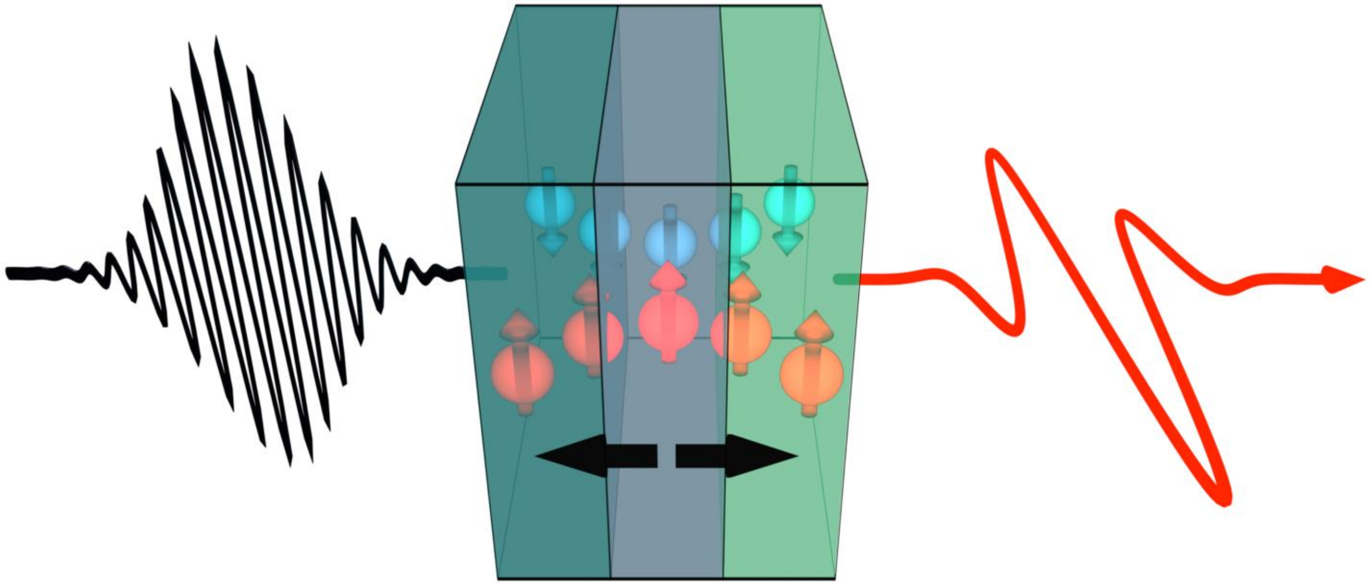
Terahertz spinorbitronics

Driving and probing spin and orbital currents at highest frequencies



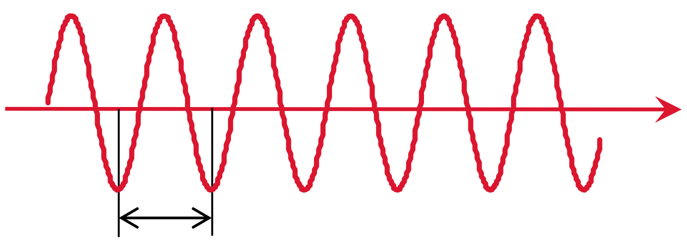
Tom S. Seifert
Project Leader
Terahertz Physics Group
Freie Universität Berlin

Freie Universität Berlin

Terahertz (THz) radiation

Electromagnetic wave

1 THz



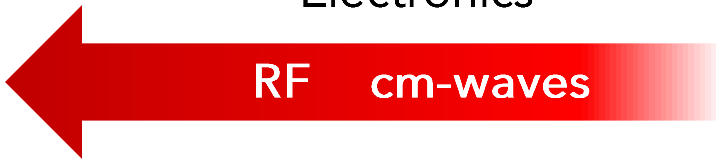
300 µm

1 THz
$= 10^{12}$ Hz
$= (1\text{ps})^{-1}$

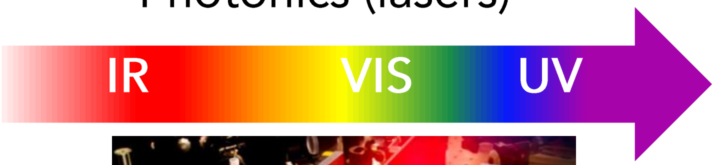
Electronics

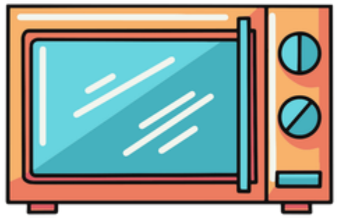
THz window

Photonics (lasers)

