

Evidence of Ultrashort Orbital Transport in Heavy Metals Revealed by Terahertz Emission Spectroscopy

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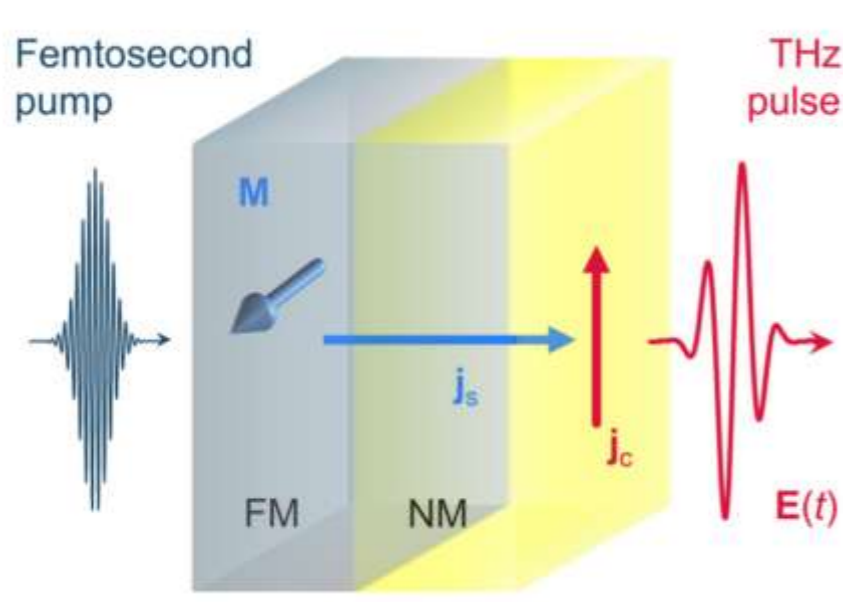
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Motivation

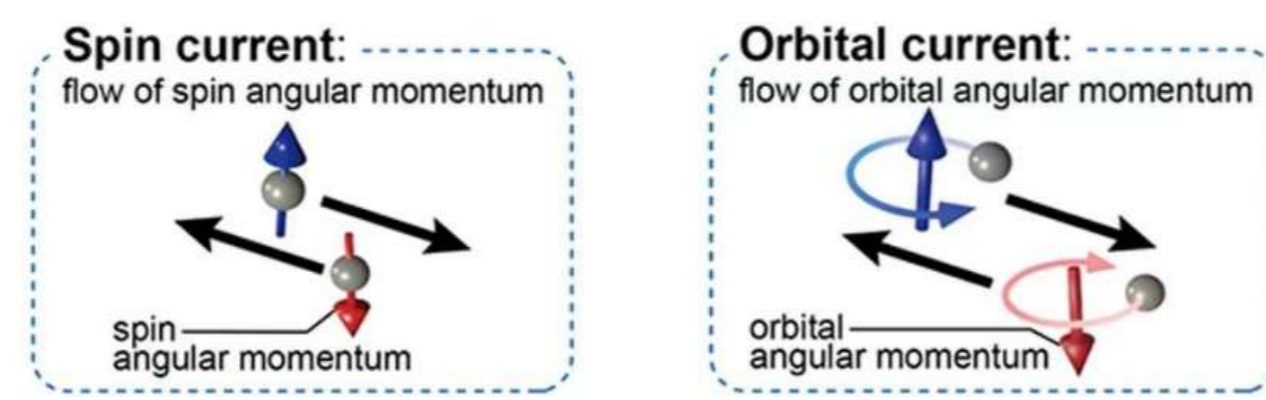
Spintronic Terahertz Emission



- ✓ Broadband THz radiation
- ✓ High electric-field amplitude
- ✓ Magnetically tunable polarization
- ✓ Ultrafast dynamics information

Promising and powerful

Orbital Current J_L



Orbital relaxation length from first-principles scattering calculations

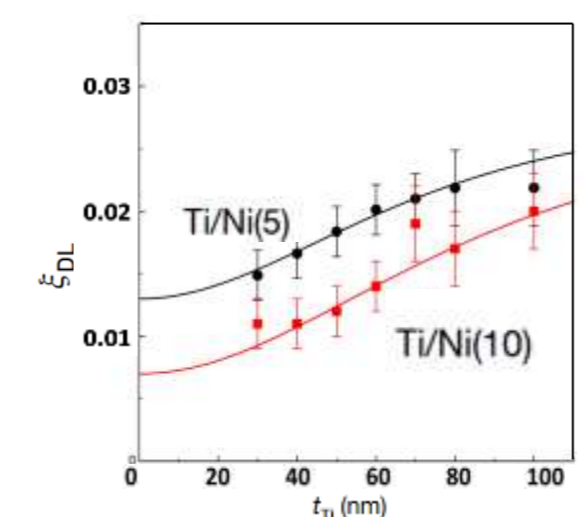
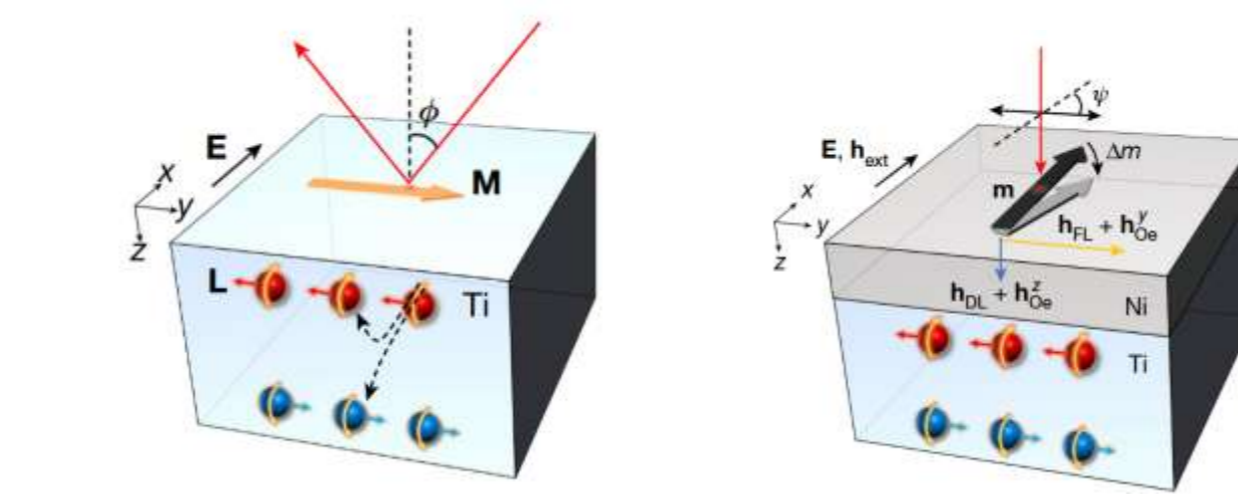
Max Rang and Paul J. Kelly
Faculty of Science and Technology and MESA⁺ Institute for Nanotechnology, University of Twente,
P.O. Box 217, 7500 AE Enschede, The Netherlands

1/e at layer No.	d (nm) per atomic layer	l_{of} (nm)	
Pt	3	0.23	0.6
Cu	2	0.21	0.4
V	3	0.14	0.5
Cr	2-5	0.15	0.3-0.8

M. Rang, *et al.*, Phys. Rev. B **109**, 214427 (2024).

Small λ_L from calculation

Experimental measurement of J_L



Y.-G. Choi, *et al.*, Nature **619**, 52 (2023).

