

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

Corresponding Author: Professor Zuanming Jin

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

This manuscript has been previously reviewed at another journal. This document only contains information relating to versions considered at Communications Physics.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors study THz emission from heterostructures containing magnetically ordered and disordered materials. Specifically, they laser-trigger fs electronic angular momentum currents from a Co layer into nearby layers that are Zr and W. The authors argue that due to the low spin orbit coupling of Zr, only the orbital angular momentum can be converted into a measurable in-plane charge inside the Zr layer. On the other hand, insertion of a W layer, i.e. Co|W|Zr, enhances the detected THz signal amplitude, which the author attribute to a spin-to-orbital-angular-momentum conversion inside W. The important role of orbital angular momentum currents in their experiment is further supported by THz waveform delays observed vs the magnetically-disordered film thickness.

In fact, numerous studies have shown similar effects observed in the current manuscript and I wonder what further insights it actually provides. The actual novelty is THz emission involving a Zr layer. However, in light of recent DC-type experiments [Fukunaga, Riko, et al. "Orbital torque originating from orbital Hall effect in Zr." *Physical Review Research* 5.2 (2023): 023054.] and the general trend that whatever can be measured in those DC-type experiments can also be observed in THz emission studies, this does not come as a surprise.

In fact, the only true claimed novelty remains to be the enhanced signal amplitudes when inserting W. But I would argue that this part of the paper is flawed in that it draws no fair comparison between Co|W and Co|Zr. Specifically, it does not show data for Co|W with a reasonable W thickness, i.e., of around 3 nm. For much thinner W films, angular momentum currents will be reflected off the back surface and thereby suppress the measured signal amplitude.

Moreover, many parts of the paper seem to be inspired to a large extent by existing publication, if not to say they are in parts plagiarized. Just to name a few examples: Fig. 1 a-c closely resembles figures in Ref 23. Large parts of the first part of the manuscript closely resemble Ref. 40. The last part of the manuscript including the analysis vs pump fluence largely resembles Ref. 46. None of these three references is properly cited, in the sense that already from their order of appearance it seems clear that the authors do not wish to draw too much attention to those sources.

For these reasons, I cannot recommend publication.

Reviewer #2

(Remarks to the Author)

In the submitted paper the authors perform THz emission spectroscopy experiments and study the orbital-to-charge current conversion in Co/Zr/Al₂O₃ and Co/W/Zr/Al₂O₃ heterostructures. A long-distance ballistic flow of orbital current is observed at the Zr layers, and the strong emitted THz signal is further enhanced by the insertion of the W layer. A critical pump fluence value is identified that enables a faster flow of orbital current. In order to draw conclusions, the authors perform additional experiments to explore the dependence on the thickness of the Zr layers, on the pump fluence intensity, and on the temperature.

The paper is well written, and the presented work is solid and well justified. The study follows previous experimental works (Refs. [40, 41, 46-49]), the results are in accord with them and a new material platform is proposed for the generation of light-induced orbital currents. I believe that the work will benefit the community given the rising interest in opto-orbital effects.

I can suggest publication if the following points are addressed by the authors:

- 1) Why was Co the choice for the FM layer and not Ni since Ni has higher spin-orbit correlation?
- 2) Is the reason behind the choice of Zr only the dominating OHC over the SHC in this material?
- 3) How does the calculated orbital velocity of 0.27 nm/fs compare to previous experiments? The readers might benefit from this additional information.
- 4) What is the orbital diffusion length and velocity for the Co/W/Zr/Al₂O₃ heterostructure?
- 5) Can the authors comment on the influence of an increase to the thickness of the W inserted layers?
- 6) The quality of the figures in the Supplementary is very poor. It was very difficult to read the plots and understand since there is no information in the axis labels and the legends. This was contradictory to the good quality of the main text.

Reviewer #3

(Remarks to the Author)

Xu et al. report on orbital transport in Co/Zr/Al₂O₃ and Co/W/Zr/Al₂O₃ heterostructures, probed through ultrafast demagnetization using THz optical pulses. The core concept is the generation of orbital and spin currents from the ultrafast demagnetization of the ferromagnet via spin and orbital pumping, which are subsequently converted into inverse spin Hall and orbital Hall effects, producing a transverse charge current. The authors attempt to disentangle multiple simultaneous processes, which, while intriguing, are challenging to detect unambiguously. I can recommend this work for publication, provided the following queries are addressed.

- 1) A comparison of ferromagnets with different spin-orbit coupling strengths (η) is essential to fully understand the entangled mechanisms. It would be important to compare the results for Fe or Fe-rich FeCoB with Co or Ni films, where the generated orbital current would flip polarity, providing direct evidence.
- 2) The authors perform sanity checks by comparing Co(2 nm)/Al₂O₃ with Co/Zr(3 nm)/Al₂O₃ in Fig. 1(e). However, this comparison could be problematic, as the 2 nm Co layer may partially oxidize when in contact with Al₂O₃. I suggest performing additional control experiments using symmetric stacks, such as Al₂O₃/Zr/Co/Zr/Al₂O₃, to mitigate this issue.
- 3) It is surprising that the orbital Hall effect (OHE), or inverse OHE, is not observed in the Co/Zr(t) layers, especially when Zr is predicted to exhibit a large OHE. The absence of this effect requires further explanation.
- 4) Instead of plotting the THz amplitude in arbitrary unit, the authors should present normalized values in the main paper, with the raw data included in the supplementary material (SM).
- 5) In Figure 2, when analyzing the normalized THz amplitude, did the authors account for current shunting in their analysis? As the Zr thickness increases, the generated transverse charge current or voltage would naturally be reduced. This was the rationale behind attributing the effect to the inverse orbital Rashba-Edelstein effect (IOREE) rather than the inverse orbital Hall effect (IOHE).
- 6) The explanation for the decrease in time delay in Fig. 2b for Zr thicknesses greater than 15 nm is unclear and unconvincing. The authors should provide additional data points for Zr thicknesses above 15 nm to support their claim.
- 7) The results on Co/W/Zr/Al₂O₃ are ambiguous, and the coexisting mechanisms are not convincingly disentangled. Introducing a symmetric stack or varying the W thickness, as mentioned earlier, could help clarify these mechanisms.

Overall, while the data presented for various samples is valuable, the analysis remains ambiguous, and the claims are not sufficiently robust. The authors should focus on one aspect of the study and strengthen their claims with more controlled experiments, which are currently lacking. If these queries are addressed and the manuscript is accordingly revised, I believe it would be suitable for publication in this journal.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

I would like to thank the authors for responding to the raised concerns. However, I stand by my previous assessment that the current work does not advance the field to such an extent that it deserves to be published in Communication Physics. I am still convinced that the interpretation stands on shaky grounds regarding fluence thresholds to overcome collisions or critical temperatures to ease orbital current flow. None of these concepts are based on solid theoretical grounds.

Reviewer #2

(Remarks to the Author)

The authors addressed all my comments, therefore I recommend publication of the revised manuscript.

Reviewer #3

(Remarks to the Author)

The response by Xu et al. has been partially satisfactory, as some of the questions, such as W thickness variation, have been addressed effectively, while others could benefit from further clarification. The additional experimental data involving W is commendable and adds value to the work.

The comparison between Co and Ni was not intended since Ni is not ideal for ultrafast measurements. Rather I suggested Fe or Fe-rich FeCoB alloys as more suitable alternatives for comparison to verify that Fe does not produce such signals. The explanation for the notably small OHE or IOHE of Zr remains unclear, and a deeper qualitative understanding would be appreciated and it could be pursued in future work.

Having said this, I acknowledge that the quality of the work has significantly improved, and I recommend it for publication. To further enrich the manuscript, the authors could consider discussing emerging mechanisms for generating orbital currents, such as orbital pumping and spin fluctuations, and incorporating relevant references into the discussion.

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Dear Professor Jin, Your manuscript entitled "Manipulation of light-induced orbital transport in Co/W/Zr/Al₂O₃ heterostructure with ultrafast terahertz emission probing" has now been seen by 3 referees, whose comments are appended below. You will see from their comments that while they find your work of potential interest, they have raised quite substantial concerns that must be addressed. Especially, authors should pay attention to the critics raised by reviewer 1. The novelty of this study should be properly justified. In light of these comments, we believe that your study will require substantial revision before being reconsidered in this journal. We would still be interested in considering a revised version that addresses these serious concerns, if the authors feel this is possible. We hope you will find the referees' comments useful as you decide how to proceed. Should further experimental data or analysis allow you to address these criticisms, we would be happy to look at a substantially revised manuscript.

