(3)/Al₂O₃, and Co(2)/Zr(5)/Al₂O₃,

respectively. In order to quantitatively evaluate the deformation of THz waveform as the pump fluence increases, we took the peak-to-peak duration measured at 0.19 mJ cm⁻² as a baseline and subtracted it from measurements obtained at higher pump fluences. Figure 3c shows the relative peak-to-peak durations of the THz pulses for Co(2)/Zr(d_{Zr})/Al₂O₃, which increases proportionally with the increment in pump fluence, indicating a broadening deformation. When the pump fluence exceeds a certain critical fluence threshold, the waveforms of THz pulses tend to be narrow. This behavior was observed in all Co(2)/Zr(d_{Zr})/Al₂O₃ samples (Supplementary Note 9). The critical fluence threshold can be extracted as a function of d_{Zr} , as summarized in Fig. 3d. It can be found that the critical fluence threshold increases with d_{Zr} linearly.

These experimental results can be qualitatively interpreted. The increase in pump fluence increase the scattering probability of charge carriers, which perturbs the orbital wave function. Thus, more collisions impeded the orbit transport⁴⁸. As a consequence, the THz pulse experiences a chirp, resulting in waveform broadening deformation. When the pump fluence increases beyond a critical value, the non-equilibrium orbital wave function absorbs additional angular momentum from the lattice via the crystal field potential by $\frac{1}{i\hbar} \langle [L, V_{CF}] \rangle$ in Eq. (1). The enhancement of the J_L can surpass the collisions and facilitates a more efficient ballistic transport. As d_{Zr} increases, the orbital carriers require more energy to overcome the collisions and to move over a larger distance ballistically, which is indicated by the