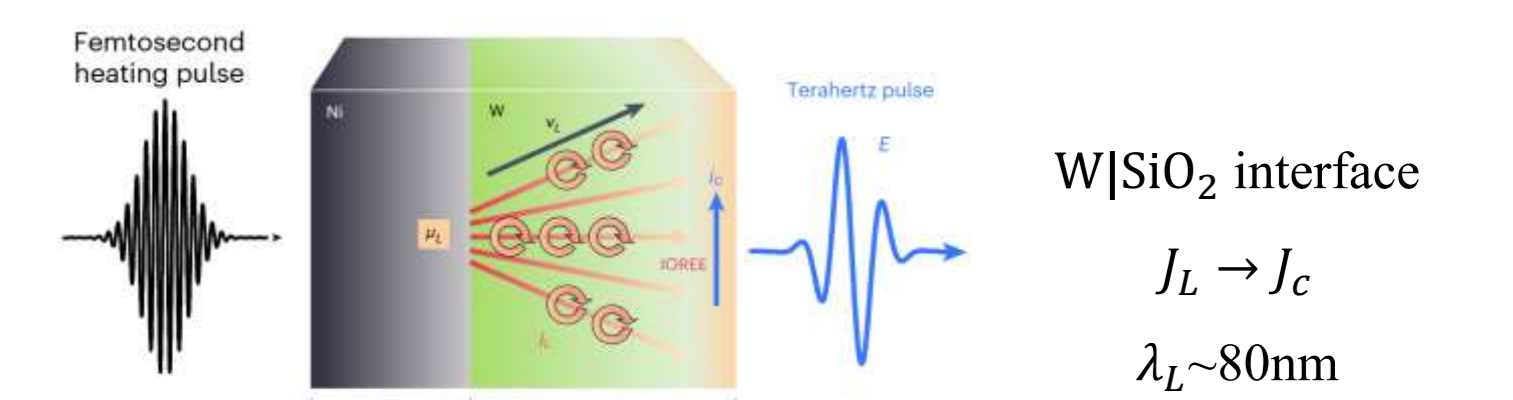
Orbital hall effect in Ti

$$J_L \rightarrow J_C$$

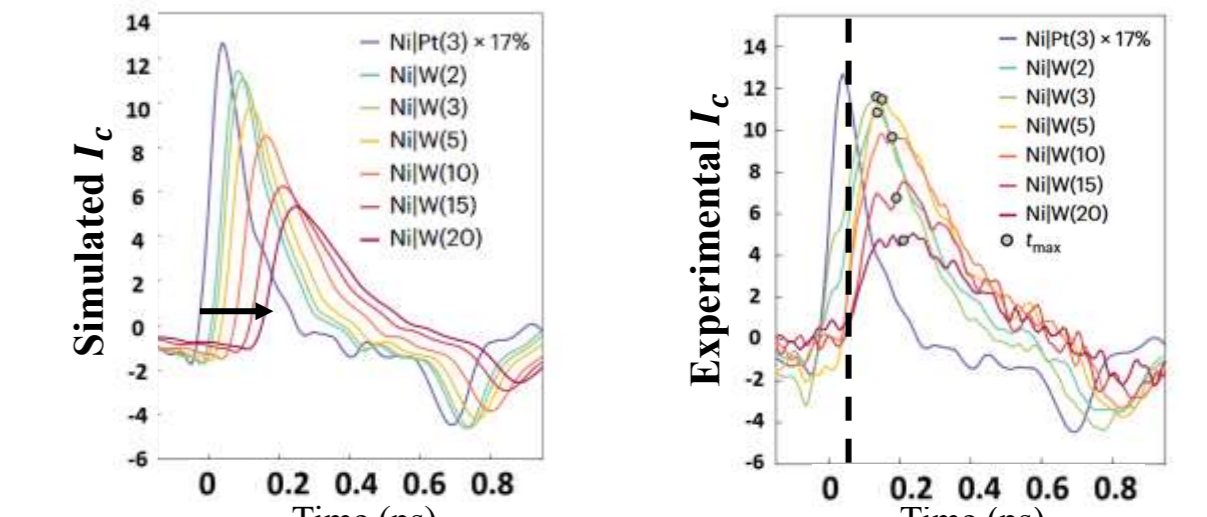
$$\lambda_L = 50 \pm 15 \text{ nm}$$

Large λ_L in LM from experiments
→ λ_L in Heavy metals(HM)?

J_L -induced THz Emission in Ni/W



IOREE: Inverse Orbital Rashba-Edelstein effect
T. S. Seifert, *et al.*, Nat. Nanotechnol. **18**, 1132 (2023).



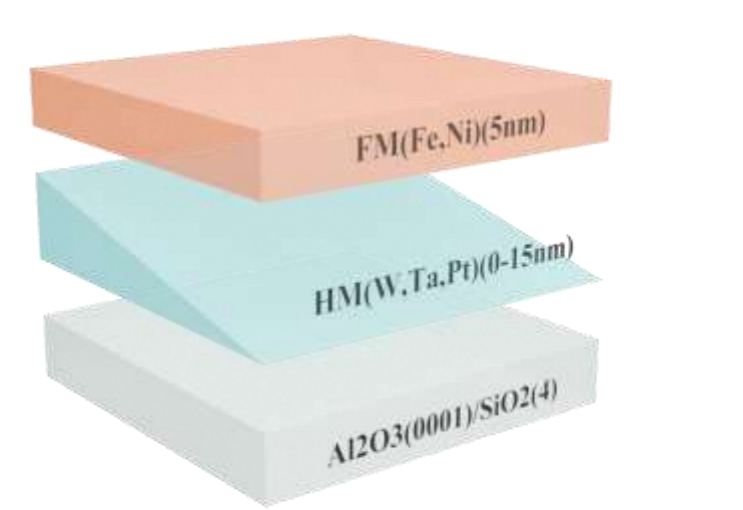
Continuously shift with d_W

Almost fixed with $d_W > 5 \text{ nm}$

THz emission mechanism in Ni/W?