<Response: We thank all the reviewers for providing insightful comment and critical assessment to our manuscript. We have now taken care of all the issues pointed out by the referees in his/her specific comments as below. This has indeed helped us to improve the presentation of our manuscript. To validate our conclusions, we have performed additional experiments, which are now incorporated in our revised manuscript.

The comments from the reviewers are reproduced verbatim as below (black color). We answer all the comments in our best possible manner (blue color). The revisions in the main manuscript and the supplementary information are highlighted in yellow color. >

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The authors study THz emission from heterostructures containing magnetically ordered and disordered materials. Specifically, they laser-trigger fs electronic angular momentum currents from a Co layer into nearby layers that are Zr and W. The authors argue that due to the low spin orbit coupling of Zr, only the orbital angular momentum can be converted into a measurable in-plane charge inside the Zr layer. On the other hand, insertion of a W layer, i.e. Co|W|Zr, enhances the detected THz signal amplitude, which the author attribute to a spin-to-orbital-angular-momentum conversion inside W. The important role of orbital angular momentum currents in their experiment is further supported by THz waveform delays observed vs the magnetically-disordered film

thickness. In fact, numerous studies have shown similar effects observed in the current manuscript and I wonder what further insights it actually provides. The actual novelty is THz emission involving a Zr layer. However, in light of recent DC-type experiments [Fukunaga, Riko, et al. "Orbital torque originating from orbital Hall effect in Zr." *Physical Review Research* 5.2 (2023): 023054.] and the general trend that whatever can be measured in those DC-type experiments can also be observed in THz emission studies, this does not come as a surprise.

<Response: We sincerely thank the reviewer for raising an important point regarding the novelty and insights of our manuscript. We agree that Zr is relatively new and less explored material in that regard, although Zr exhibits large orbital conductivity. Several authors in this paper used the orbital Hall effect of the Zr with weak spin-orbit coupling to generate the orbital torque, realizing the efficient reversal of the magnetic moment of materials with perpendicular magnetic anisotropy. The critical switching current density can be as low as 2.6×10^6 A/cm², which is lower than that of heavy metal heterojunctions (*Nat Commun* **15**, 8645 (2024)).

As mentioned by the Reviewer, the study of Fukunaga et al. highlighted the orbital torque driven by the orbital Hall effect in Zr through DC measurements [*Physical Review Research* 5 (2023): 023054]. In this paper, only the orbital diffusion length of 14.2 ± 0.8 nm can be obtained, however, the orbit orbital velocity cannot be accessed. In addition, the inverse orbital Hall effect of Zr and its modulation on the ultrafast time scales are still unclear, which is the problem to be solved in this paper. Here, for the first time, we demonstrate that light metal Zr as a promising nonmagnetic layer in the heterostructures enables coherent terahertz radiation driven by *LCC*. The THz emission from Co/Zr and Co/W/Zr heterostructure are observed via long-range ballistic orbital transport, as evidenced by the delay and chirping in the emitted THz pulse correlating with increased Zr thickness. Both the orbital velocity and orbital diffusion length can be accessed from the THz emission spectroscopy. Furthermore, 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. Our work broadens the scope of research on spintronic terahertz radiation sources.

Following the comments, we change the title of our manuscript. "Generation and manipulation of light-induced orbital transport in Co/Zr/Al₂O₃ heterostructure probed with ultrafast terahertz

emission” in Page 1. >

In fact, the only true claimed novelty remains to be the enhanced signal amplitudes when inserting W. But I would argue that this part of the paper is flawed in that it draws no fair comparison between Co|W and Co|Zr. Specifically, it does not show data for Co|W with a reasonable W thickness, i.e., of around 3 nm. For much thinner W films, angular momentum currents will be reflected off the back surface and thereby suppress the measured signal amplitude.

<Response: We are grateful to the Reviewer for this comment. We appreciate your concern regarding the role of the W interlayer in the enhancement of THz emission from Co/Zr heterostructures. We agree that a comparison between Co/W, Co/Zr, and Co/W/Zr would be helpful in clarifying the role of the insertion layer.

Firstly, new samples of Co(2)/W(3)/Zr(3), Co(2)/W(5)/Zr(3), Co(2)/W(3), Co(2)/W(5) have been fabricated and characterized. Next, we make a comparison between Co/W, Co/Zr bilayers, and Co/W/Zr trilayer, as shown in the following Fig. 4 (a)-(c). Based on the experimental observations, the following points are made.

Observation (1): The THz signal emitted from Co(2)/W(1) is 3.65 times higher than that from Co(2)/Zr(3). The THz signal emitted from Co(2)/W(3) is 9.3 times higher than that from Co(2)/Zr(3). The THz signal emitted from Co(2)/W(5) is 4.04 times higher than that from Co(2)/Zr(3). The polarities of THz emission from Co/W heterostructures are opposite to that from Co/Zr.

Conclusion (1): As the spin conductivity and orbital conductivity of W layer have the opposite sign, thus, the SCC and LCC induced currents should have opposite polarities. However, the spin-orbit correlation factor of W, $\eta_{L-S}^W < 0$. Since the sign of $\theta_{SCC}^W < 0$ and $\theta_{LCC}^W > 0$, thus the mechanisms of SCC and LCC add up constructively, as shown in Fig. 4 (e), because they coincide with the same signs of $\theta_{SCC}^W < 0$ and $\eta_{L-S}^W \theta_{LCC}^W < 0$. Note 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 polarity of the Co(2)/Zr(3) and Co(2)/W should be the same. However, they are different, indicating that the LCC dominates in the Co(2)/Zr(3)/Al₂O₃ heterostructures.

Observation (2): For the case of Co/W(d_w) bilayer, THz signal strength firstly increases with increasing the W-layer from 1 nm to 3 nm. While, the THz signal strength decreases when increasing the W layer from 3 nm to 5 nm.

Conclusion (2): This clearly indicates that the spin diffusion length in the W layer is around 2-3 nm, which agrees well with the literature [Physical Review Materials, 2, 014403 (2018)]. It can be expected that for a thicker W-insertion layer ($W > 3$ nm) would have significantly reduced the spin current transmitted to the adjacent layer.

Observation (3): Co(2)/W(1)/Zr(3) sample emits nearly 1.7 and 6.24 times higher THz signal as compared to that from Co(2)/W(1) and Co(2)/Zr(3), respectively. However, the THz emission from Co(2)/W(3)/Zr(3) is nearly 60.7% of that from Co(2)/W(3). The THz emission from Co(2)/W(5)/Zr(3) is nearly 45.6% of that from Co(2)/W(5).

Conclusion (3): As the $\eta_{L-S}^W < 0$ is opposite with the $\eta_{L-S}^{Co} > 0$, the THz emission from Co(2)/W(1)/Zr(3)/Al₂O₃ shows opposite polarity of Co(2)/Zr(3)/Al₂O₃. If the emission in the Co(2)/W(1)/Zr(3) structure was only due to the ISHE, one would expect that the THz signal amplitude to be nearly equal to the sum of the THz signals emitted from Co(2)/W(1) and Co(2)/Zr(3). In contrast, the measured THz signal from the Co/W(1)/Zr(3) to be nearly 1.7 times stronger than the Co(2)/W(1) and 6.24 times stronger than the Co(2)/Zr(3). It can also be found that $E_{Co(2)/W(1)/Zr(3)/Al_2O_3}^{THz} - E_{Co(2)/W(1)/Al_2O_3}^{THz}$ and $E_{Co(2)/Zr(3)/Al_2O_3}^{THz}$ also show a 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 $E_{Co(2)/Zr(3)/Al_2O_3}^{THz}$.

Therefore, the role of the 1 nm W-insertion layer acts as an additional orbital transport channel rather than simply as SCC and LCC effects itself. The physical picture is that the spin current from Co can be converted to the orbital current in W and then transmitted into the adjacent Zr layer within the Co/W/Zr/Al₂O₃ heterostructure. And then, the additional orbital current flows into charge current, which explains the magnitude enhancement of THz emission from the trilayer. While, the presence of a thicker W insertion layer ($W \geq 3$ nm) would have significantly suppressed the spin current, which consequently reduces the spin-to-orbit conversion in W and finally decreases the THz emission. We believe that the W-thickness dependent THz emission from Co/W/Zr/Al₂O₃ will be a valuable knowledge to this research field. Specifically, an optimum 1 nm thickness of the inserting W layer provides an additional large orbital current by its large spin-orbit coupling.