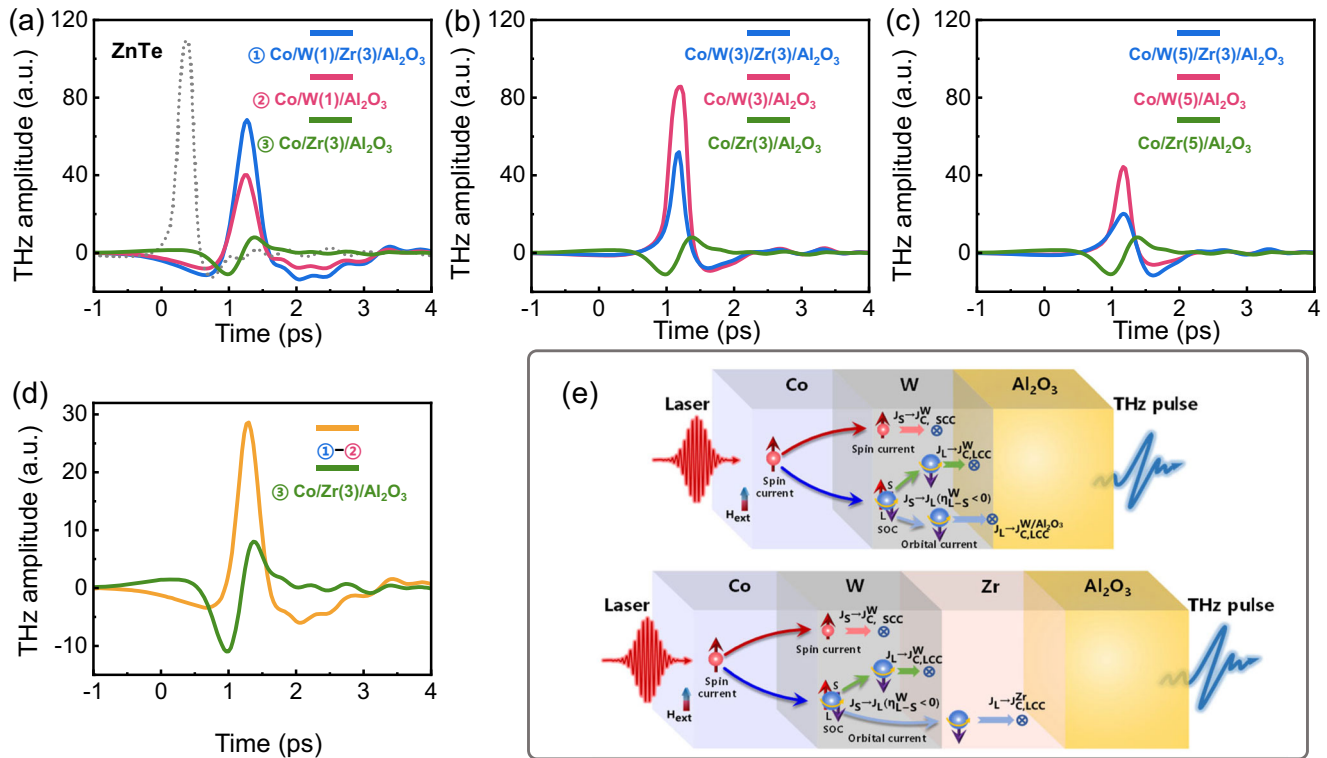


Fig. 4 | Modulation of THz emission assisted by additional spin-orbital current conversion. The time domain THz emissions from **a** Co(2)/W(1)/Zr(3)/Al₂O₃ and Co(2)/W(1)/Al₂O₃, **b** Co(2)/W(3)/Zr(3)/Al₂O₃ and Co(2)/W(3)/Al₂O₃, **c** Co(2)/W(5)/Zr(3)/Al₂O₃ and Co(2)/W(5)/Al₂O₃ compared with Co(2)/Zr(3)/Al₂O₃ and 1.0-mm thick ZnTe, measured under same conditions. **d** The extracted difference of THz waveforms between ① and ② in **a** compared with THz emission from Co(2)/Zr(3)/Al₂O₃. **e** Conception diagram of the THz emission from different channels in Co/W/Al₂O₃ and Co/W/Zr/Al₂O₃. For Co/W/Al₂O₃, (1) the spin current J_S

generated is converted into the charge current in W layer $J_{C,SCC}^W$, (2) the orbital current J_L is converted into the charge currents both $J_{C,LCC}^W$ in W layer and $J_{C,LCC}^{W/Al_2O_3}$ at the interface of W and Al₂O₃. For Co/W/Zr/Al₂O₃, (1) the spin current J_S generated is converted into the charge current in W layer $J_{C,SCC}^W$, (2) the orbital current J_L is converted into the charge currents both $J_{C,LCC}^W$ in W layer and $J_{C,LCC}^{Zr}$ in Zr layer. SCC and LCC label the spin-to-charge-current conversion and the orbit-to-charge current conversion, respectively.

larger critical pump fluence. In Fig. 3d, the slope of the linear correlation can be represented as a critical energy density, ϵ_c , which denotes the energy per unit volume required to overcome collisions and facilitates a ballistic orbital transport. ϵ_c is quantified as 624.2 J cm⁻³, which is larger than 343.7 J cm⁻³ in Ni/Pt heterostructure⁴⁸.

Inspired by the work of S. Kumar et al.⁵⁶, we aim to gain a deeper understanding of the angular momentum transfer between orbitals and crystal lattice. To achieve this, we have measured the THz emissions from Co(2)/Zr(3)/Al₂O₃ and Co(2)/Zr(5)/Al₂O₃ at low temperatures. The time-domain data are shown in Supplementary Note 10. In Fig. 3e, the ΔPK of two samples increase proportionally with the samples' temperature cooling down from 300 K to around 80 K. Below 80 K, ΔPK starts to be flat. In addition, as shown in Fig. 3f, the relative peak-to-peak durations narrow by approximately 16.5 fs and 24 fs, from 20 K to 80 K for Co(2)/Zr(3)/Al₂O₃ and Co(2)/Zr(10)/Al₂O₃, respectively. Above 80 K to 300 K, the relative peak-to-peak durations shift back to the similar value measured at around 20 K. The non-monotonic temperature dependences demonstrate that the orbital degree of angular momentum is strongly influenced by the temperature-sensitive crystal field potential. Our experimental results can be interpreted qualitatively. The drop of temperature (from 300 K to 80 K) decreases the scattering probability of charge carriers and the collision between orbital carriers and lattices, which facilitates more efficient orbit transport and results in the negative shift of durations by ~16.5 fs for Co(2)/Zr(3)/Al₂O₃ and ~24 fs for Co(2)/Zr(5)/Al₂O₃, respectively. While, at lower temperature below 80 K, on the one hand, the phonon freezing slows down both spin-lattice and orbit-lattice interactions. On the other hand, the charge carriers may transfer additional angular momentum back to the lattice, thereby impeding the

ballistic transport of orbit current, which leads to the positive shift of peak-to-peak duration. This semi-quantitative analysis could provide a qualitative way to understand the modulation of THz emission by the angular momentum transfer among spin, electron, orbital and crystal, with varying pump fluence and temperature. While, from the theoretical perspective, a more accurate quantum scattering model is desired to microscopically describe the dynamics and conversion of orbit and spin currents, which is required to interpret the pump fluence thresholds or the critical temperatures.