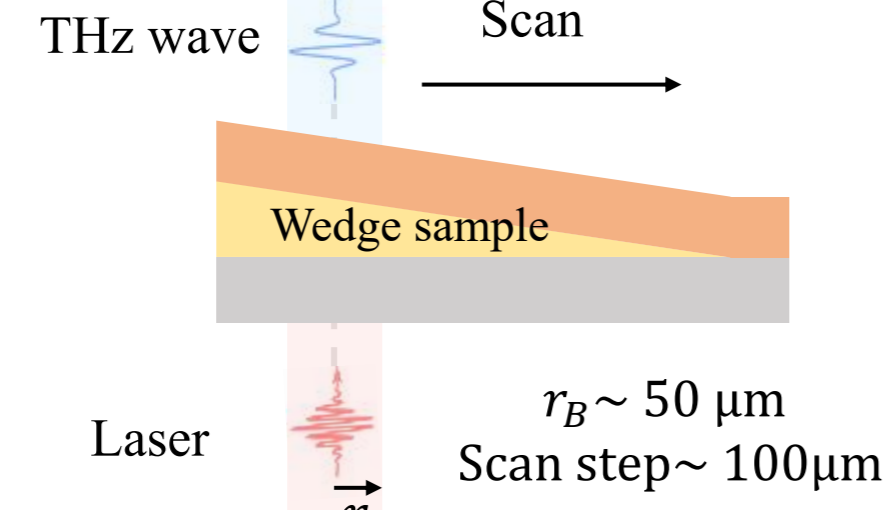
Sample Preparation and Measurement

Sample Structure



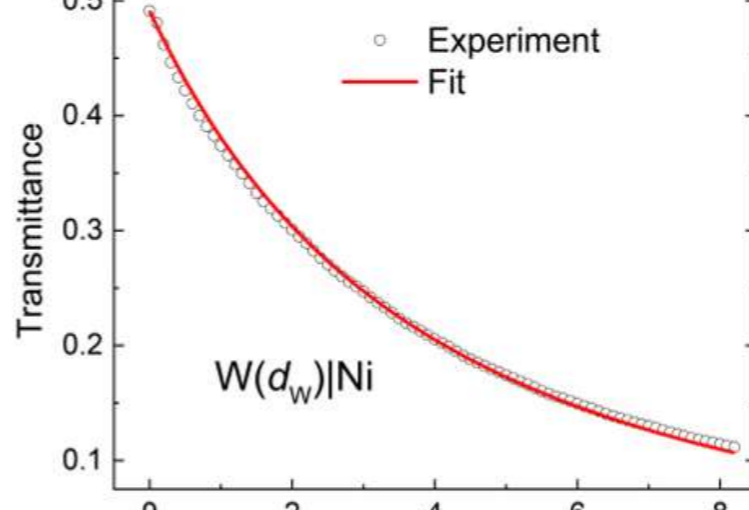
HM (W, Ta, Pt) wedge samples

Measurement



Scan along wedge direction

NIR Transmittance

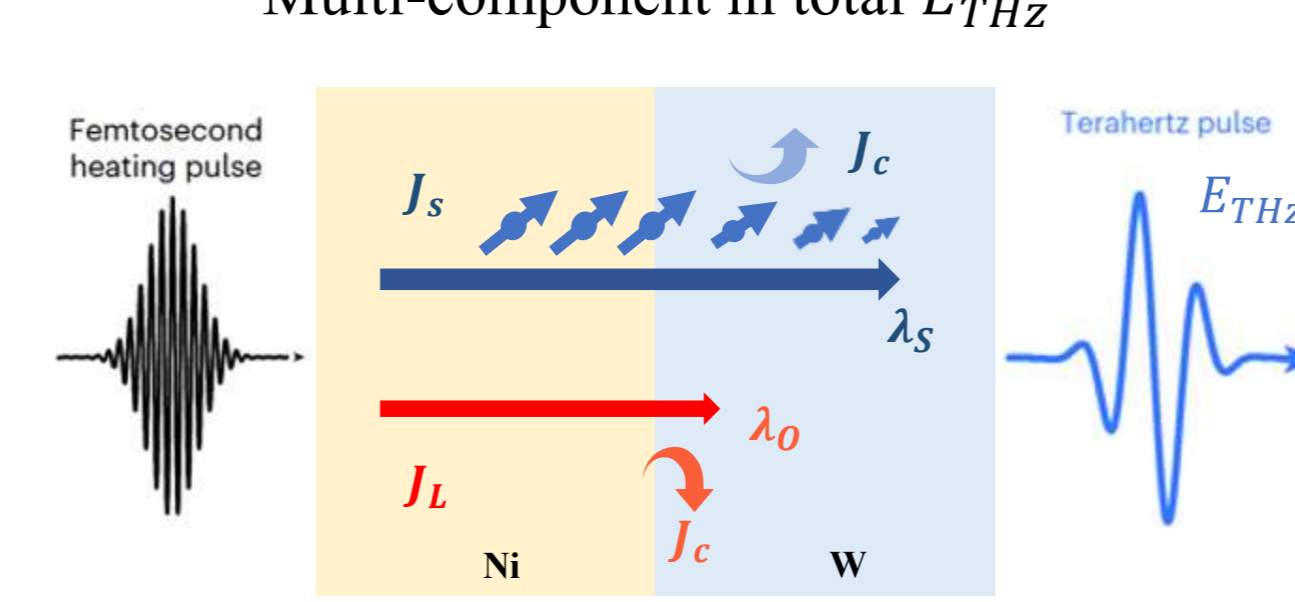


Film thickness verification

High-resolution thickness-dependent measurement on one wedge sample

Multi-component THz Emission Model

Multi-component THz Emission Model (MC-TEM)