We have updated the main text and Figure 4 with corresponding discussion. >

Moreover, many parts of the paper seem to be inspired to a large extent by existing publication, if not to say they are in parts plagiarized. Just to name a few examples: Fig. 1a-c closely resembles figures in Ref 23.

<Response: We thank the Reviewer for this important comment concerning the novelty of our findings. Several recent works have highlighted the importance of the orbital degree of freedom in condensed matter physics and kicked off the emergent research field of orbitronics. Indeed, our study follows previous experimental works, our experimental results are in accord with them and a new material platform is proposed for the generation and manipulation of light-induced orbital currents in this study. Although the exploration of orbital transport is just beginning and we believe that new materials and structures of spintronic terahertz emitters are certainly of high interest for the broader scientific community. As shown in previous work, inserting a W layer with high spin-orbit conversion efficiency has been shown to enhance the THz emission (npj Quantum Materials 28 (2023)). However, the influence of thickness dependency of the W inserted layers on THz emission still need to be explored. In addition, further experimental studies are required to confirm the role of effectively active control in generation ultrafast orbital current for THz generation.

We apologize that in the first version of the paper, Fig. 1 (a)-(c) closely resembles figures in Ref 23. We revised the Figure 1 (a)-(c), as shown in the following Figure 1.

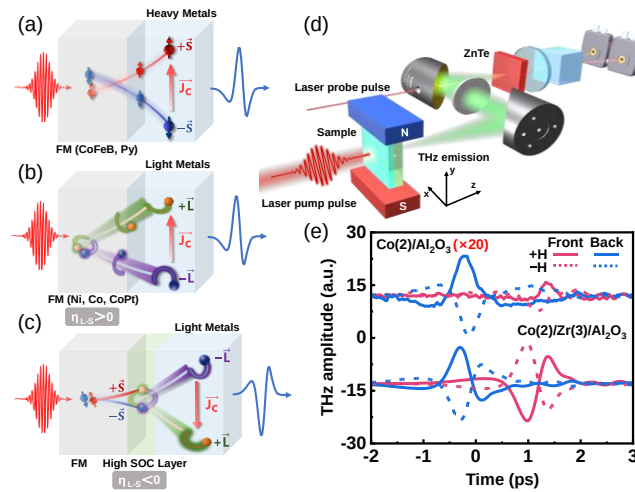


Figure 1

Large parts of the first part of the manuscript closely resemble Ref. 40. The last part of the manuscript including the analysis vs pump fluence largely resembles Ref. 46. None of these three references is properly cited, in the sense that already from their order of appearance it seems clear

that the authors do not wish to draw too much attention to those sources. For these reasons, I cannot recommend publication.

<Response: We will do our best to convince the Reviewer of originality and accuracy of our results in our response to comments below.

1. Additional experiments are performed on the Co/Zr bilayer with varying thickness of Zr layer to reiterate our original findings. To better quantitatively understand the concave shape of τ_D as a function of d_{Zr} , we used the formalism presented in [52], by introducing a probability of orbital flip or orbital decoherence, to express the τ_D . From the fit of Eq (2), the orbital velocity and orbital diffusion length of Zr are extracted. In addition, 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). This particular observation will be a new value addition to the existing literature.

All the literatures are cited properly and we have updated the main text and Figure 2 with corresponding discussion.

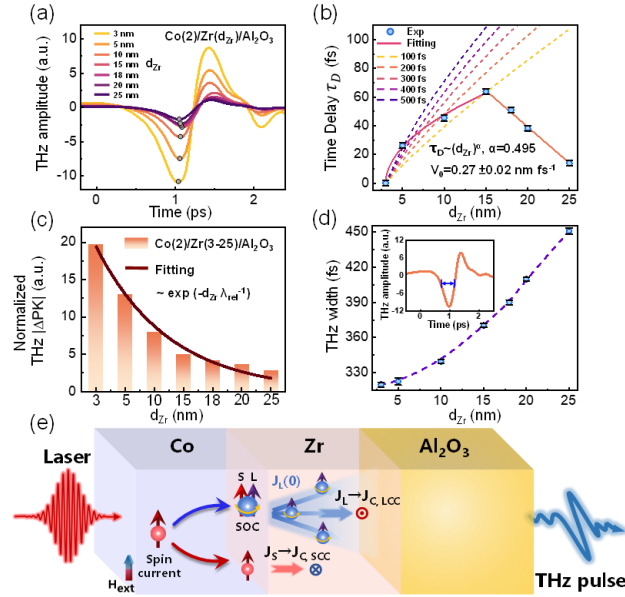


Figure 2

In addition, we have compared the orbital velocity and orbital diffusion length with that reported values in other heterostructures, such as Ni/W, Ni/Cu, Ni/Pt, and CoPt/Cu, as summarized in the Table 2.

Heterostructures	Orbital velocity	Orbital diffusion
	(nm fs ⁻¹)	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 [52]	9.0 [52]
Al ₂ O ₃ (substrate)/Zr/Co/MgO(capping layer)	0.27±0.02	7.0~15.6

2. We have provided new experimental results on Co/W and Co/W/Zr having different thicknesses of the W layer to further strengthen our claim on the orbital current modulation in the THz signal from the Co/W/Zr heterostructure. Undoubtedly, the Co/Zr and Co/W/Zr systems are studied in our manuscript is a suitable for exploring orbital transport phenomena.

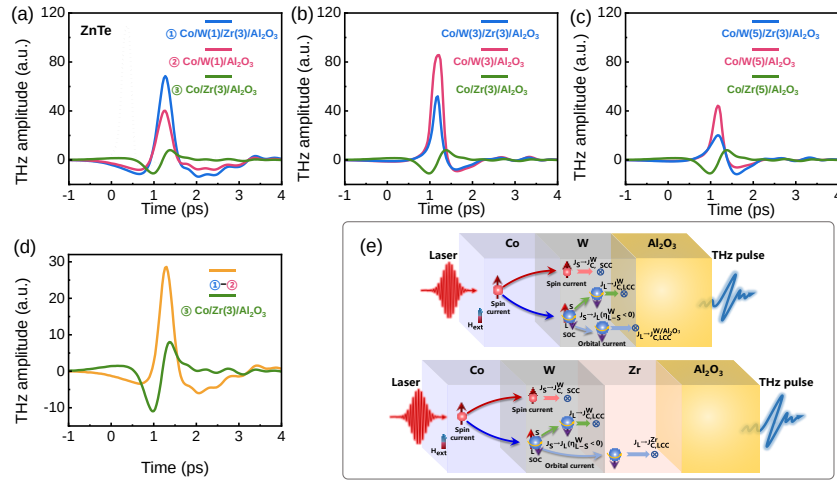


Figure 4

We have updated the main text and Figure 4 with corresponding discussion.

3. Furthermore, we performed temperature-dependent THz emission studies, which creates unique challenges and opportunities that were not addressed in earlier works.

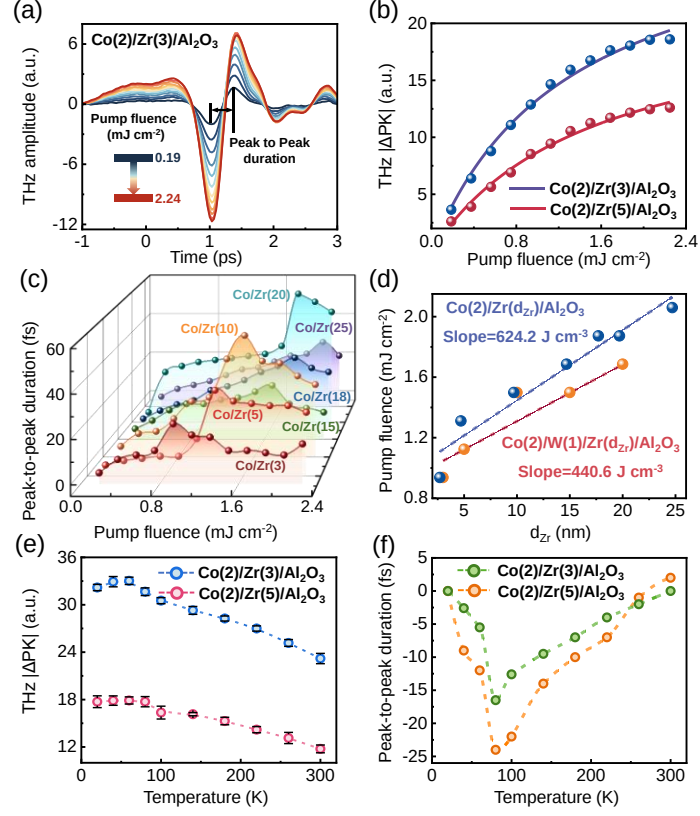


Figure 3

We have updated the main text and Figure 3 with corresponding discussion.