Finally, to further active modulate the orbital transport, a W layer with high-efficient SOC was inserted between Co and Zr layer. Figure 4a–c shows the typical time-domain waveforms of THz radiations from the Co(2)/W(1)/Zr(3)/Al₂O₃, Co(2)/W(3)/Zr(3)/Al₂O₃ and Co(2)/W(5)/Zr(3)/Al₂O₃ compared with Co(2)/W(1)/Al₂O₃, Co(2)/W(3)/Al₂O₃, Co(2)/W(5)/Al₂O₃ and Co(2)/Zr(3)/Al₂O₃, respectively, under the same front side excitation with +H.

We firstly found that the polarities of THz emissions from Co/W/Al₂O₃ are opposite to that from Co/Zr/Al₂O₃. Owing to the spin conductivity and the orbital conductivity of W layer have opposite sign, thus, the SCC and LCC induced currents should have opposite polarities. However, the spin-orbit correlation factor of W, $\eta_{L-S}^W < 0$ ⁴⁰. Thus, the mechanisms of SCC and LCC add up constructively, as shown in Fig. 4e, because they coincide with the same signs of $\theta_{SCC}^W < 0$ and $\eta_{L-S}^W \theta_{LCC}^W < 0$. Noted that the spin conductivity of Zr has the same polarity with that of W, if the THz emission mainly derive from the ISHE in Zr, the THz polarities of the Co(2)/Zr(3)/Al₂O₃ and Co(2)/W/Al₂O₃ should be the same. However, they are opposite, indicating that the LCC dominates the THz emission from the Co(2)/Zr(3)/Al₂O₃ heterostructures.

Secondly, we compare the THz emissions from Co/W/Zr/Al₂O₃ and Co/W/Al₂O₃. As shown in Fig. 4b and c, the THz emission from Co(2)/W(3)/Zr(3)/Al₂O₃ is nearly 60.7% of that from Co(2)/W(3)/Al₂O₃. The THz emission from Co(2)/W(5)/Zr(3)/Al₂O₃ is nearly 45.6% of that from Co(2)/W(5)/Al₂O₃. The experimental results can be interpreted, as shown by the schematic diagrams in Fig. 4e. In the Co/W/Al₂O₃, the J_S can be converted into $J_{C,SCC}^W$, while the J_L can be converted into both $J_{C,LCC}^W$ and $J_{C,LCC}^{W/Al_2O_3}$. The sum of $J_{C,SCC}^W + J_{C,LCC}^W + J_{C,LCC}^{W/Al_2O_3}$ is responsible for THz generation. While, in the Co/W/Zr/Al₂O₃, the Zr layer between W and Al₂O₃ can destroy the $J_{C,LCC}^{W/Al_2O_3}$ via interfacial IOREE, which would significantly reduce the THz emission.