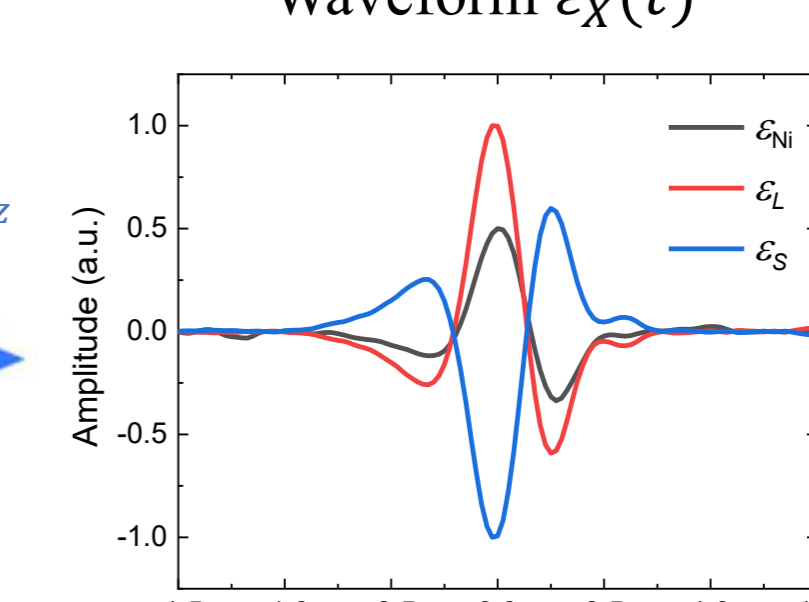
$$E_{THz} = E_S + E_L + E_{Ni}$$

$$E_S: \text{ISHE} \quad E_L: \text{IOHE}$$

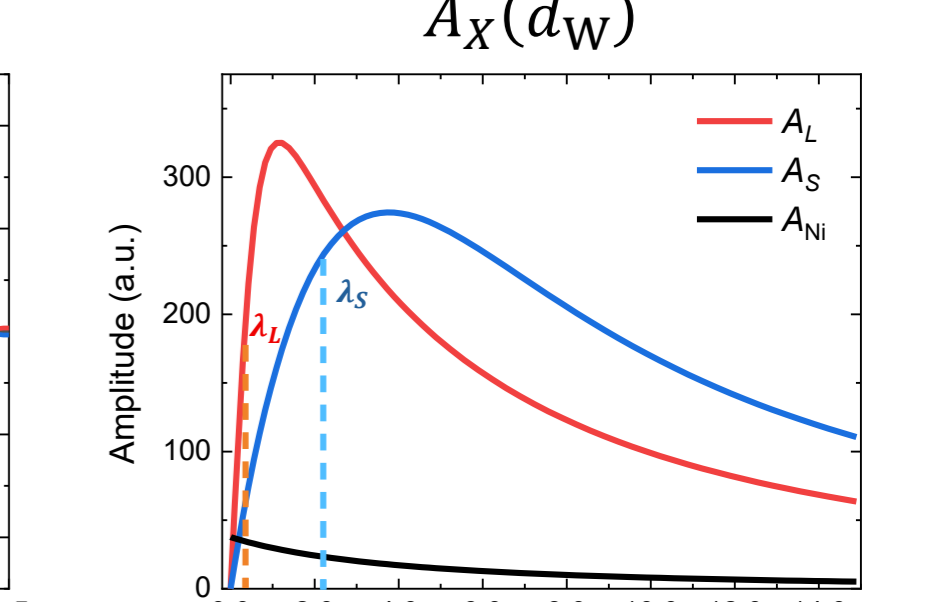
$$E_{Ni}: \text{MDE} + \text{AHE} \dots$$

$$E_X = A_X(d_W) \cdot \varepsilon_X(t) \quad (E_X \rightarrow E_S, E_L, E_{Ni})$$

Waveform $\varepsilon_X(t)$



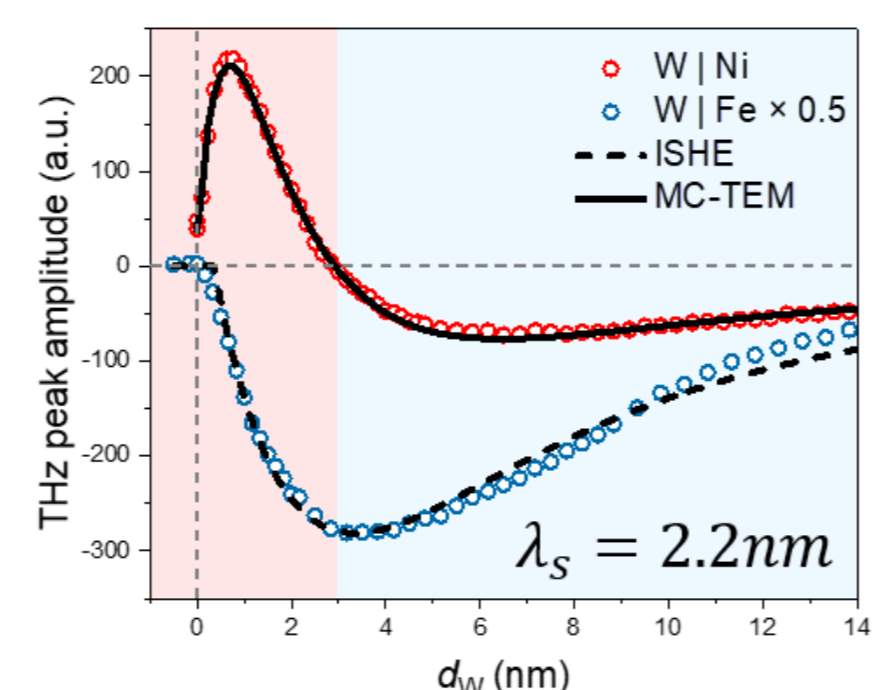
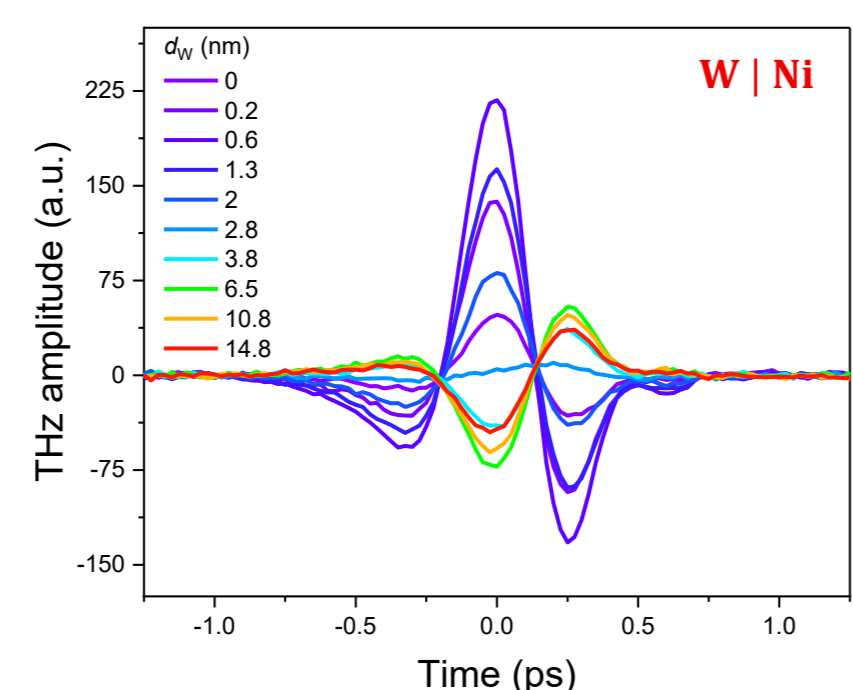
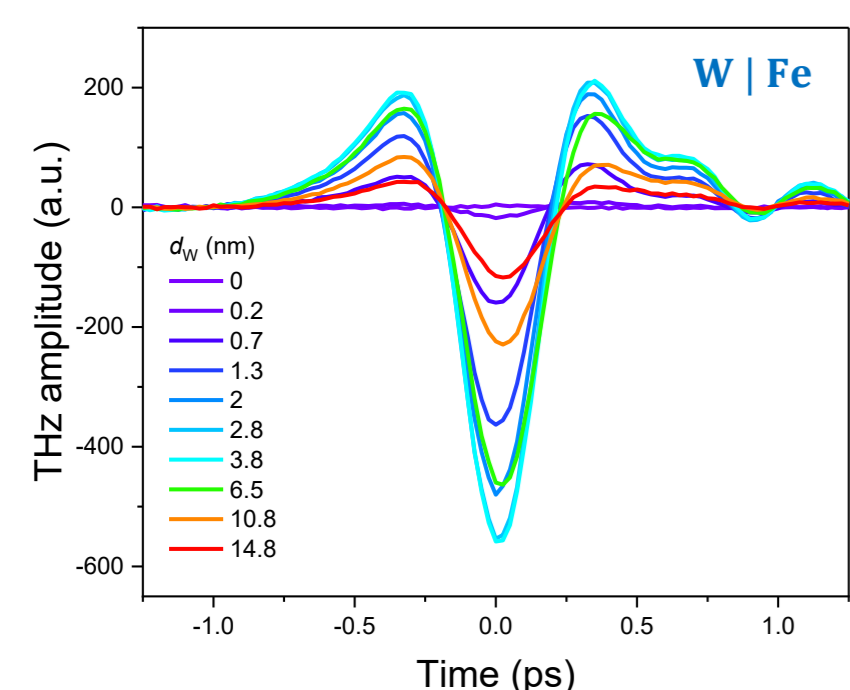
d_W -dependent amplitude $A_X(d_W)$