Summary, 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 . 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. 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.

We hope that additional experimental results and the corresponding rewritten alleviate the concern of the Reviewer about the novelty of this study. We believe that the differences in the material systems and manipulation approach used in our work contribute to the advancement of such an emerging field. >

Reviewer #2 (Remarks to the Author):

In the submitted paper the authors perform THz emission spectroscopy experiments and study the orbital-to-charge current conversion in Co/Zr/Al₂O₃ and Co/W/Zr/Al₂O₃ heterostructures. A long-distance ballistic flow of orbital current is observed at the Zr layers, and the strong emitted THz signal is further enhanced by the insertion of the W layer. A critical pump fluence value is identified that enables a faster flow of orbital current. In order to draw conclusions, the authors perform additional experiments to explore the dependence on the thickness of the Zr layers, on the pump fluence intensity, and on the temperature.

The paper is well written, and the presented work is solid and well justified. The study follows previous experimental works (Refs. [40, 41, 46-49]), the results are in accord with them and a new material platform is proposed for the generation of light-induced orbital currents. I believe that the work will benefit the community given the rising interest in opto-orbital effects.

<Response: We are very grateful to the Reviewer for his/her positive evolution of our work, and for supporting its publication in *Communications Physics*. In particular, your recognition of the new material platform we proposed for generating photoinduced orbital currents has been a great encouragement to our research group.>

I can suggest publication if the following points are addressed by the authors:

1) Why was Co the choice for the FM layer and not Ni since Ni has higher spin-orbit correlation?

<Response: We certainly agree with the Reviewer that Ni has higher spin-orbit correlation. At present, most of the orbital currents generated by femtosecond lasers have been observed in Ni-based systems, such as Nature nanotechnology 18 1132, (2023) and Nature Communications 15 4568 (2024). Actually, Co has a significantly higher saturation magnetization (M_s) than Ni. The M_s per atom for Co and Ni is approximately $1.6 \mu_B$ and $0.6 \mu_B$, respectively [*Phys. Rev. B* 77, 155435 (2008)]. In addition, the Curie temperature of Ni is relatively low, limiting the potential for

developing high performance spintronic THz emitters.

In literature [*Phys Rev Research* **4**, 033037 (2022)], the effective spin-orbital conductivity ξ_{LS} were measured in FM/NM layers. Note that the ξ_{LS} of Co/Cr is larger than that of Ni/Cr with flip polarity, the ξ_{LS} of Co/Mn is larger than that of Ni/Mn with flip polarity, while the ξ_{LS} of Co/Pt is large than Ni/Pt with the same sign. It is important to see that ξ_{LS} depends on the materials, combinations and interfaces, etc.

As wish by the Reviewer, we fabricate a Ni/Zr bilayer. As shown in the Figure R1, we find that the THz emission from Co/Zr is much larger than that of Ni/Zr. Therefore, our experimental results indicate that Co(2) and Co(2)/W(1) are a good material choice for orbital current source. In this paper, we provide strong evidence for conversion from a light-induced both spin current and orbital current within Co into charge currents for generation of free-space THz pulse radiation.

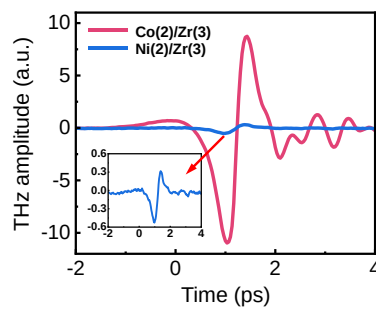


Figure R1 Comparison between the emitted THz waveforms obtained from Co(2)/Zr(3) and Ni (2)/Zr(3) bilayers>

2) Is the reason behind the choice of Zr only the dominating OHC over the SHC in this material?

<Response: We are grateful to the Reviewer for this comment. In previous work, several authors in this paper investigated the orbital current converted to charge current in the Ti and Mn layers via the inverse orbital Hall effect. Indeed, the light metal Zr layer with large OHC has never been investigated. On the other hand, several authors in this paper used the orbital Hall effect of the Zr with weak spin-orbit coupling to generate the orbital torque, realizing the efficient reversal of the magnetic moment of materials with perpendicular magnetic anisotropy. The critical switching current density can be as low as 2.6×10^6 A/cm², which is lower than that of heavy metal heterojunctions (*Nat Commun* **15**, 8645 (2024)). Actually, both the spin-charge and orbit-charge conversions of Zr on ultrafast time scales are still unexplored, which is the issue to be solved in this

paper.

We have added the following in our revised manuscript.

Zr is a 4d light metal for orbital transport due to its $4d^25s^2$ orbital configuration, the unfilled d cell facilitates the efficient transportation of $\mathbf{L}$ [32]. Although Zr has been used to generate the orbital torque [39], and further realizing the efficient reversal of the magnetic moment with perpendicular magnetic anisotropy [57]. 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 $\mathbf{J}_L$ is not fully understood. >

3) How does the calculated orbital velocity of 0.27 nm/fs compare to previous experiments? The readers might benefit from this additional information.

<Response: We thank the reviewer for the comment. We would like to emphasize that the Zr thickness dependent THz emission spectroscopy helps to provide an estimation of the group velocity of the orbital carriers of the Zr layer within the Co/Zr heterostructure, which yields a value of about $v_o = 0.27 \pm 0.02$ nm/fs. As suggested by the Reviewer, we have compared it with that reported values in other heterostructures, such as Ni/W, Ni/Cu, Ni/Pt, and CoPt/Cu, as summarized in the Table 2.

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

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

Among the reported orbital velocities, the Co/Zr/Al₂O₃ heterostructure in this work exhibits comparable and high orbital velocity, compared with the values reported by Xu and Seifert for Ni/Cu/MgO is 0.26±0.08 nm/fs and Ni/W/SiO₂ is 0.14 nm/fs, respectively.

We have added the Table 2 into the revised manuscript, and following in our revised manuscript.

Although Zr is a relatively new candidate as an LCC material, its large orbital conductivity and orbital velocity makes it a highly promising material for THz emission driven by ultrafast orbital currents. >

4) What is the orbital diffusion length and velocity for the Co/W/Zr/Al₂O₃ heterostructure?

<Response: We thank the reviewer for the comment. Figure R2 shows the THz emission waveforms of Co(2)/W(1)/Zr(d_{Zr})/Al₂O₃ with different Zr thicknesses.

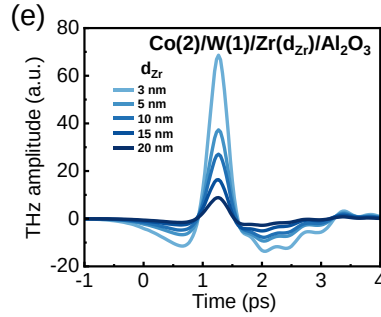


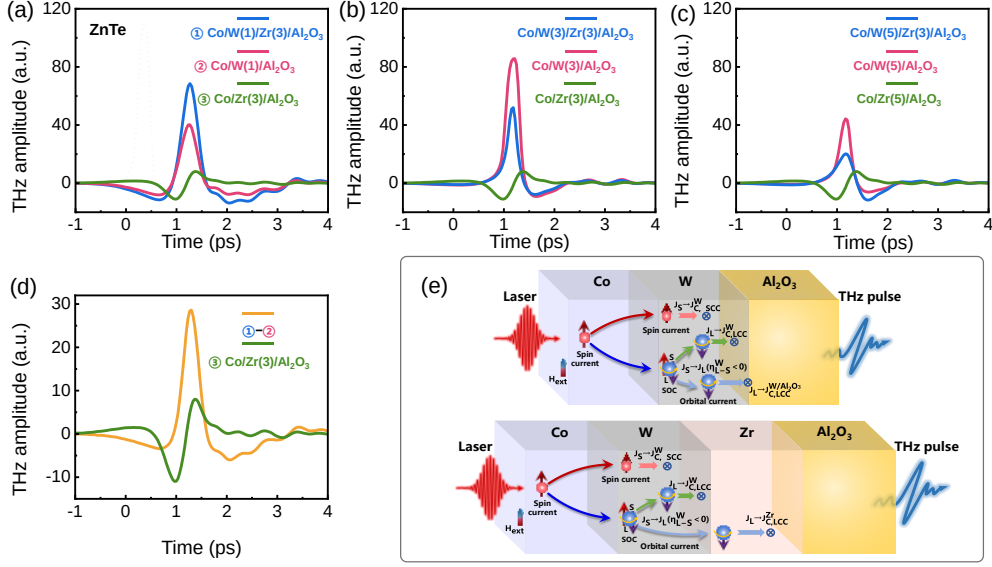
Figure R2

In the Co(2)/W(1)/Zr(d_{Zr})/Al₂O₃ heterostructures, the THz emission not only comes from conversion of orbital current but also from a significant bulk spin-ISHE effects of W within the whole heterostructures. Therefore, the time delay in the THz peak is not visible clearly, compared with the Co(2)/Zr(3-25)/Al₂O₃ heterostructures under the same experimental geometry. As the values of the orbital velocity and orbital diffusion length are derived from the measured time delay derived from Eq. (2). Therefore, we cannot obtain the orbital diffusion length and velocity from the Co(2)/W(1)/Zr(d_{Zr})/Al₂O₃ heterostructures.