Previous reports have demonstrated that an insertion W-insertion layer can provide an additional J_L . To investigate the LCC in the Zr layer, the thickness of W was decreased to 1 nm in the Co/W/Zr/Al₂O₃. As shown in Fig. 4a, the Co(2)/W(1)/Zr(3)/Al₂O₃ (marked as ①) produces nearly 1.7 and 6.24 times higher THz signal as compared to that from Co(2)/W(1)/Al₂O₃ (marked as ②) and Co(2)/Zr(3)/Al₂O₃ (marked as ③), respectively. Figure 4d shows $E_{Co(2)/W(1)/Zr(3)/Al_2O_3}^{THz} - E_{Co(2)/W(1)/Al_2O_3}^{THz}$ (marked as ①–②) and $E_{Co(2)/Zr(3)/Al_2O_3}^{THz}$ (marked as ③), which have an significant phase shift. In addition, the absolute peak value of $E_{Co(2)/W(1)/Zr(3)/Al_2O_3}^{THz} - E_{Co(2)/W(1)/Al_2O_3}^{THz}$ is 2.95 times larger than that of $E_{Co(2)/Zr(3)/Al_2O_3}^{THz}$. The role of the 1 nm W-insertion layer acts as an additional orbital current channel rather than simply as SCC and LCC effects itself. This additional orbital current $J_L^{Co/W}$ has an opposite sign compared with the J_L^{Co} of Co layer. As shown in Fig. 4e, the J_C produced in Co(2)/W(1)/Zr(3)/Al₂O₃ responsible for enhanced THz emission is the vectorial sum of the three dominating charge currents, expressed as $J_C \propto J_{C,SCC}^W + J_{C,LCC}^W + J_{C,LCC}^{Zr}$. Therefore, the 1 nm thick W is the optimized structure for THz generation from Co/W/Zr/Al₂O₃. As shown in Fig. 4a, the synergistic effect of both LCC and SCC enhances the THz generation signal from Co(2)/W(1)/Zr(3)/Al₂O₃, which can be almost as large as the emission from a 1.0-mm thick ZnTe crystal. The experimental setup and crystal's orientation are optimized to achieve the maximum THz emission signal from ZnTe. Noted that, the peak-to-peak duration of Co(2)/W(1)/Zr(d_{Zr})/Al₂O₃ has a similar pump fluence dependence with the Co(2)/Zr(d_{Zr})/Al₂O₃, and the ϵ_c is quantified as 440.6 J cm⁻³, as shown in Fig. 3d.

Conclusions

In summary, we have demonstrated the generation and manipulation of ultrafast J_L in Co/Zr/Al₂O₃ and Co/W/Zr/Al₂O₃ heterostructures excited by femtosecond laser pulses. The ballistic orbital transport through Zr propagates a long-distance with an orbital diffuse length in the range of 7.0–15.6 nm and a velocity of 0.27 ± 0.02 nm fs⁻¹. Furthermore, the orbital transport can be controlled by the incident femtosecond laser pulses and temperature. Usually, the increasing collision disperses the orbital transport. Beyond a critical pump fluence, carriers overcome the collisions, and the orbital transport occurs at a swifter pace. Below a critical temperature, the ballistic transport of orbit current is impeded. Finally, the Co(2)/W(1) bilayer turns out to be an efficient source of J_L , with efficiency associated with the inseting of the large SOC of 1 nm thick W between the Co and Zr layers. Zr is a candidate as LCC material, its large orbital conductivity and orbital velocity makes it a highly promising material for THz emission driven by ultrafast orbital currents. The spin-orbitronic THz sources can be assembled to the ready-to-use device for compact THz spectroscopy and communication applications^{68–71}.

Methods

Sample preparation

In this study, the multilayer films with a structure of Co(2)/Zr(3–25) were deposited on Al₂O₃ single crystal substrates via an ultrahigh vacuum magnetron sputtering system with a base pressure value lower than 5.0×10^{-8} Torr. During the deposition process, the Ar pressure was maintained at a constant level of 3 mTorr. The growth rate is 0.256 Å s⁻¹ for Zr and

0.206 Å s⁻¹ for Co, respectively. The Zr layer was deposited at 300 °C to obtain polycrystalline Zr layer. The MgO capping layer was 5.0 nm thick to protect the structure from oxidation. Figure S1 depicts XRD pattern of the polycrystalline Zr thin film on the Al₂O₃ substrate. We found two peaks at $2\theta = 33.06^\circ$ and 34.75° , corresponding to the (100) and (002) reflections of hcp-structured Zr, suggesting that Zr film is polycrystalline. In addition, the MgO/Co(2)/W(1,3,5)/Zr(3–20)/Al₂O₃ heterostructures were fabricated. The Co(2)/Al₂O₃ and Co(2)/W(1)/Al₂O₃ heterostructures were employed as the control samples. The thickness of the FM Co layer remains constant at 2.0 nm throughout the experiment.