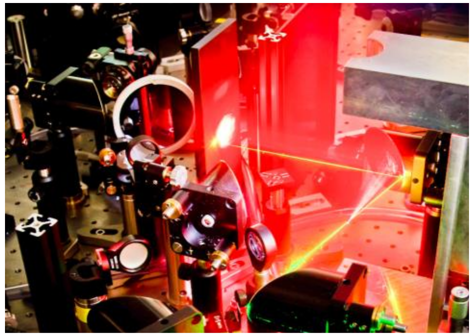


0.3 … 30 THz





1 THz ≙ 4 meV

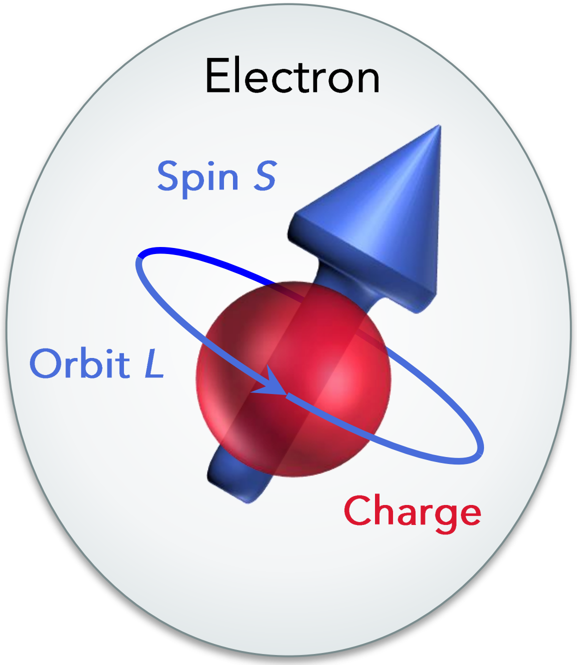


Why combining THz & spin/orbital transport?

Review: P. Lu,..TSS,.., M. Jarrahi, Nanophot. 10.1515 (2022)

Spin and orbit as information carrier

Spinorbitronics



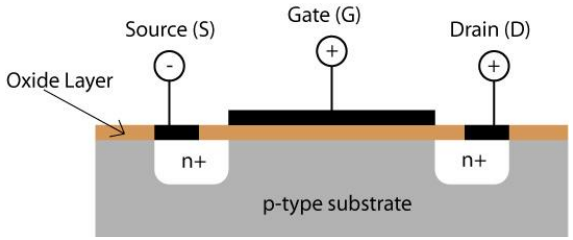
Encode information by *S* or *L*

Conventional electronics: towards terahertz rates

Next-gen WiFi (6G): 0.3 THz

Zhang *et al.*, IEEE VTM (2019)

fastest FET: ~1 THz cut-off

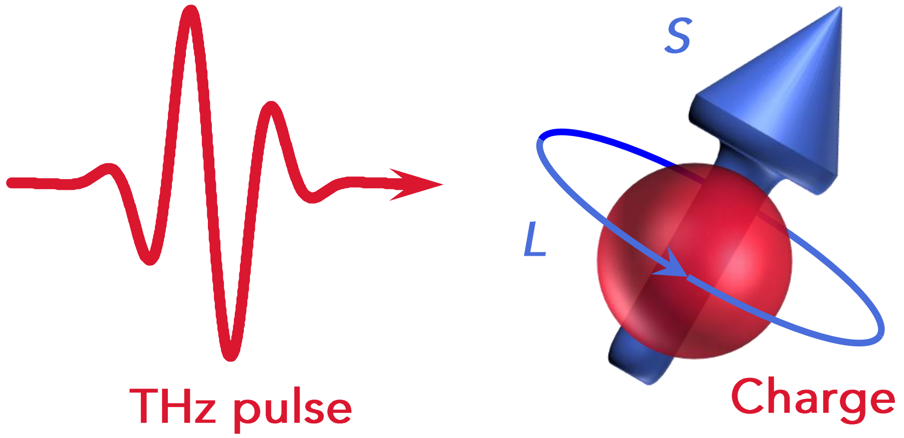


Del Alamo, Nature (2011)

Low-dissipation spinorbitronics → overcome GHz data-processing limit

Spinorbitronics needs to work at THz rates

Terahertz spinorbitronics

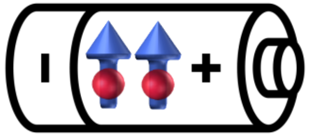


Guiding questions:

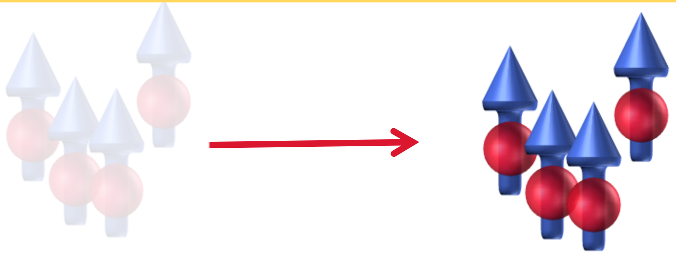
1. Spinorbitronic effects operative at THz rates?
2. Can we discriminate *S* vs *L* dynamically?
3. Exploite in terms of THz photonics?