We reorganized the paper and this figure is deleted in the revised manuscript. >

5) Can the authors comment on the influence of an increase to the thickness of the W inserted layers?

<Response: The Reviewer indeed raises a very important point, which we address here by performing additional experiments. New samples have been fabricated and characterized. In the revised Fig. 4 (a)-(c) show the normalized THz emission results obtained from Co(2)/W($d_W=1, 3, 5$)/Zr(3)/Al₂O₃ heterostructures, in each figure, are direct compared with Co(2)/W($d_W=1, 3, 5$) and Co(2)/Zr(3) heterostructures.



We made a qualitative analysis of W thickness dependent THz emission from Co(2)/W($d_W=1, 3, 5$)/Al₂O₃ and Co(2)/W($d_W=1, 3, 5$)/Zr(3)/Al₂O₃, by the following observations in Figure 4 (a)-(c):

Observation (1): The THz signal emitted from Co(2)/W(1) is 3.65 times higher than that from Co(2)/Zr(3). The THz signal emitted from Co(2)/W(3) is 9.3 times higher than that from Co(2)/Zr(3). The THz signal emitted from Co(2)/W(5) is 4.04 times higher than that from Co(2)/Zr(3). The polarities of THz emission from Co/W heterostructures are opposite to that from Co/Zr.

Conclusion (1): As the spin conductivity and orbital conductivity of W layer have the opposite sign, thus, the SCC and LCC induced currents should have opposite polarities. However, the spin-orbit correlation factor of W, $\eta_{L-S}^W < 0$. Since the sign of $\theta_{SCC}^W < 0$ and $\theta_{LCC}^W > 0$, thus the mechanisms of SCC and LCC add up constructively, as shown in Fig. 4 (e), because they coincide with the same signs of $\theta_{SCC}^W < 0$ and $\eta_{L-S}^W \theta_{LCC}^W < 0$. Note 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 polarity of the Co(2)/Zr(3) and Co(2)/W should be the same. However, they are different, indicating that the LCC dominates in the Co(2)/Zr(3)/Al₂O₃ heterostructures.

Observation (2): When we increase the thickness of W, the THz amplitude of Co(2)/W(3) is larger than that of Co(2)/W(1). While, the Co(2)/W(5) is smaller than that of Co (2)/W (3) bilayer.

Conclusion (2): This clearly indicates that the spin diffusion length in the W layer is limited. It can be expected that for a thicker W-insertion layer ($W > 3$ nm) would have significantly reduced the spin current transmitted to the adjacent layer.

Observation (3): Co(2)/W(1)/Zr(3) sample emits nearly 1.7 and 6.24 times higher THz signal as compared to that from Co(2)/W(1) and Co(2)/Zr(3), respectively. However, the THz emission from Co(2)/W(3)/Zr(3) is nearly 60.7% of that from Co(2)/W(3). The THz emission from Co(2)/W(5)/Zr(3) is nearly 45.6% of that from Co(2)/W(5).

Conclusion (3): As the $\eta_{L-S}^W < 0$ is opposite with the $\eta_{L-S}^{Co} > 0$, the THz emission from Co(2)/W(1)/Zr(3)/Al₂O₃ shows opposite polarity of Co(2)/Zr(3)/Al₂O₃. It can be found that $E_{Co(2)/W(1)/Zr(3)/Al_2O_3}^{THz} - E_{Co(2)/W(1)/Al_2O_3}^{THz}$ and $E_{Co(2)/Zr(3)/Al_2O_3}^{THz}$ also show 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 $E_{Co(2)/Zr(3)/Al_2O_3}^{THz}$. Therefore, the role of the 1 nm W-insertion layer acts as an additional orbital transport channel rather than simply as SCC and LCC effects itself. The physical picture is that the spin current from Co can be converted to the orbital current in W and then transmitted into the adjacent Zr layer within the Co/W/Zr/Al₂O₃ heterostructure. And then, the addition orbital current flows into charge current, which explains the magnitude enhancement of THz emission. While, the presence of a thicker W insertion layer ($W \geq 3$ nm) would significantly suppress the spin current, which consequently reduces the spin-to-orbit conversion in W and finally decreases the THz emission. We believe that the W-thickness dependent THz emission from Co/W/Zr/Al₂O₃ will be a valuable knowledge to this research field. Specifically, an optimum 1 nm thickness of the inserting W layer provides an additional large orbital current by its large spin-orbit coupling.

We make a corresponding comment into the main text of the revised manuscript. >

6) The quality of the figures in the Supplementary is very poor. It was very difficult to read the plots and understand since there is no information in the axis labels and the legends. This was contradictory to the good quality of the main text.

<Response: We certainly agree with the Reviewer. (1) All axis labels and legends have been updated to include detailed and clear information to enhance readability and comprehension. (2) The resolution of the figures has been improved to ensure they meet the same quality standards as the main text. (3) Additional annotations have been added where necessary to provide further clarification.

We have updated the supplementary material in the revised submission. >

Reviewer #3 (Remarks to the Author):

Xu et al. report on orbital transport in Co/Zr/Al₂O₃ and Co/W/Zr/Al₂O₃ heterostructures, probed through ultrafast demagnetization using THz optical pulses. The core concept is the generation of orbital and spin currents from the ultrafast demagnetization of the ferromagnet via spin and orbital pumping, which are subsequently converted into inverse spin Hall and orbital Hall effects, producing a transverse charge current. The authors attempt to disentangle multiple simultaneous processes, which, while intriguing, are challenging to detect unambiguously. I can recommend this work for publication, provided the following queries are addressed.

<Response: We are very grateful to the Reviewer for his/her overall very positive evaluation of our work, and recommending our work for publication in *Communications Physics*. We hope our additional experimental results and clarifications adequately address your concerns.>

1) A comparison of ferromagnets with different spin-orbit coupling strengths (η) is essential to fully understand the entangled mechanisms. It would be important to compare the results for Fe or Fe-rich FeCoB with Co or Ni films, where the generated orbital current would flip polarity, providing direct evidence.

<Response: We thank the Reviewer for raising this point. As mentioned in literature [*Nat Commun* **12**, 6710 (2021)], The orbital-to-spin conversion efficient C_{FM} of Fe, CoFe, Co and Ni are shown in the following Figure, which indicates that they have the same polarity.

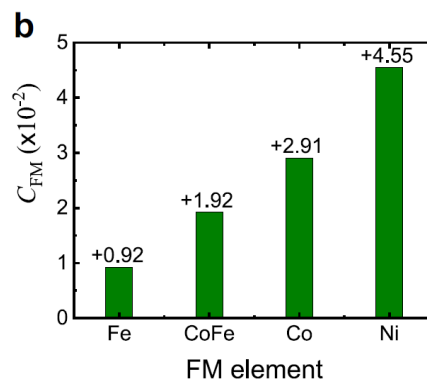


Figure Orbital-to-spin conversion efficiency C_{FM} of Fe, CoFe, Co, and Ni.

[*Nat Commun* **12**, 6710 (2021)]

In addition, in literature of *Phys Rev Research* **4**, 033037 (2022), the effective spin-orbital conductivity ξ_{LS} were measured in several FM/NM layers. The ξ_{LS} of Co/Cr is larger than that of

Ni/Cr with flip polarity, the ξ_{LS} of Co/Mn is larger than that of Ni/Mn with flip polarity, while the ξ_{LS} of Co/Pt is large than Ni/Pt with the same sign. Note that the ξ_{LS} depends on the materials, combinations, interfaces, etc, and hence demands an explicit verification.