Experimental data → $\varepsilon_X(t)$ → $\tilde{A}_S, \tilde{A}_L, \lambda_S, \lambda_L \rightarrow A_X(d_W)$
 $E_{THz} = \sum_X A_X(d_W) \cdot \varepsilon_X(t)$ → fit experimental data
 $\tilde{A}_S, \tilde{A}_L, \lambda_S, \lambda_L$

d_W -dependent Polarity Reversal

d_W -dependent THz Emission Spectroscopy



$\lambda_S = 2.2 \text{ nm}$

ISHE: Inverse Spin Hall Effect

MC-TEM: Model in this work

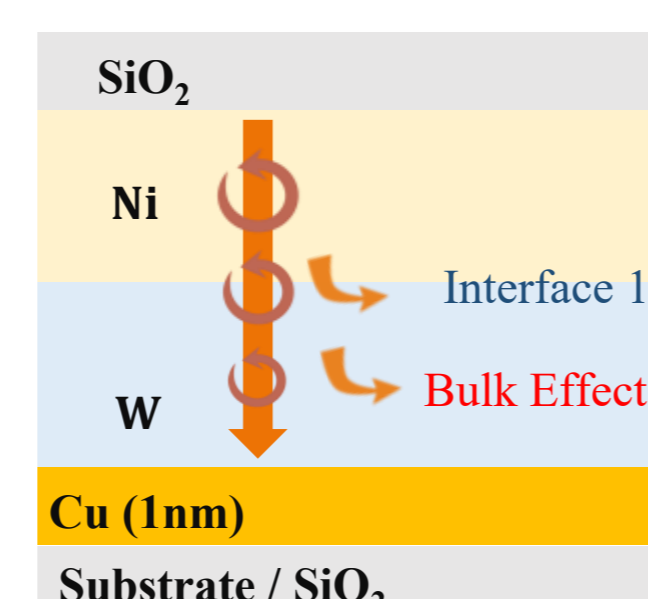
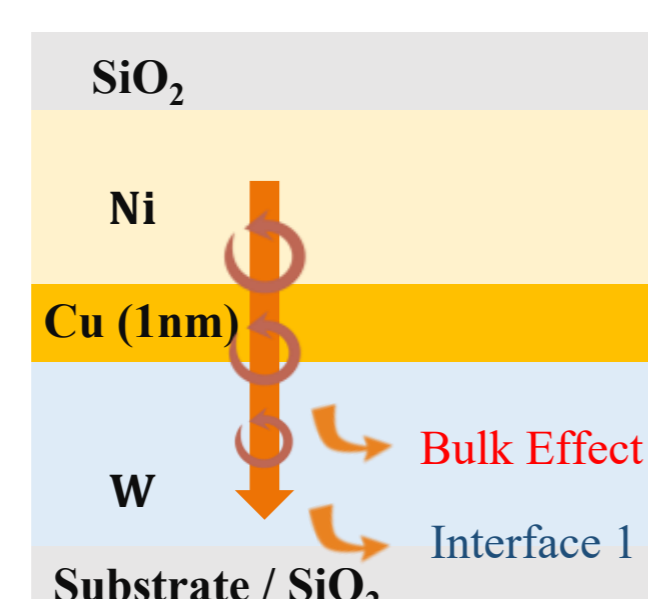
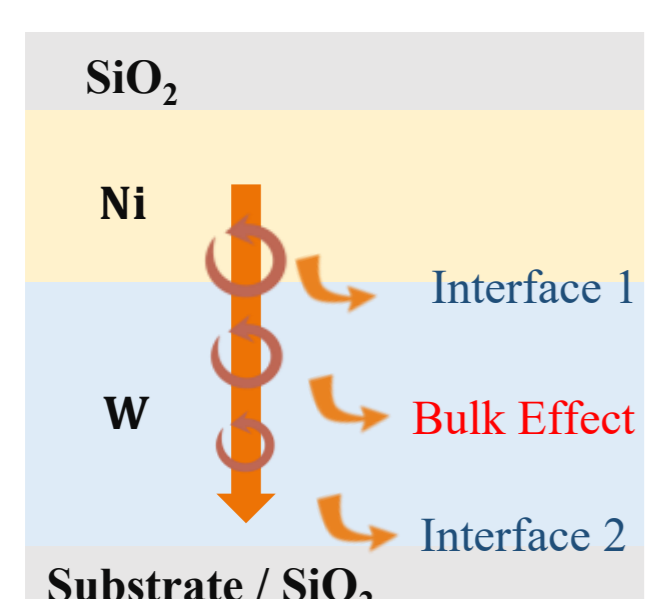
➤ Polarity reversal with d_W was observed in W|Ni