Ultrafast spinorbitronics

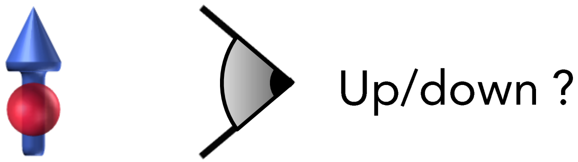
1. Need an angular momentum battery



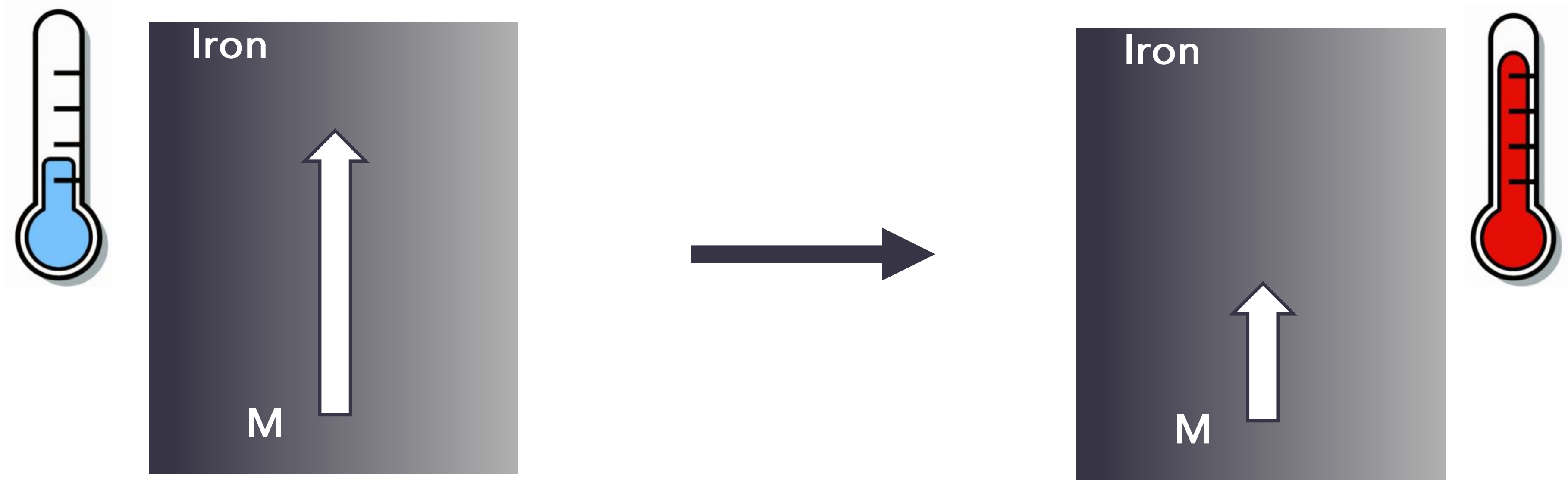
2. Transport & manipulate



3. Sense

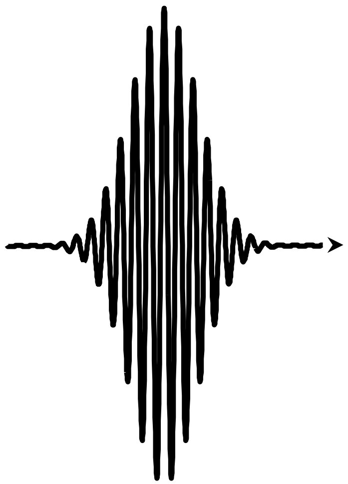


Heating a ferromagnet

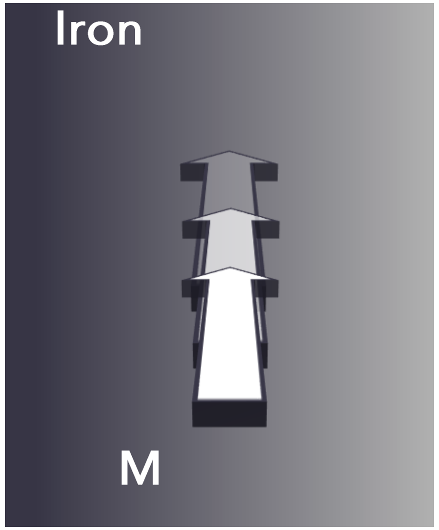


Ultrafast?