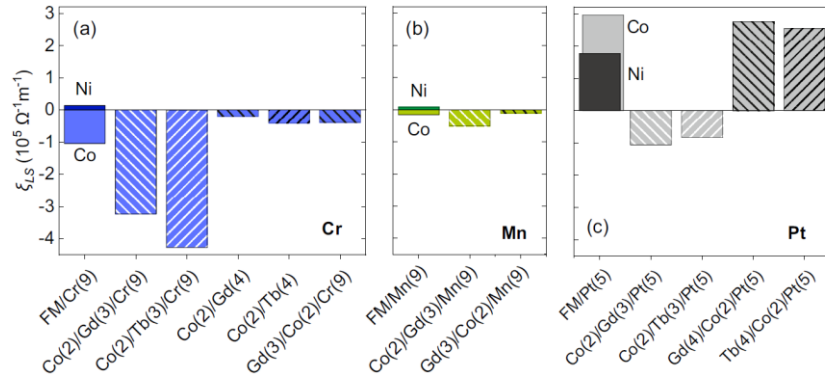


Figure Comparison of the effective spin-orbital conductivity ξ_{LS} measured in NM/FM and NM/X/FM layers where (a) NM = Cr, (b) NM = Mn, and (c) NM = Pt; FM = Co, Ni, X = Gd, Tb.

[Phys Rev Research 4, 033037 (2022)]

In previous work, Xu et al., have demonstrated that extremely small differences between MgO/CoFeB and MgO/Cu/CoFeB, which exclude the existence of significant orbital effect. Therefore, the orbital current generated from CoFeB is considered to be much less pronounced than Ni and Co.

As wish by the Reviewer, as shown in the Figure R1, additional experiments were conducted on the Ni/Zr under identical photoexcited conditions. The results indicate that THz emission from Co/Zr is much stronger than that from Ni/Zr. Notably, the polarities of THz emissions from Co/Zr and Ni/Zr are the same.

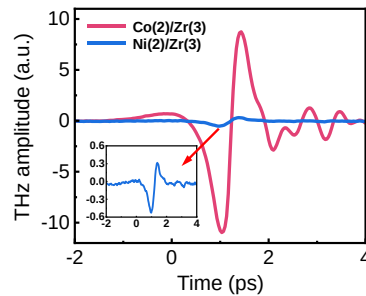


Figure R1 Comparison between the emitted THz waveforms obtained from Co(2)/Zr(3) and Ni(2)/Zr(3) bilayers

In our experiments, we compare the THz emission from Co/W/Al₂O₃ and Co/Zr/Al₂O₃, as the

spin-orbit correlation factor of W, $\eta_{L-S}^W < 0$. Since the sign of $\theta_{SCC}^W < 0$ and $\theta_{LCC}^W > 0$, thus the mechanisms of *SCC* and *LCC* add up constructively, because they coincide with the same signs of $\theta_{SCC}^W < 0$ and $\eta_{L-S}^W \theta_{LCC}^W < 0$. Note that the spin conductivity of Zr has the same polarity with that of W, if the THz emission mainly derive from the *SCC* in Zr, the THz polarity of the Co(2)/Zr(3) and Co(2)/W should be the same. However, as shown in Figure 4 (a-c), the THz polarity of Co(2)/Zr(3) and Co(2)/W are nearly opposite, which is a direct evidence that the *LCC* dominates in the Co(2)/Zr(3)/Al₂O₃.

We include the corresponding comments in the revised manuscript. >

2) The authors perform sanity checks by comparing Co(2 nm)/Al₂O₃ with Co/Zr(3 nm)/Al₂O₃ in Fig. 1(e). However, this comparison could be problematic, as the 2 nm Co layer may partially oxidize when in contact with Al₂O₃. I suggest performing additional control experiments using symmetric stacks, such as Al₂O₃/Zr/Co/Zr/Al₂O₃, to mitigate this issue.

<Response: We thank the reviewer for the comment. Actually, we would emphases in the paper that the polarity of the THz emission remains unchanged when the Co(2 nm)/Al₂O₃ is flipped, which indicates that the dominant physical mechanism is ultrafast demagnetization, the anomalous Hall effect is not a player in the THz emission process in current case. In contrast, the polarity of THz emission from Co/Zr(3)/Al₂O₃ is reversed when the sample is flipped, which indicates the electric dipole contribution is dominate. As mentioned by the Reviewer, the 2 nm Co layer may partially oxidize when in contact with the substrate Al₂O₃. Therefore, the magnitude comparison between Co(2)/Al₂O₃ and Co/Zr(3)/Al₂O₃ is unreasonable. Unfortunately, the fabrication of an exactly symmetric stacks such as Al₂O₃/Zr/Co/Zr/Al₂O₃ is presently beyond the capabilities of our experiments.

Therefore, the sentence "Note that, the THz emission from Co/Zr/Al₂O₃ is about 20 times larger than that from Co single-layer." has been deleted in the revised manuscript. In addition, we have updated the main text with corresponding discussion. >

3) It is surprising that the orbital Hall effect (OHE), or inverse OHE, is not observed in the Co/Zr(t) layers, especially when Zr is predicted to exhibit a large OHE. The absence of this effect requires further explanation.

<Response: We are very grateful to the Reviewer for this comment. Indeed, it is not correct to

neglect any effect. As suggested by the Reviewer, both the ISHE, the IOHE, and IOREE should coexist in the THz emission from Co/Zr bilayers, according to the following observation.

(1) In Figure 2 (c), we do not see any progressive increase of active region for IOHE based THz emission at increasing thickness of Zr, which indicates that the experimental results cannot be attributed to a conversion only by IOHE in the bulk of Zr.

(2) In Figure 2 (b) plots the time delay τ_D as a function of d_{Zr} , which shows a non-monotonic dependency. When d_{Zr} increases from 3 nm to 15 nm, $\tau_D \propto (d_{Zr})^{0.495}$ associated to a concave shape, which indicates a ballistic transport regime with a long decoherent process. When d_{Zr} increases from 15 nm to 25 nm, the τ_D decreases linearly with increasing d_{Zr} .

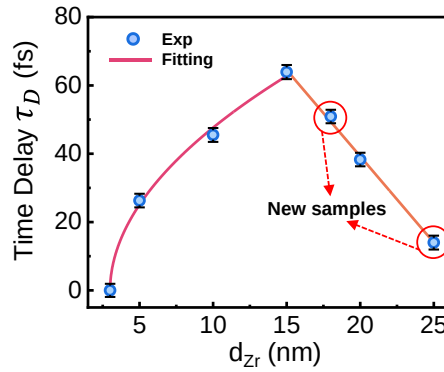


Figure 2 (b) The extracted time delay (τ_D) as a function of Zr thickness measured at pump fluence of 1.87 mJ cm^{-2}

To understand the variation, it is necessary to consider the interplay between the orbital and spin current. The orbital contribution induced by orbital-to-charge conversion (*LCC*) and a spin contribution induced by spin-to-charge conversion (*SCC*). When the thickness of Zr is larger than the orbital diffusion length of Zr, the contribution from *LCC* decreases significantly, while the *SCC* contribution increases. Thus, the τ_D becomes smaller accordingly. This non-monotonic dependence of $\tau_D(d_{Zr})$ demonstrates a crossover from the *LCC* based THz emission with delay time to the *SCC*, which generates a THz signal without time delay. At $d_{Zr} = 25 \text{ nm}$, it still remains a residual time delay of 14 fs, indicating that *LCC* still have a contribution. In summary, the THz signal in the Co/Zr (3–25 nm) heterostructures is generated by the combined contributions of *LCC* and *SCC*.

To be honest, however, our current paper does not present a quantitative decomposition of all THz emission mechanisms. In light of the complexity arising from different contributions, we remind the reader that our work demonstrates a critical role played by *LCC* in Co/Zr/Al₂O₃ heterostructures.

We have updated the main text with corresponding discussion. We add the following sentence in the revised manuscript. “Therefore, the THz generation from Co/Zr/Al₂O₃ should consists of two electric dipole contributions from both LCC and SCC mechanisms.”

4) Instead of plotting the THz amplitude in arbitrary unit, the authors should present normalized values in the main paper, with the raw data included in the supplementary material (SM).

<Response: We are very grateful to the Reviewer for this comment. Following the comment of the Reviewer, we therefore normalize the temporal THz emission spectra, and include the revised figures in the revised paper, the raw data are included in the supplementary material.