Laser-pumped orbital currents J_L in Ni

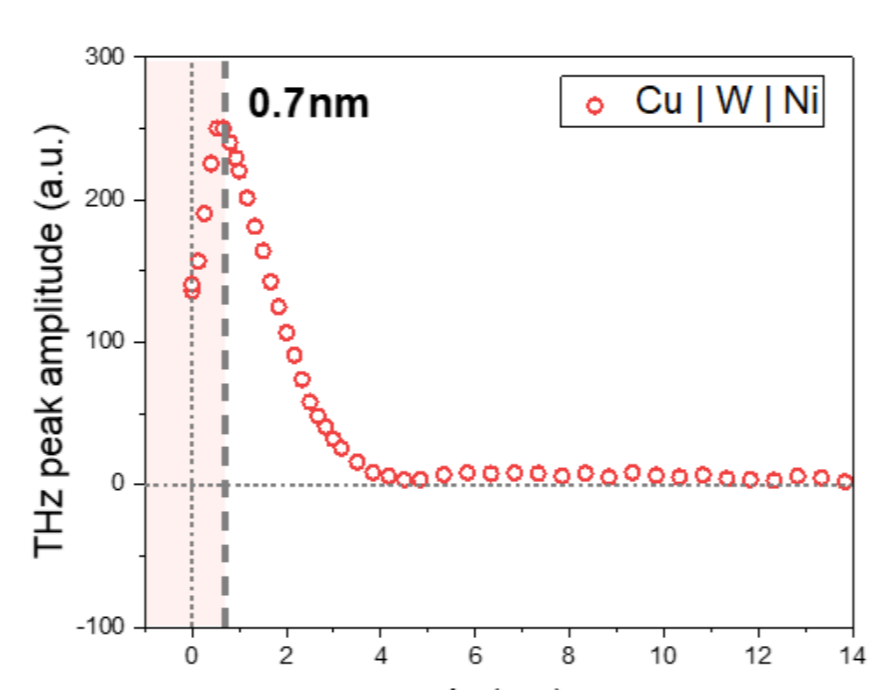
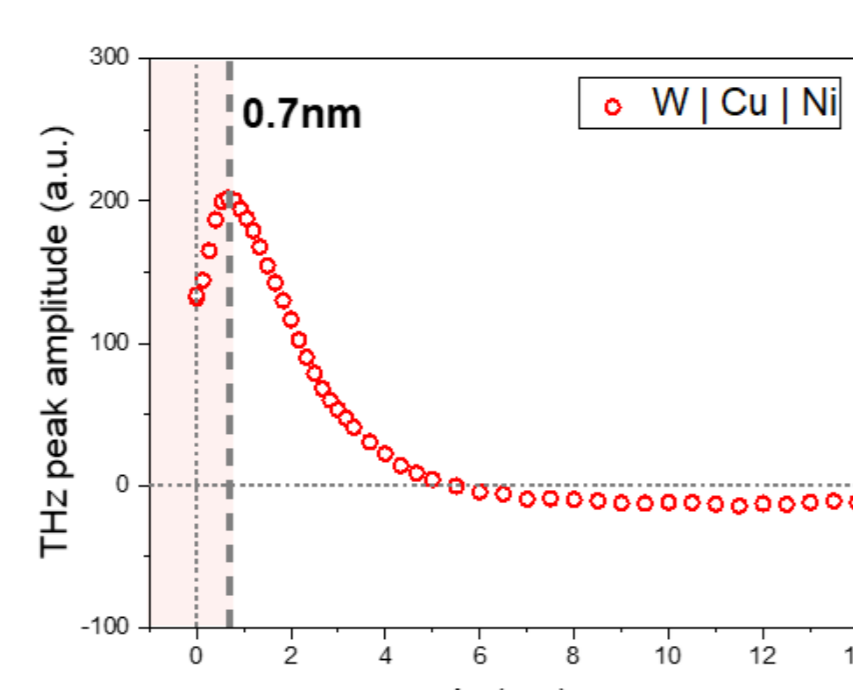
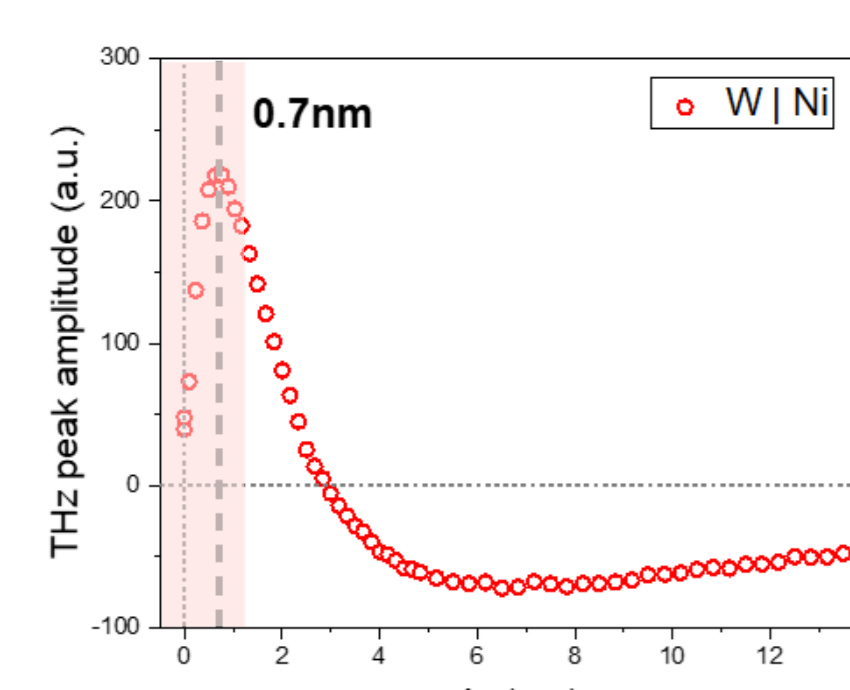
- W|Fe: THz spectroscopy agrees well with ISHE THz emission model → $\lambda_S = 2.2 \text{ nm}$
- W|Ni: Opposite polarity to W|Fe with $d_W < 2 \text{ nm}$ → origins from J_L in Ni