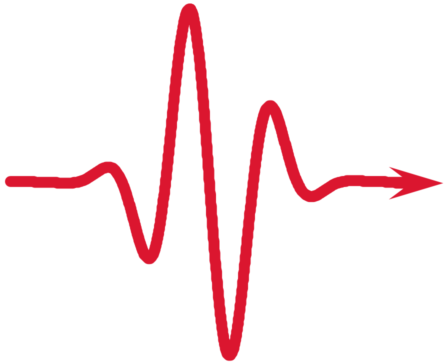
Ultrafast heating of a ferromagnet



Femtosecond heating pulse



Ultrafast magnetization quenching

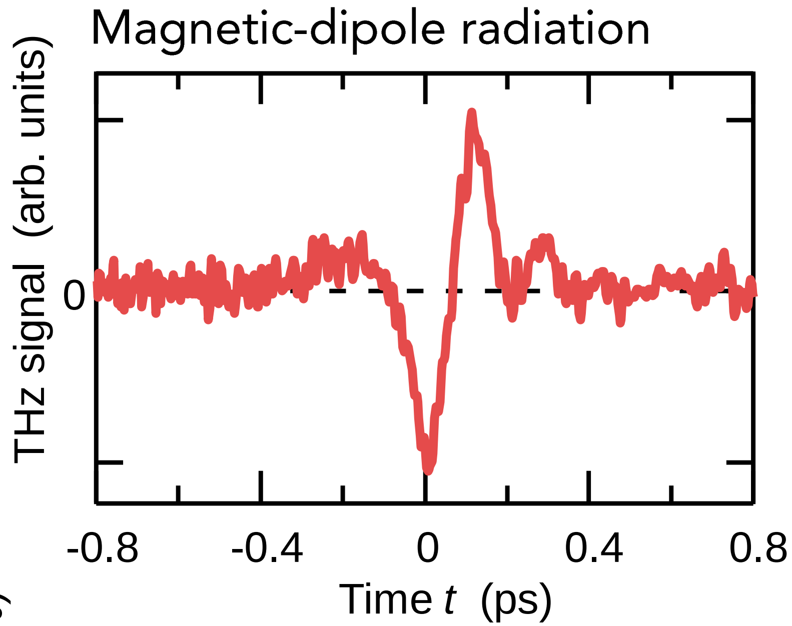


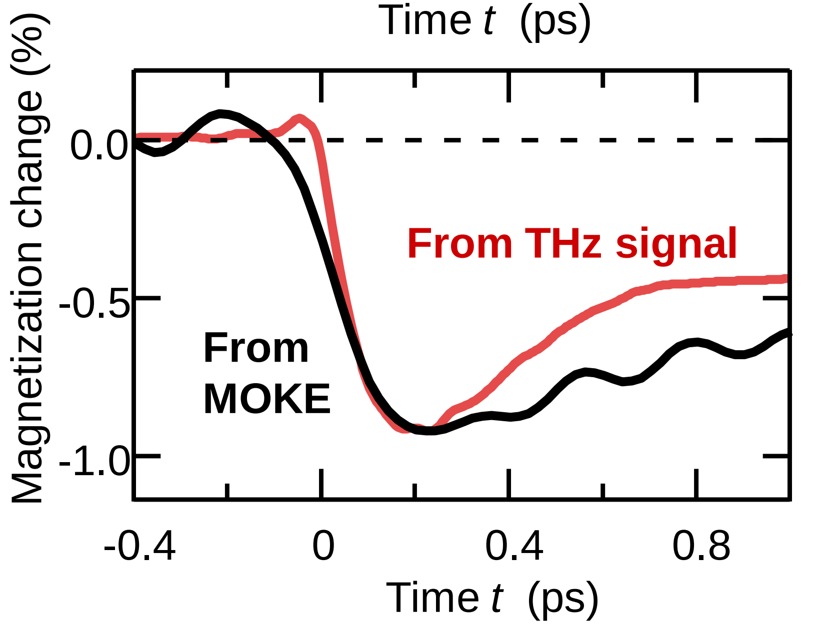
$$E_{\mathrm{MD}}(t) \propto \partial_t{}^2 M(t)$$

Beaurepaire *et al.*, PRL (1996)

Magnet wants to lose spins within 200 fs.

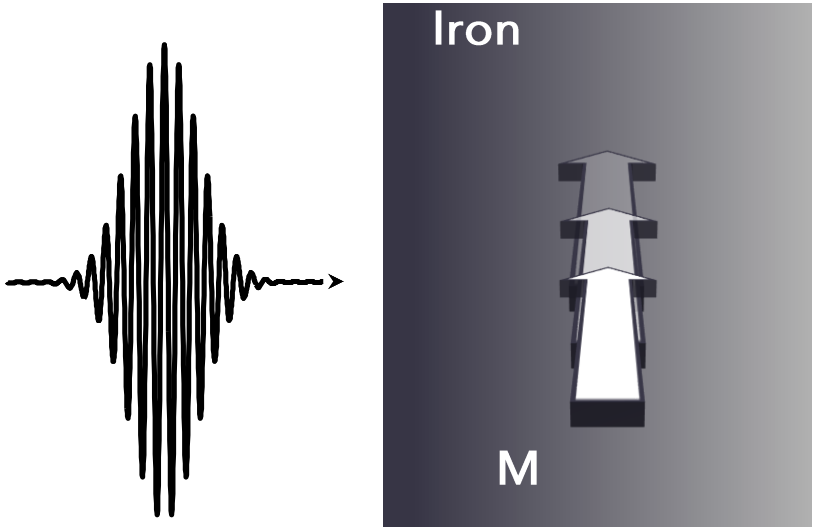
How to understand?