This procedure eliminates any impact of sample exchange on the pump-pulse-absorption efficiency and sample impedance. The normalized THz pulses in the article are obtained by dividing the raw data of 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 1:

$$A_{FM} = A_{total} \frac{d_{Co}}{d_{total}} \quad (1)$$

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

For the bilayer heterostructure, Z is represented by:

$$Z = \frac{Z_0}{1 + n + Z_0(\sigma_{Co}d_{Co} + \sigma_{Zr}d_{Zr})} \quad (2)$$

for the trilayer heterostructure, Z is represented by:

$$Z = \frac{Z_0}{1 + n + Z_0(\sigma_{Co}d_{Co} + \sigma_Wd_W + \sigma_{Zr}d_{Zr})} \quad (3)$$

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:

$$E = \frac{E_{THz}}{A_{FM}Z} \quad (4)$$

As shown in Figure R3 (a) and R3 (b), we provide the raw data and the normalized data of the terahertz pulses emitted from the Co/W(1)/Zr(3)/Al₂O₃. Therefore, the normalized THz signal can be used to estimate the spin/orbit-charge conversion efficiency of NM layers.

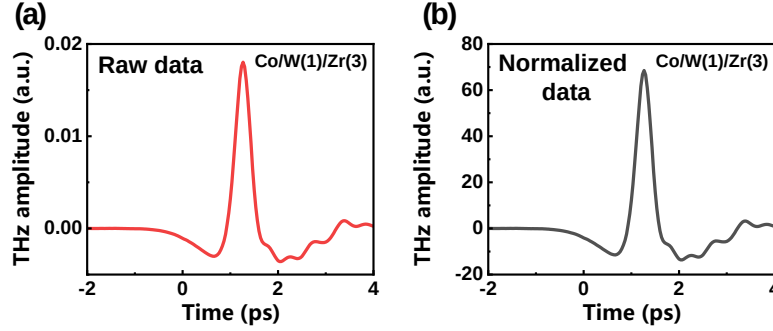


Figure R3 (a) Raw data and (b) normalized THz signal with respect to the A_{FM} and the Z for Co/W(1)/Zr(3) sample.

We present normalized values in the revised main paper, and the raw data are included in the supplementary material. >

5) In Figure 2, when analyzing the normalized THz amplitude, did the authors account for current shunting in their analysis? As the Zr thickness increases, the generated transverse charge current or voltage would naturally be reduced. This was the rationale behind attributing the effect to the inverse orbital Rashba-Edelstein effect (IOREE) rather than the inverse orbital Hall effect (IOHE).

<Response: As the referee pointed out, the rapid decrease of the signal can be due to several factors acting in parallel. When changing thicknesses and adding more interfaces, the conductivity of the stack will be changed, which could change the total laser absorptance of the FM layer and the impedance of THz emission. Therefore, for obtained the accurate spin/orbit-charge conversion efficiency by using the THz signal, we normalized the THz signals following the equation $E = \frac{E_{THz}}{A_{FM}Z}$.

As mentioned by the Reviewer, the dependence of the E as a function of Zr thickness can not be used to disentangle the effect of IOREE or IOHE. As mentioned above, according to our experimental findings, the LCC mediated THz emission is a combined effect of both interface IOREE and bulk IOHE.>

6) The explanation for the decrease in time delay in Fig. 2b for Zr thicknesses greater than 15 nm is unclear and unconvincing. The authors should provide additional data points for Zr thicknesses above 15 nm to support their claim.

<Response: We apologize that our explanation was unfortunately not clear enough. As suggested by the reviewer, additional measurements for d_{Zr} greater than 15 nm will provide a clearer trend and more direct evidence for the observed reduction in time delay. We fabricate new samples with Zr thickness above 14 nm: Co(2)/Zr(18)/Al₂O₃, and Co(2)/Zr(25)/Al₂O₃. We present new experimental results on the THz emission from Co(2)/Zr(d_{Zr})/Al₂O₃ samples, which are shown in Fig. 2 (b). To ensure consistence in our comparisons, we repeated the same experiment on Co(2)/Zr(20)/Al₂O₃, which is already included in the revised manuscript.

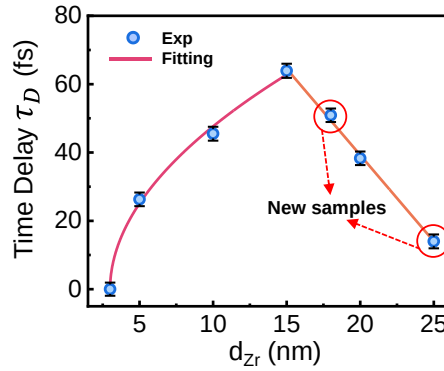


Figure 2 (b) The extracted time delay (τ_D) as a function of Zr thickness measured at pump fluence of 1.87 mJ cm^{-2}

In Figure 2 (b) plots the time delay τ_D in the peak of the emitted THz pulse as a function of d_{Zr} , which shows a non-monotonic dependency. When d_{Zr} increases from 3 nm to 15 nm, $\tau_D \propto (d_{\text{Zr}})^{0.495}$ associated to a concave shape, which indicates a ballistic transport regime with a long decoherent process. When d_{Zr} increases from 15 nm to 25 nm, the τ_D decreases linearly with increasing d_{Zr} . To understand the variation, it is necessary to consider the interplay between the orbital and spin current.

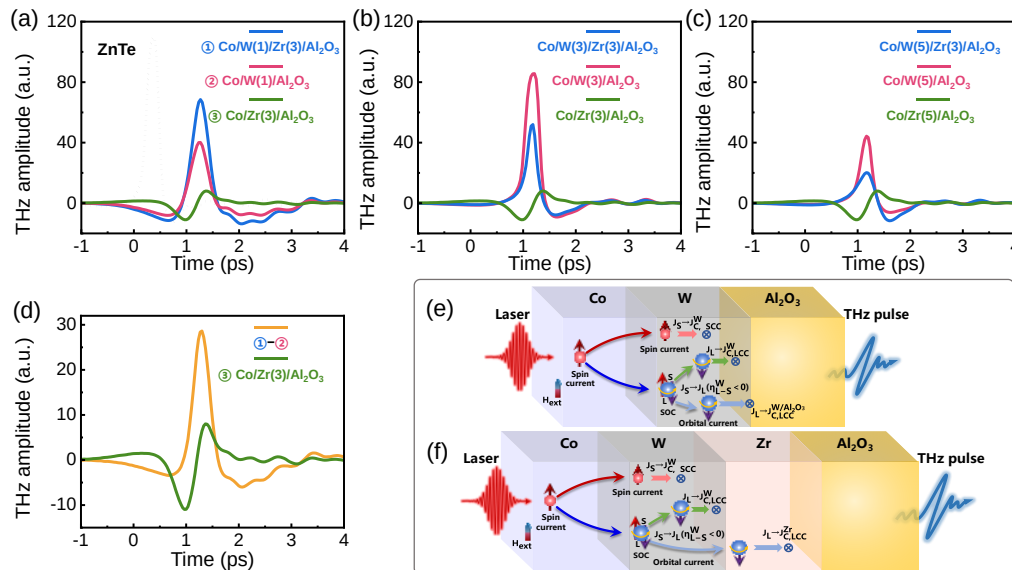
The orbital contribution induced by orbital-to-charge conversion (*LCC*) and a spin contribution induced by spin-to-charge conversion (*SCC*). When the thickness of Zr is larger than the orbital diffusion length of Zr, the contribution from *LCC* is suppressed significantly, while the *SCC* contribution increases. Thus, the τ_D becomes smaller accordingly. The non-monotonic

dependence of $\tau_D(d_{Zr})$ demonstrates a crossover from the *LCC* based THz emission with delay time to the *SCC*, which generates a THz signal without time delay. At $d_{Zr} = 25$ nm, it still remain a residual time delay of 14 fs, indicating that *LCC* still makes a minor contribution. In summary, the THz signal in the Co/Zr (3–25 nm) heterostructures is generated by the combined contributions of *LCC* and *SCC*.

We have also updated Figure 2 (b) to incorporate these additional data points and have revised the manuscript to offer a more comprehensive discussion of the potential mechanisms influencing time delay in thicker Zr layers. >

7) The results on Co/W/Zr/Al₂O₃ are ambiguous, and the coexisting mechanisms are not convincingly disentangled. Introducing a symmetric stack or varying the W thickness, as mentioned earlier, could help clarify these mechanisms.

<Response: The Reviewer indeed raises a very important point, which we address here by performing additional experiments. New samples have been fabricated and characterized. In the revised Fig. 4 (a)-(c) show the normalized THz emission results obtained from Co(2)/W($d_W=1, 3, 5$)/Zr(3)/Al₂O₃ heterostructures, in each figure, are direct compared with Co(2)/W($d_W=1, 3, 5$) and Co(2)/Zr(3) heterostructures.