THz emission setup

In the optical experiment, the system employs a titanium sapphire laser with a central wavelength of 800 nm, a duration of 35 fs, and a repetition frequency of 1 kHz, and a spot diameter of ~4 mm. As depicted in Fig. 1, the fs laser was split into two beams (8:2) by beam splitter. The high-power laser pump beam was collimated with the linearly polarized laser spot on the sample of 4 mm. A static in-plane magnetic field of $H_{ext} = 100$ mT was applied along the $\pm y$ direction to keep the system in the saturated magnetization state. A lock-in amplifier modulated by a chopper of ~490 Hz was used to measure the signal. A 1.0 mm thick ZnTe (110) crystal was utilized as the THz detector. Based on the electro-optic sampling method, the time-domain signal of the THz pulse was measured. For the electro-optic sampling, a quarter-wave plate and a Wollaston prism are employed to separate the s and p polarization of the probe beam. The changes in the probe beam are subsequently identified via a balanced photodiode that measures the intensity difference between the s and p-polarized beams. A lock-in amplifier is used to enhance the signal-to-noise ratio and to plot the THz electric field. All measurements were conducted at room temperature in a dry-air environment. The temperature-dependent THz time-domain waveform were measured in the temperature range from 20 K to 300 K (the temperature interval of 20 K between 20 K and 100 K, and 40 K between 100 K and 300 K).

THz time-domain spectroscopy (THz-TDS)

Broadband THz-TDS was carried out on the Co/Zr/Al₂O₃ and Co/W/Zr/Al₂O₃ heterostructures with the PCA-scanner system. A femtosecond laser pulse (70 mW, 100 fs duration, 1550 nm wavelength, 100 MHz repetition rate) was split into two beams: generation and detection laser beams. The generated laser beam has a power of 27.5 mW, whereas the detection laser beam has a power of 21.2 mW. The generated laser beam is focused onto a 100 μ m InGaAs antenna as the emitter. The generated THz beam is focused by a parabolic mirror and transmitted through a Co/Zr/Al₂O₃ and Co/W/Zr/Al₂O₃ heterostructures. The amplitude and phase of the transmitted THz pulses are detected by another InGaAs photoconductive switching, by scanning a variable optical delay stage. The dynamic range exceeds 80 dB. All static THz spectra were obtained in a room-temperature with dry air environment.

Models for orbital carrier transport

The delay time τ_D depends on the Zr thickness (d_{Zr}) and the group velocity of orbital carriers in the case of both ballistic and diffusive transports. (1) The ballistic transport occurs when the coherence time is short, resulting in a linear relationship between the traveling distance and delay time, $\tau_D \propto d_{Zr}$. (2) The ballistic transport is accompanied by a long decoherence process, where the τ_D increases with the distance according: $\tau_D \propto (d_{Zr})^\alpha$ with $\alpha < 1$. (3) For the diffusive transport regime, the τ_D increases with the d_{Zr} according: $\tau_D \propto (d_{Zr})^2$. (4) The super-diffusive regime corresponds to an intermediate between purely ballistic and diffusive regimes ($1 < \alpha < 2$), corresponding to the excitation of secondary source of electrons.

Model for THz saturation

The peak-to-peak value of THz signal ΔPK increases with an increasing pump fluence, and then reaches saturation. The experimental data of saturation effect can be fitted by the saturation formula $\Delta PK(F_P) = B \times F_P /$

$(F_p + F_{sat})^{72}$, where B characterizes the factors in terms of the THz radiation, F_p is the pump fluence and F_{sat} is the saturation fluence.

Data availability

The experimental raw data for this work can be found in Supplementary Data 1. All other relevant data are available from the corresponding author upon reasonable request.