Ruling Out Interfacial Effects

Control Samples with Cu Insertion Layers



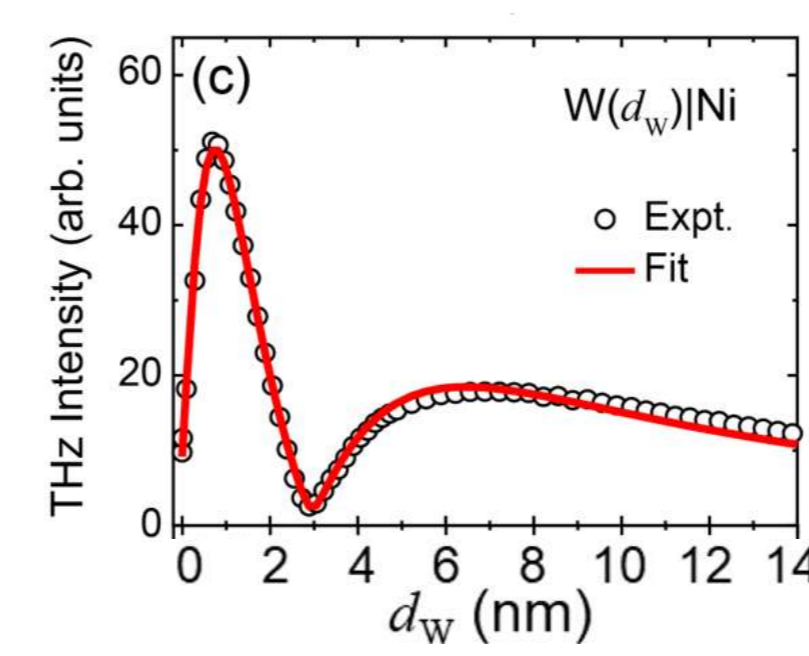
d_W -dependent THz Peak Amplitude



Rapid THz signal increase with $d_W (< 0.7 \text{ nm})$ remains consistent

× Interfacial effect(IOREE...) ✓ Bulk conversion(IOHE)

Fitting Results Using MC-TEM

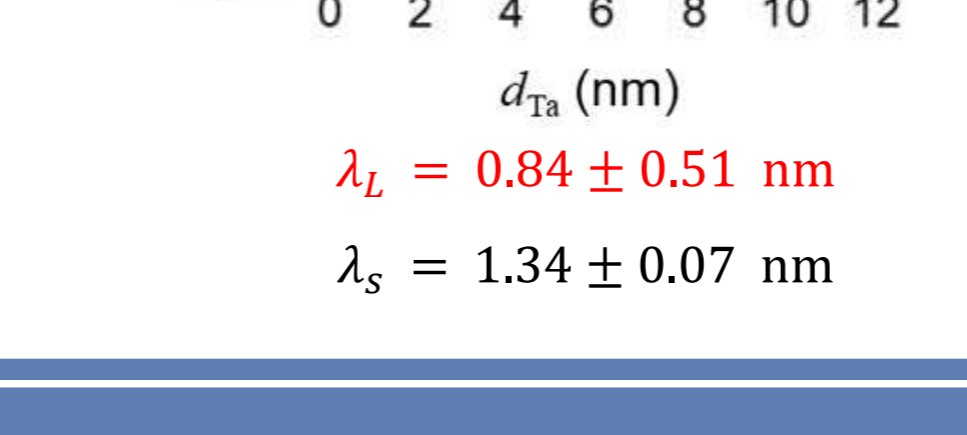
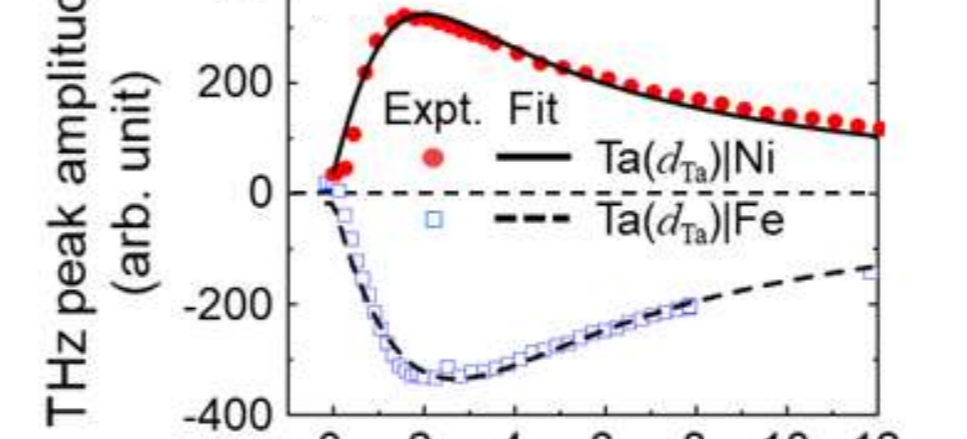
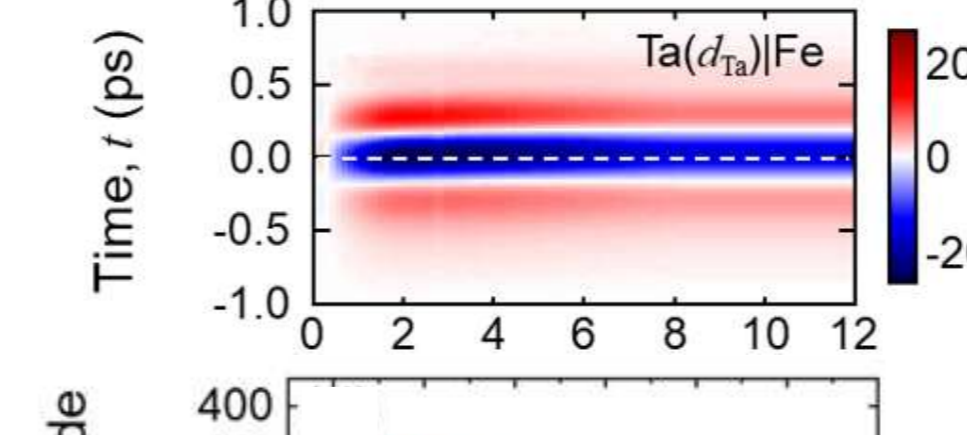
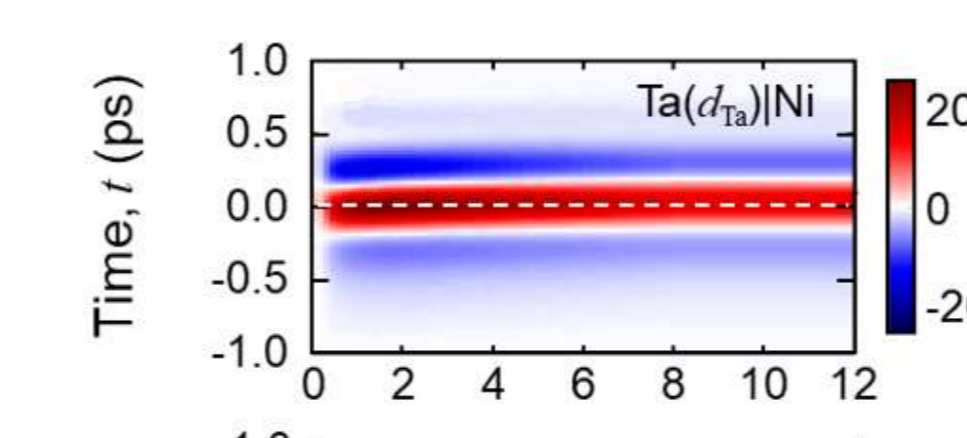