We made a qualitative analysis of W thickness dependent THz emission from Co(2)/W($d_W=1, 3, 5$)/Al₂O₃ and Co(2)/W($d_W=1, 3, 5$)/Zr(3)/Al₂O₃, by the following observations in Figure 4 (a)-(c):

Observation (1): The THz signal emitted from Co(2)/W(1) is 3.65 times higher than that from

Co(2)/Zr(3). The THz signal emitted from Co(2)/W(3) is 9.3 times higher than that from Co(2)/Zr(3). The THz signal emitted from Co(2)/W(5) is 4.04 times higher than that from Co(2)/Zr(3). The polarities of THz emission from Co/W heterostructures are opposite to that from Co/Zr.

Conclusion (1): As the spin conductivity and orbital conductivity of W layer have the opposite sign, thus, the *SCC* and *LCC* induced currents should have opposite polarities. However, the spin-orbit correlation factor of W, $\eta_{L-S}^W < 0$. Since the sign of $\theta_{SCC}^W < 0$ and $\theta_{LCC}^W > 0$, thus the mechanisms of *SCC* and *LCC* add up constructively, as shown in Fig. 4 (e), because they coincide with the same signs of $\theta_{SCC}^W < 0$ and $\eta_{L-S}^W \theta_{LCC}^W < 0$. Note 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 polarity of the Co(2)/Zr(3) and Co(2)/W should be the same. However, they are different, indicating that the *LCC* dominates in the Co(2)/Zr(3)/Al₂O₃ heterostructures.

Observation (2): When we increase the thickness of W, the THz amplitude of Co(2)/W(3) is larger than that of Co(2)/W(1). While, the Co(2)/W(5) is smaller than that of Co (2)/W (3) bilayer.

Conclusion (2): This clearly indicates that the spin diffusion length in the W layer is limited. It can be expected that for a thicker W-insertion layer (W=3 nm) would have significantly reduced the spin current transmitted to the adjacent layer.

Observation (3): Co(2)/W(1)/Zr(3) sample emits nearly 1.7 and 6.24 times higher THz signal as compared to that from Co(2)/W(1) and Co(2)/Zr(3), respectively. However, the THz emission from Co(2)/W(3)/Zr(3) is nearly 60.7% of that from Co(2)/W(3). The THz emission from Co(2)/W(5)/Zr(3) is nearly 45.6% of that from Co(2)/W(5).

Conclusion (3): As the $\eta_{L-S}^W < 0$ is opposite with the $\eta_{L-S}^{Co} > 0$, the THz emission from Co(2)/W(1)/Zr(3)/Al₂O₃ shows opposite polarity of Co(2)/Zr(3)/Al₂O₃. It can be found that $E_{Co(2)/W(1)/Zr(3)/Al_2O_3}^{THz} - E_{Co(2)/W(1)/Al_2O_3}^{THz}$ and $E_{Co(2)/Zr(3)/Al_2O_3}^{THz}$ also show 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 $E_{Co(2)/Zr(3)/Al_2O_3}^{THz}$. Therefore, the role of the 1 nm W-insertion layer acts as an additional orbital transport channel rather than simply as an *SCC* and *LCC* effect itself. The physical picture is that the spin current from Co can be converted to the orbital current in W and then transmitted into the adjacent Zr layer within the Co/W/Zr/Al₂O₃ heterostructure. And then, the addition orbital

current flows into charge current, which explains the magnitude enhancement of THz emission. While, the presence of a thicker W insertion layer ($W \geq 3$ nm) would have significantly suppress the spin current, which consequently reduces the spin-to-orbit conversion in W and finally decreases the THz emission. We believe that the W-thickness dependent THz emission from Co/W/Zr/Al₂O₃ will be a valuable knowledge to this research field. Specifically, an optimum 1 nm thickness of the inserting W layer provides an additional large orbital current by its large spin-orbit coupling.

We make a corresponding comment into the main text of the revised manuscript.>

Overall, while the data presented for various samples is valuable, the analysis remains ambiguous, and the claims are not sufficiently robust. The authors should focus on one aspect of the study and strengthen their claims with more controlled experiments, which are currently lacking. If these queries are addressed and the manuscript is accordingly revised, I believe it would be suitable for publication in this journal.

<Response: In conclusion, we would like to thank the Reviewers for the constructive and relevant comments, which certainly helped us improve the quality of our manuscript. We rewrite our paper and hope that in our response we were able to address all the issues raised by the Reviewer. >

Dear Editor, dear Reviewers,

We thank all the reviewers for providing insightful comment to our manuscript. We have now taken care of all the issues pointed out by the referees in his/her specific comments as below. The revisions in the main manuscript and the supplementary information are highlighted in yellow color. >

Zuanming Jin, on behalf of all co-authors

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

I would like to thank the authors for responding to the raised concerns. However, I stand by my previous assessment that the current work does not advance the field to such an extent that it deserves to be published in Communication Physics. I am still convinced that the interpretation stands on shaky grounds regarding fluence thresholds to overcome collisions or critical temperatures to ease orbital current flow. None of these concepts are based on solid theoretical grounds.

<Response: We thank the reviewer for the valuable comments. Orbitronics has become a promising research field as the same as spintronics. Terahertz emission spectroscopy is a powerful tool to directly investigate the conversion of spin-to-orbital-to-charge current, which can help us to deeply understand the physical mechanisms and factors for orbital-current generation and transport. Our experimental results provide deep understanding of the angular momentum transfer between orbitals and crystal lattice, which will be benefit for designing the orbitronic memory devices and orbitronic THz emitters. In addition, we modified several sentences to explain the reviewer's comment in the manuscript.

(1) *We added the citation in the sentence of "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."*

(2) *The sentence of "To better understand the angular momentum transfer between orbital and crystal lattice, the THz emissions from Co(2)/Zr(3)/Al₂O₃ and Co(2)/Zr(5)/Al₂O₃ are measured at low temperatures." has been revised into "Inspired by the work of S. Kumar et al.,⁵³ 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.”

(3) We revised the following sentence in the revised manuscript. *“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 critical temperatures.”>*

Reviewer #2 (Remarks to the Author):

The authors addressed all my comments, therefore I recommend publication of the revised manuscript.

<Response: We are very grateful to the Reviewer for his/her recommending our work for publication in *Communications Physics*.>

Reviewer #3 (Remarks to the Author):

The response by Xu et al. has been partially satisfactory, as some of the questions, such as W thickness variation, have been addressed effectively, while others could benefit from further clarification. The additional experimental data involving W is commendable and adds value to the work. The comparison between Co and Ni was not intended since Ni is not ideal for ultrafast measurements. Rather I suggested Fe or Fe-rich FeCoB alloys as more suitable alternatives for comparison to verify that Fe does not produce such signals. The explanation for the notably small OHE or IOHE of Zr remains unclear, and a deeper qualitative understanding would be appreciated and it could be pursued in future work. Having said this, I acknowledge that the quality of the work has significantly improved, and I recommend it for publication.

<Response: We are very grateful to the Reviewer for his/her overall very positive evaluation of our work, and recommending our work for publication in *Communications Physics*. Following the Reviewer’s suggestion, distinguishing between IOHE and IOREE will be investigated in our future work with the advance in fabrication and characterization of heterostructures.

To further enrich the manuscript, the authors could consider discussing emerging mechanisms for generating orbital currents, such as orbital pumping and spin fluctuations, and incorporating

relevant references into the discussion.

<Response: We thank the Reviewer for raising this point. We have updated the following sentence in the revised manuscript.

“The orbital current can be generated by magnetization dynamics, the process can be referred to as orbital pumping and/or spin fluctuations^{24,57-59}. 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^{40,41,48,52-56}, the J_S in the FM layer can partially transform into the ultrafast J_L due to their relatively large η_{L-S} ”.

The following relevant references are added into the revised paper.

- [1]. Brataas, A. *Quantum scattering theory of spin transfer torque, spin pumping, and fluctuations*. *Phys. Rev. B*. **106**, 064402 (2022).
- [2]. Han, S., Ko, H. -W. & Oh, J. H. et al. *Orbital Pumping Incorporating Both Orbital Angular Momentum and Position*. *Phys. Rev. Lett.* **134**, 036305 (2025).
- [3]. Tserkovnyak, Y., Brataas, A. & Bauer, G. E. W. *Enhanced Gilbert Damping in Thin Ferromagnetic Films*. *Phys. Rev. Lett.* **88**, 117601 (2002). ” >

<Response: In conclusion, we would like to thank the Reviewers for the relevant comments. We hope that in our response we were able to address all the issues raised by the Reviewer. >