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References

- Chen, X. et al. Observation of the antiferromagnetic spin Hall effect. *Nat. Mater.* **20**, 800 (2021).
- Xie, B. et al. Higher-order quantum spin Hall effect in a photonic crystal. *Nat. Commun.* **11**, 3768 (2020).
- Bindel, J. R. et al. Probing variations of the Rashba spin-orbit coupling at the nanometre scale. *Nat. Phys.* **12**, 920 (2016).
- Maguid, E. et al. Disorder-induced optical transition from spin Hall to random Rashba effect. *Science* **358**, 1411 (2017).
- Nakayama, H. et al. Molecular engineering of Rashba spin-charge converter. *Sci. Adv.* **4**, eaar3899 (2018).
- Wesenberg, D., Liu, T., Balzar, D., Wu, M. & Zink, B. L. Long-distance spin transport in a disordered magnetic insulator. *Nat. Phys.* **13**, 987 (2017).
- Zucchetti, C. et al. Doping dependence of the electron spin diffusion length in germanium. *APL Mater.* **7**, 101122 (2019).
- Huang, L. et al. Antiferromagnetic Inverse Spin Hall Effect. *Adv. Mater.* **34**, 2205988 (2022).
- Jeon, J. et al. Spin transport at a Pt/InAs quantum well interface using spin Hall and Rashba effects. *Appl. Phys. Lett.* **4**, 042403 (2020).
- Wang, Y. et al. Spin pumping during the antiferromagnetic-ferromagnetic phase transition of iron-rhodium. *Nat. Commun.* **11**, 275 (2020).
- Kimling, J. et al. Picosecond Spin Seebeck Effect. *Phys. Rev. Lett.* **118**, 057201 (2017).
- Gao, Y. et al. Photoexcitation induced magnetic phase transition and spin dynamics in antiferromagnetic MnPS₃ monolayer. *npj. Comput. Mater.* **9**, 107 (2023).
- Shokeen, V. et al. Spin Flips versus Spin Transport in Nonthermal Electrons Excited by Ultrashort Optical Pulses in Transition Metals. *Phys. Rev. Lett.* **119**, 107203 (2017).
- Safranski, C. et al. Spin-orbit torque driven by a planar Hall current. *Nat. Nanotech.* **14**, 27 (2019).
- Khang, N. H. D. et al. A conductive topological insulator with large spin Hall effect for ultralow power spin-orbit torque switching. *Nat. Mater.* **17**, 808 (2018).
- Hibino, Y. et al. Large Spin-Orbit-Torque Efficiency Generated by Spin Hall Effect in Paramagnetic Co-Ni-B Alloys. *Phys. Rev. Appl.* **14**, 064056 (2020).
- Seifert, T. et al. Efficient metallic spintronic emitters of ultrabroadband terahertz radiation. *Nat. Photon.* **10**, 483 (2016).
- Kampfrath, T. et al. Terahertz spin current pulses controlled by magnetic heterostructures. *Nat. Nanotech.* **8**, 256 (2013).
- Jin, Z. et al. Cascaded Amplification and Manipulation of Terahertz Emission by Flexible Spintronic Heterostructures. *Laser Photonics Rev.* **16**, 2100688 (2022).
- Jungfleisch, M. B. et al. Control of Terahertz emission by ultrafast spin-charge current conversion at Rashba Interfaces. *Phys. Rev. Lett.* **120**, 207207 (2018).
- Zhou, C. et al. Broadband Terahertz generation via the interface inverse Rashba-Edelstein Effect. *Phys. Rev. Lett.* **121**, 086801 (2018).
- Shen, J. et al. Spin-to-Charge Conversion in Ag/Bi Bilayer Revisited. *Phys. Rev. Lett.* **126**, 197201 (2021).
- Choi, Y.-G. et al. Observation of the orbital Hall effect in a light metal Ti. *Nature*. **619**, 52 (2023).
- Hayashi, H., Go, D., Haku, S., Mokrousov, Y. & Ando, K. Observation of orbital pumping. *Nat Electron* **7**, 646–652 (2024).
- Huang, Q. et al. Current-induced magnetization switching in light-metal-oxide/ferromagnetic-metal bilayers via orbital Rashba effect. *Nano. Lett.* **23**, 11323 (2023).