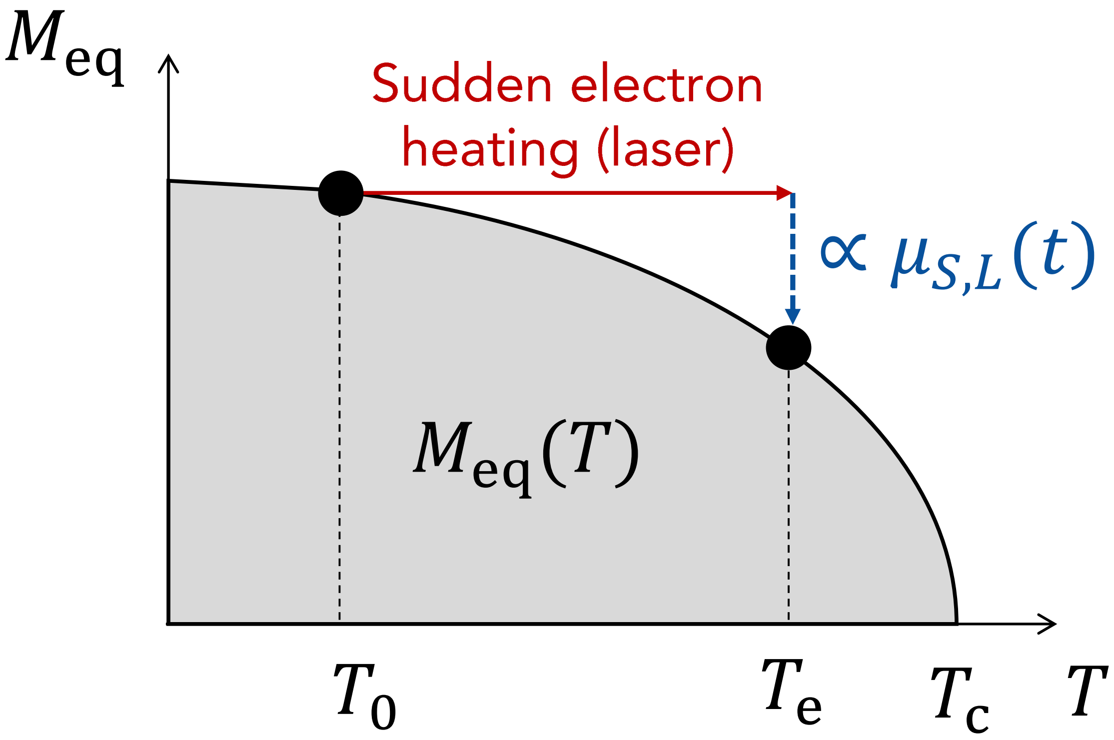


W. Zhang, TSS, Turchinovich *et al.*, Nat. Commun. 11 (2021).
R. Rouzegar,..,TSS *et al.*, PRB (2022)

Ultrafast heating of a ferromagnet



Femtosecond heating pulse



$\mu_{S,L}(t)$ is an excess magnetization: "Spin/Orbital voltage (accumulation)"

Relaxes locally

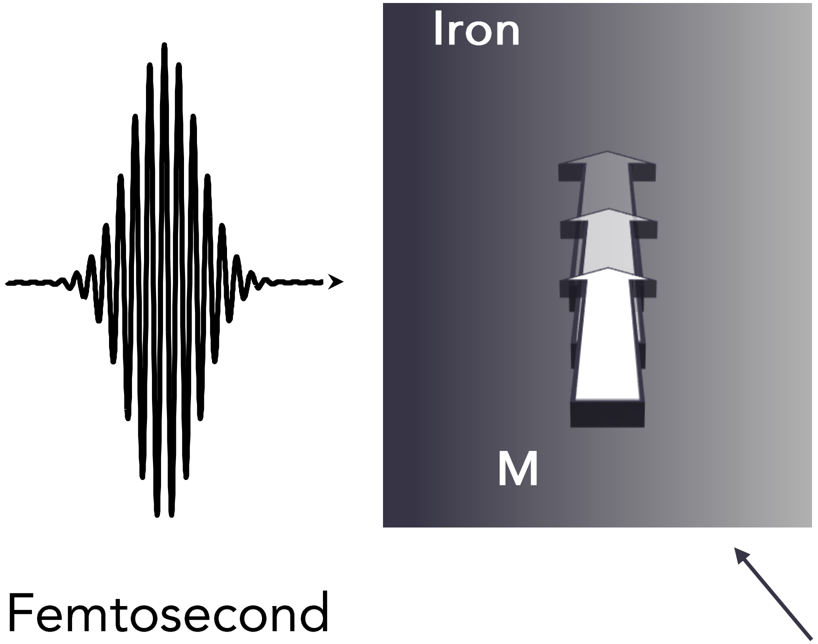
Choi, Cahill *et al.*, Nat. Comm. (2014)

Mueller, Rethfeld, PRB (2014).

R. Rouzegar,.., TSS *et al.*, PRB (2022)

Gupta, Oppeneer *et al.*, PRB (2023)

Ultrafast heating of a ferromagnet



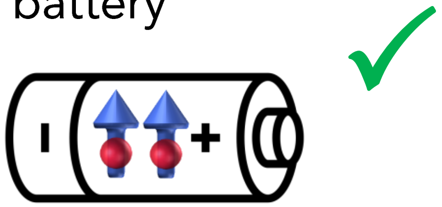
Femtosecond heating pulse

Ultrafast magnetization quenching

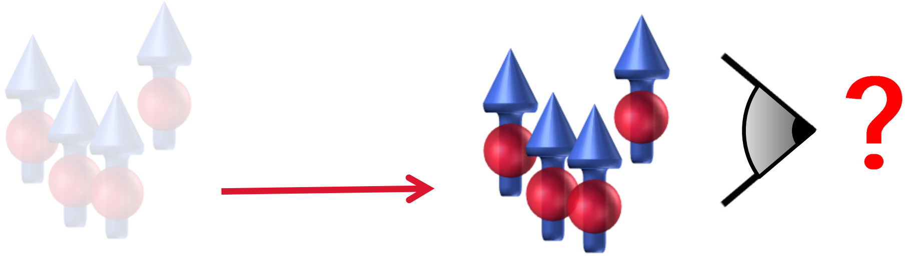
200 fs spin voltage

Spin information processing:

0. Need a THz spin battery

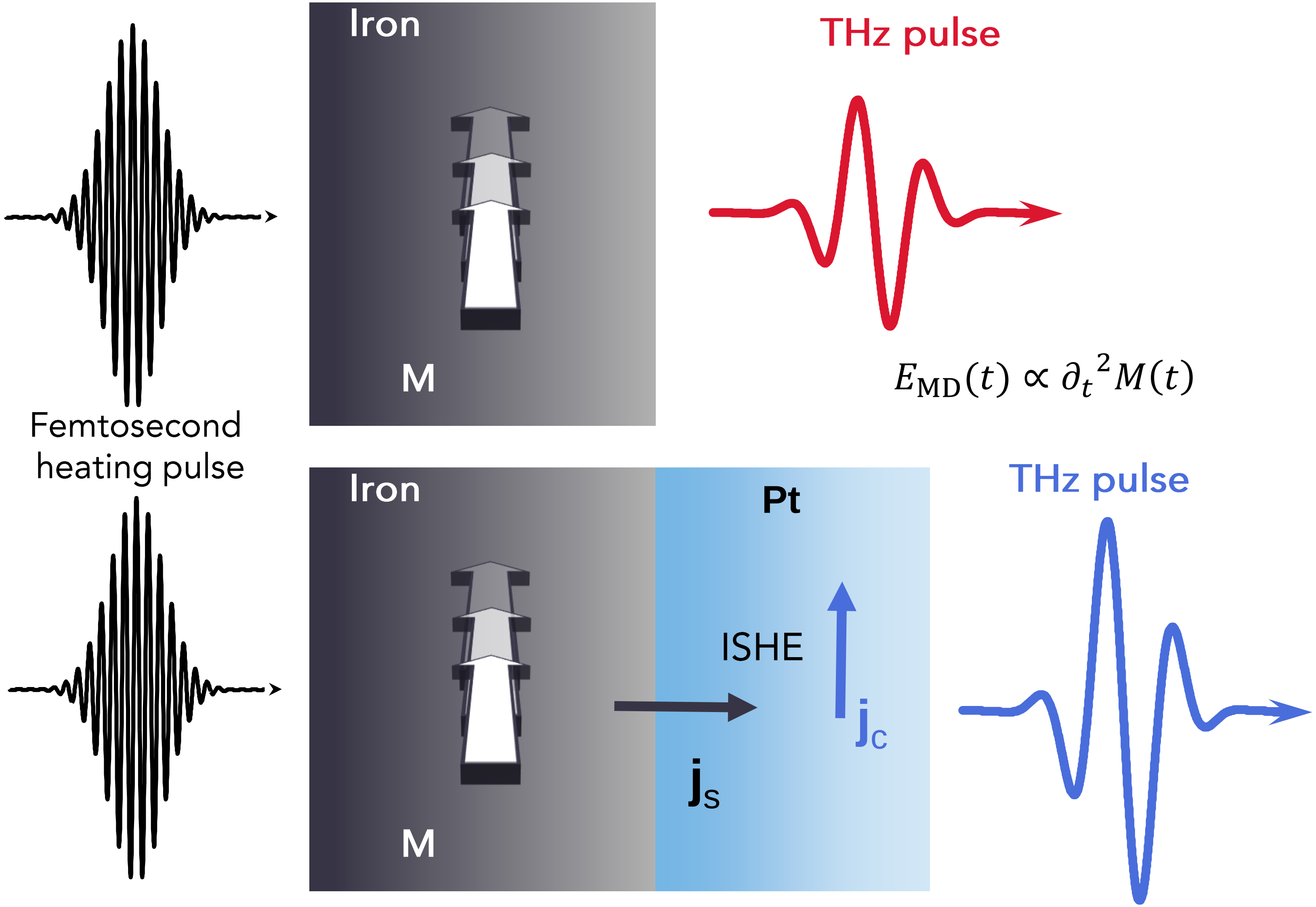


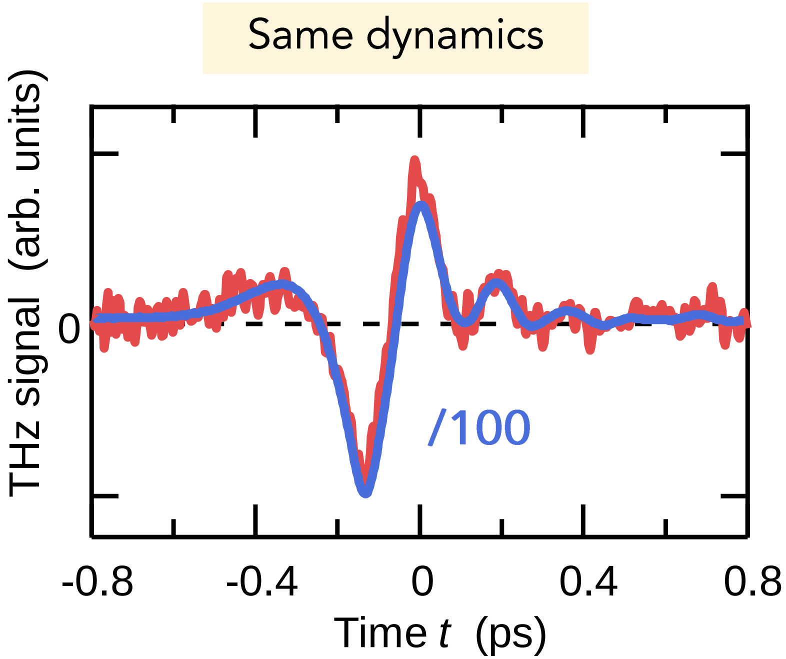
1. Transport spins through space (fast) + detect



Idea: inject a spin current into an adjacent layer.