$$\lambda_L = 0.36 \pm 0.17 \text{ nm}$$

$$\lambda_S = 2.20 \pm 0.13 \text{ nm}$$

Fitted λ_L is extremely short in W!

Fitted λ_L in other HMs (Ta, Pt)



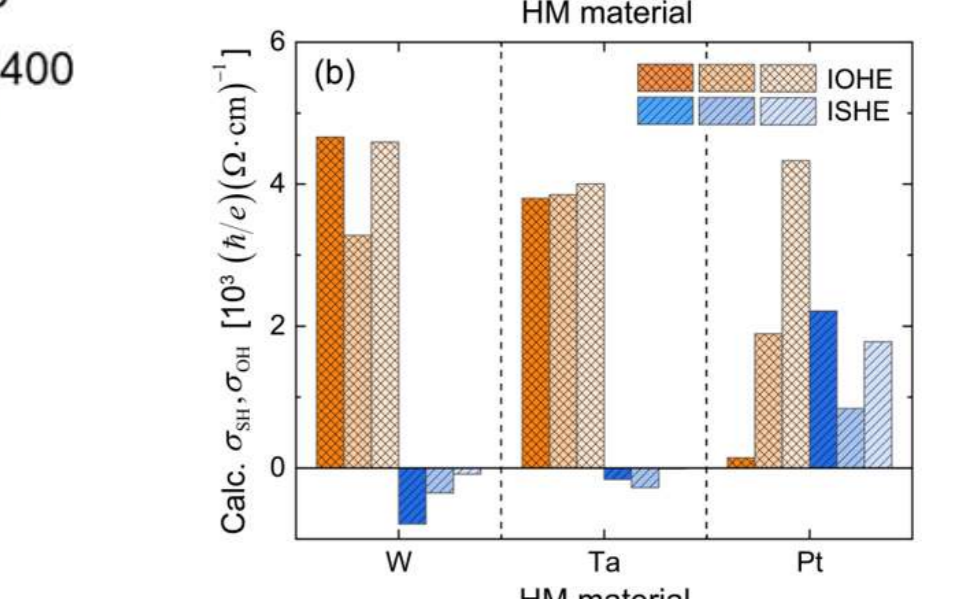
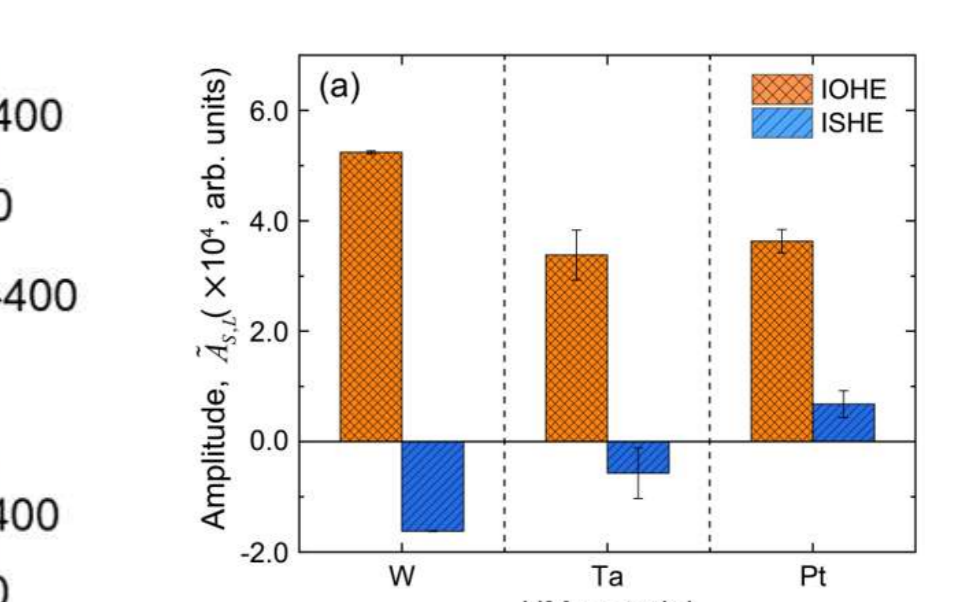
$$\lambda_L = 0.84 \pm 0.51 \text{ nm}$$

$$\lambda_S = 1.34 \pm 0.07 \text{ nm}$$

$$\lambda_L = 0.94 \pm 0.44 \text{ nm}$$

$$\lambda_S = 2.00 \pm 0.34 \text{ nm}$$

Fitting results agree well with experimental data



The signs of \tilde{A}_S, \tilde{A}_L are consistent with theoretical σ_{SH} and σ_{OH} .

➤ Ultrashort fitted $\lambda_L (< 1 \text{ nm})$ in heavy metals!

Conclusion

Ultrashort orbital transport in Heavy metals

- Multi-component THz emission spectroscopy was observed in HMs/Ni.
- Interfacial conversion mechanisms are ruled out.
- Experimental data show excellent agreement with the MC-TEM.
- Ultrashort λ_L is revealed in HMs.