- Zhang, S. & Yang, Z. Intrinsic spin and orbital angular momentum Hall Effect. *Phys. Rev. Lett.* **94**, 066602 (2005).
- Kontani, H. et al. Giant Orbital Hall effect in transition metals: origin of large spin and anomalous hall effects. *Phys. Rev. Lett.* **102**, 016601 (2009).
- Salemi, L. et al. First-principles theory of intrinsic spin and orbital Hall and Nernst effects in metallic monoatomic crystals. *Phys. Rev. Materials*. **6**, 095001 (2022).
- Lee, D. et al. Orbital torque in magnetic bilayers. *Nat. Commun.* **12**, 6710 (2021).
- Jo, D., Go, D. & Lee, H.-W. Gigantic intrinsic orbital Hall effects in weakly spin-orbit coupled metals. *Phys. Rev. B*. **98**, 214405 (2018).
- Lyalin, I. et al. Magneto-Optical Detection of the Orbital Hall Effect in Chromium. *Phys. Rev. Lett.* **131**, 156702 (2023).
- Zheng, Z. C. et al. Magnetization switching driven by current-induced torque from weakly spin-orbit coupled Zr. *Phys. Rev. Res.* **2**, 013127 (2020).
- Kim, S. et al. Optical detection of spin-orbit torque in Ta/CoFeB and W/CoFeB bilayers. *Curr. Applied. Phys.* **61**, 1567–1739 (2024).
- Bass, J. et al. Spin-diffusion lengths in metals and alloys, and spin-flipping at metal/metal interfaces: an experimentalist's critical review. *J. Phys. Condens. Matter*. **19**, 183201 (2007).
- Liu, Y. et al. Interface Enhancement of Gilbert Damping from First Principles. *Phys. Rev. Lett.* **113**, 207202 (2014).
- Chen, K. & Zhang, S. Spin Pumping in the Presence of Spin-Orbit Coupling. *Phys. Rev. Lett.* **114**, 126602 (2015).
- Lee, S. et al. Efficient conversion of orbital Hall current to spin current for spin-orbit torque switching. *Commun. Phys.* **4**, 234 (2021).
- Liu, F., Liang, B., Xu, J., Jia, C. & Jiang, C. Giant efficiency of long-range orbital torque in Co/Nb bilayers. *Phys. Rev. B*. **107**, 054404 (2023).
- Fukunaga, R., Haku, S., Hayashi, H. & Ando, K. Orbital torque originating from orbital Hall effect in Zr. *Phys. Rev. Res.* **5**, 023054 (2023).
- Seifert, T. S. et al. Time-domain observation of ballistic orbital-angular-momentum currents with giant relaxation length in tungsten. *Nat. Nanotechnol.* **18**, 1132 (2023).
- Liu, Y. et al. Efficient Orbitronic Terahertz Emission Based on CoPt Alloy. *Adv. Mater.* **36**, 2404174 (2024).
- Go, D. & Lee, H.-W. Orbital torque: Torque generation by orbital current injection. *Phys. Rev. Res.* **2**, 013177 (2020).
- Han, J. et al. Room-Temperature Spin-Orbit Torque Switching Induced by a Topological Insulator. *Phys. Rev. Lett.* **119**, 077702 (2017).
- Bose, A. et al. Detection of long-range orbital-Hall torques. *Phys. Rev. B*. **107**, 134423 (2023).
- Kim, K.-W. et al. Spin-orbit torques from interfacial spin-orbit coupling for various interfaces. *Phys. Rev. B*. **96**, 104438 (2017).
- Haney, P. M. & Stiles, M. D. Current-Induced Torques in the Presence of Spin-Orbit Coupling. *Phys. Rev. Lett.* **105**, 126602 (2010).
- Go, D. et al. Theory of current-induced angular momentum transfer dynamics in spin-orbit coupled systems. *Phys. Rev. Res.* **2**, 033401 (2020).
- Mishra, S. S., Lourembam, J., Lin, D. J. X. & Singh, R. Active ballistic orbital transport in Ni/Pt heterostructure. *Nat. Commun.* **15**, 4568 (2024).
- Zhang, W. et al. Ultrafast terahertz magnetometry. *Nat. Commun.* **11**, 4247 (2020).
- Huisman, T. J. et al. Femtosecond control of electric currents in metallic ferromagnetic heterostructures. *Nat. Nanotech.* **11**, 455 (2016).