R. Rouzegar,..,TSS *et al.*, PRB (2022)

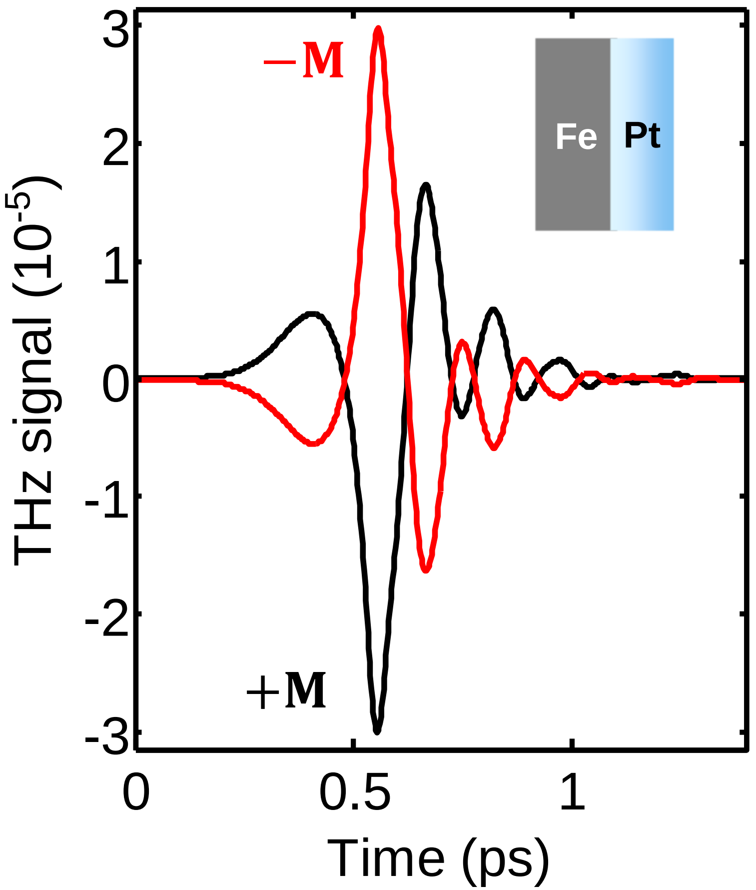


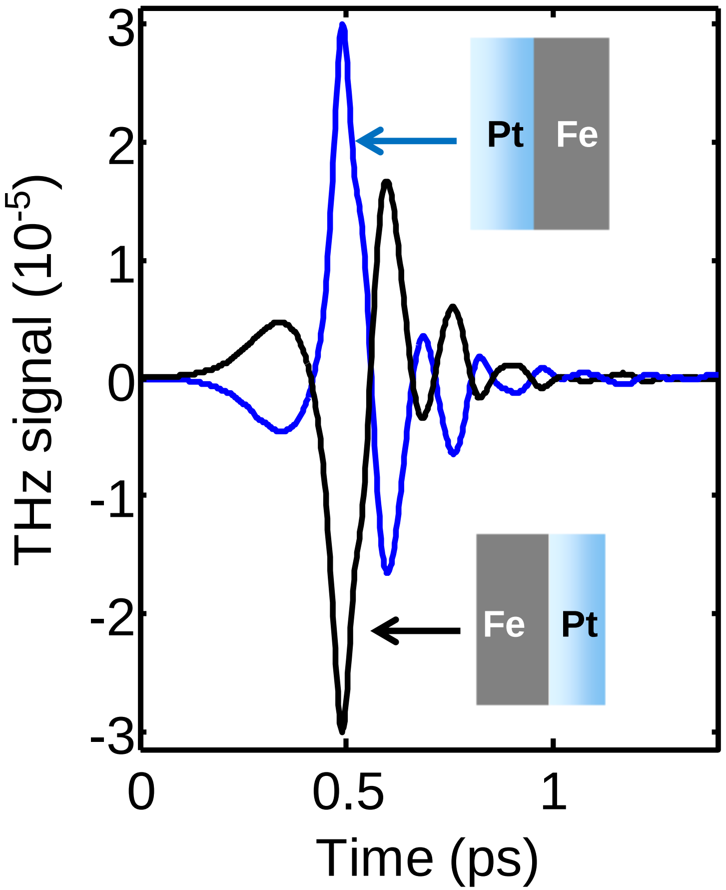


Why so strong?
Really spin transport?