51. Agarwal, P. et al. Ultrafast Photo-Thermal Switching of Terahertz Spin Currents. *Adv. Funct. Mater.* **31**, 2010453 (2021).
 52. Brataas, A. Quantum scattering theory of spin transfer torque, spin pumping, and fluctuations. *Phys. Rev. B*. **106**, 064402 (2022).
 53. Han, S. et al. Orbital pumping incorporating both orbital angular momentum and position. *Phys. Rev. Lett.* **134**, 036305 (2025).
 54. Tserkovnyak, Y., Brataas, A. & Bauer, G. E. W. Enhanced Gilbert Damping in Thin Ferromagnetic Films. *Phys. Rev. Lett.* **88**, 117601 (2002).
 55. Xu, Y. et al. Orbitronics: light-induced orbital currents in Ni studied by terahertz emission experiments. *Nat. Commun.* **15**, 2043 (2024).
 56. Kumar, S. & Kumar, S. Ultrafast THz probing of nonlocal orbital current in transverse multilayer metallic heterostructures. *Nat. Commun.* **14**, 8185 (2023).
 57. Wang, P. et al. Inverse orbital Hall effect and orbitronic terahertz emission observed in the materials with weak spin-orbit coupling. *Npj. Quantum. Mater.* **8**, 28 (2023).
 58. Sala, G. et al. Giant orbital Hall effect and orbital-to-spin conversion in 3d, 5d, and 4f metallic heterostructures. *Phys. Rev. Res.* **4**, 033037 (2022).
 59. Martini, M. et al. Engineering the Spin-Orbit-Torque Efficiency and Magnetic Properties of Tb/Co Ferromagnetic Multilayers by Stacking Order. *Phys. Rev. Appl.* **17**, 044056 (2022).
 60. Yang, Y. et al. Orbital torque switching in perpendicularly magnetized materials. *Nat Commun* **15**, 8645 (2024).
 61. Lan, Z. et al. Unveiling of Terahertz emission from ultrafast demagnetization and the anomalous Hall effect in a single ferromagnetic film. *Chin. Phys. Lett.* **41**, 044203 (2024).
 62. Zhang, Q. et al. Terahertz emission from anomalous Hall effect in a single-layer ferromagnet. *Phys. Rev. Appl.* **12**, 054027 (2019).
 63. Feng, Z. et al. Anomalous Nernst effect induced terahertz emission in a single ferromagnetic film. *Nano. Lett.* **17**, 8171 (2023).
 64. Chen, Z. et al. Light-induced ultrafast spin transport in multilayer metallic films originates from sp-d spin exchange coupling. *Sci. Adv.* **9**, eadi1618 (2023).
 65. Jin, Z. et al. Ultrafast electron transport in metallic antiferromagnetic Mn₂Au thin films probed by terahertz spectroscopy. *Phys. Rev. B*. **102**, 014438 (2020).
 66. Winkel, T. J. et al. Comparative analysis of THz signal emission from bilayer heterostructures: Wideband and high-frequency THz signal advantage of PtBi-based emitter. *Appl. Phys. Lett.* **124**, 012408 (2024).
 67. Jechumtál, J. et al. Accessing ultrafast spin-transport dynamics in copper using broadband terahertz spectroscopy. *Phys. Rev. Lett.* **132**, 226703 (2024).
 68. Lyu, J. et al. Frequency selective fingerprint sensor: the Terahertz unity platform for broadband chiral enantiomers multiplexed signals and narrowband molecular AIT enhancement. *Photonix* **4**, 28 (2023).
 69. Xie, J. et al. Integrated terahertz vortex beam emitter for rotating target detection. *Adv. Photon.* **5**, 066002 (2023).
 70. Liu, B. et al. Ultra-wideband terahertz fingerprint enhancement sensing and inversion model supported by single-pixel reconfigurable graphene metasurface. *Photonix*. **5**, 10 (2024).
 71. Shahriar, B. Y., Hopmann, E. & Elezzabi, A. Y. On-Chip waveguided spintronic sources of terahertz radiation. *ACS. Photonics*. **10**, 518 (2023).
 72. Barnes, M. E. et al. Investigation of the role of the lateral photo-Dember effect in the generation of terahertz radiation using a metallic mask on a semiconductor. *Opt. Express*. **21**, 16263 (2013).
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Author contributions

D.L.Z., Z.M.J., and Y.J. conceived the idea. H.R.X. built the terahertz emission setup and carried out the THz emission measurements, supervised by Z.M.J., Y.M.Z., Y.P. P.W., D.L.Z., Z.F., and S.L.Z. gave advice on the experiments. Y.H.Y., T.W., W.S.Y., C.C., and F.C. fabricated and characterized the samples and optimized the fabrication process, supervised by D.L.Z. and Y.J. Z.J., Y.M.Z., and Y.P. derived the analytical model. Z.M.J., H.R.X., D.L.Z., Y.M.Z., and Y.P. cowrote the manuscript with the contributions from all co-authors. All co-authors discussed the results and commented on the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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