R. Rouzegar,..,TSS *et al.*, PRB (2022)

THz inv. spin Hall effect





- THz emission is magnetic
- THz electric field $\perp \mathbf{M}$

- THz waveform reverses for reversed sample

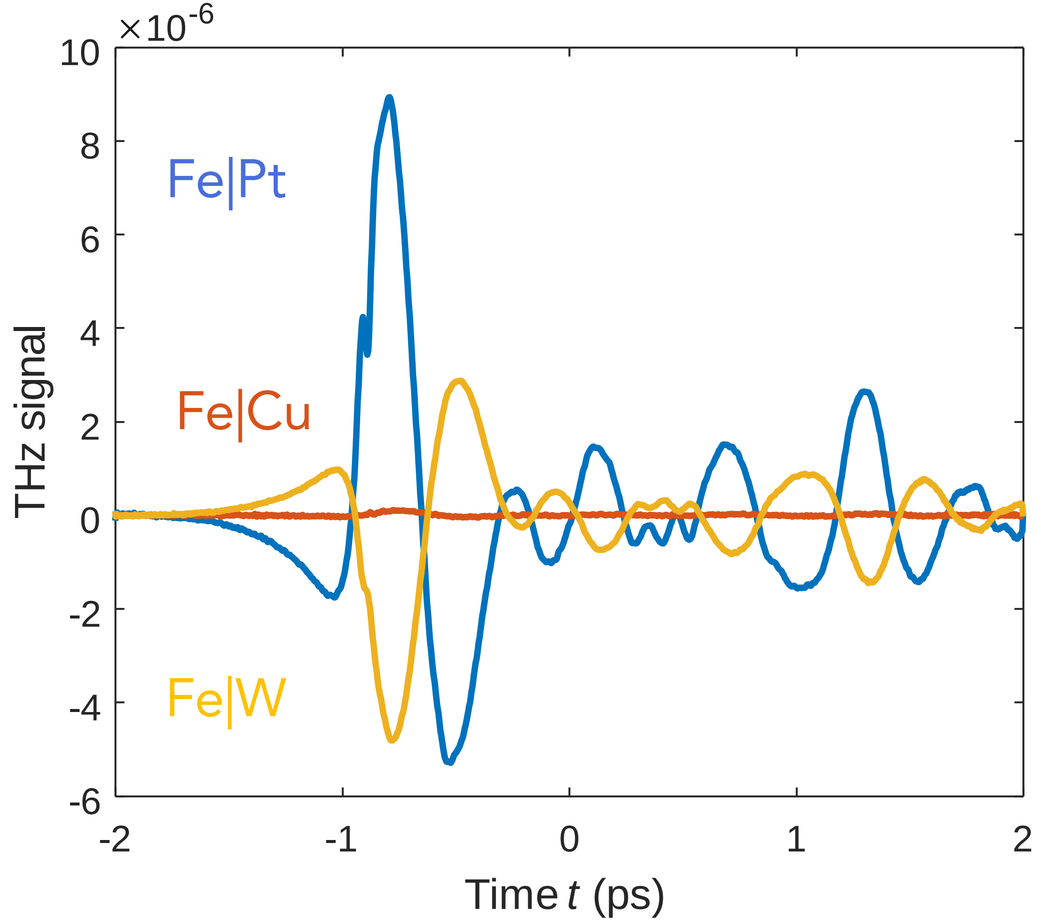
Electric dipole $\perp \mathbf{M}$ as expected from **inv. spin Hall effect**

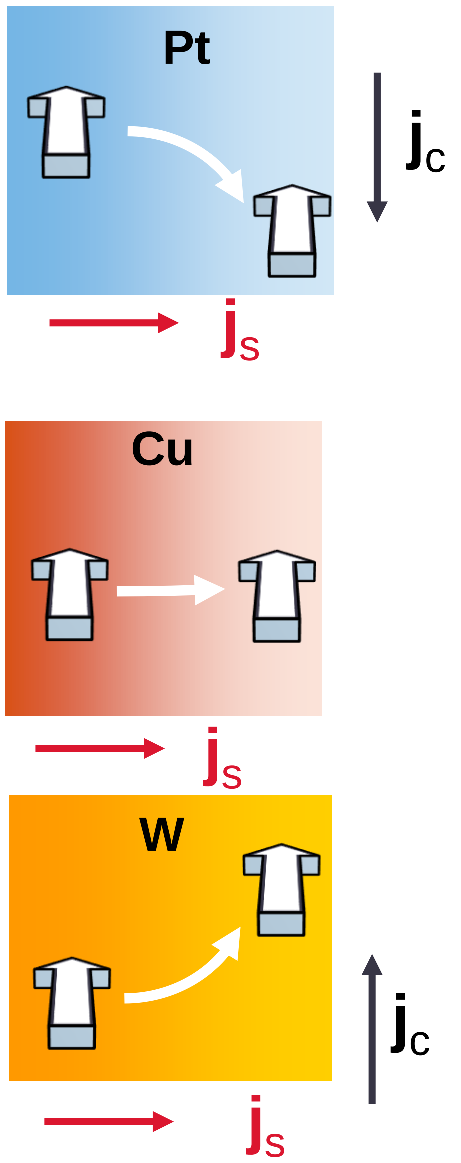
Final check: exchange Pt

TSS *et al.*, Nat. Phot. (2016)

THz inv. spin Hall effect

Change the paramagnet





Pt vs W:

- Strong opposite spin Hall angles

Saitoh *et al.*, APL (2006)

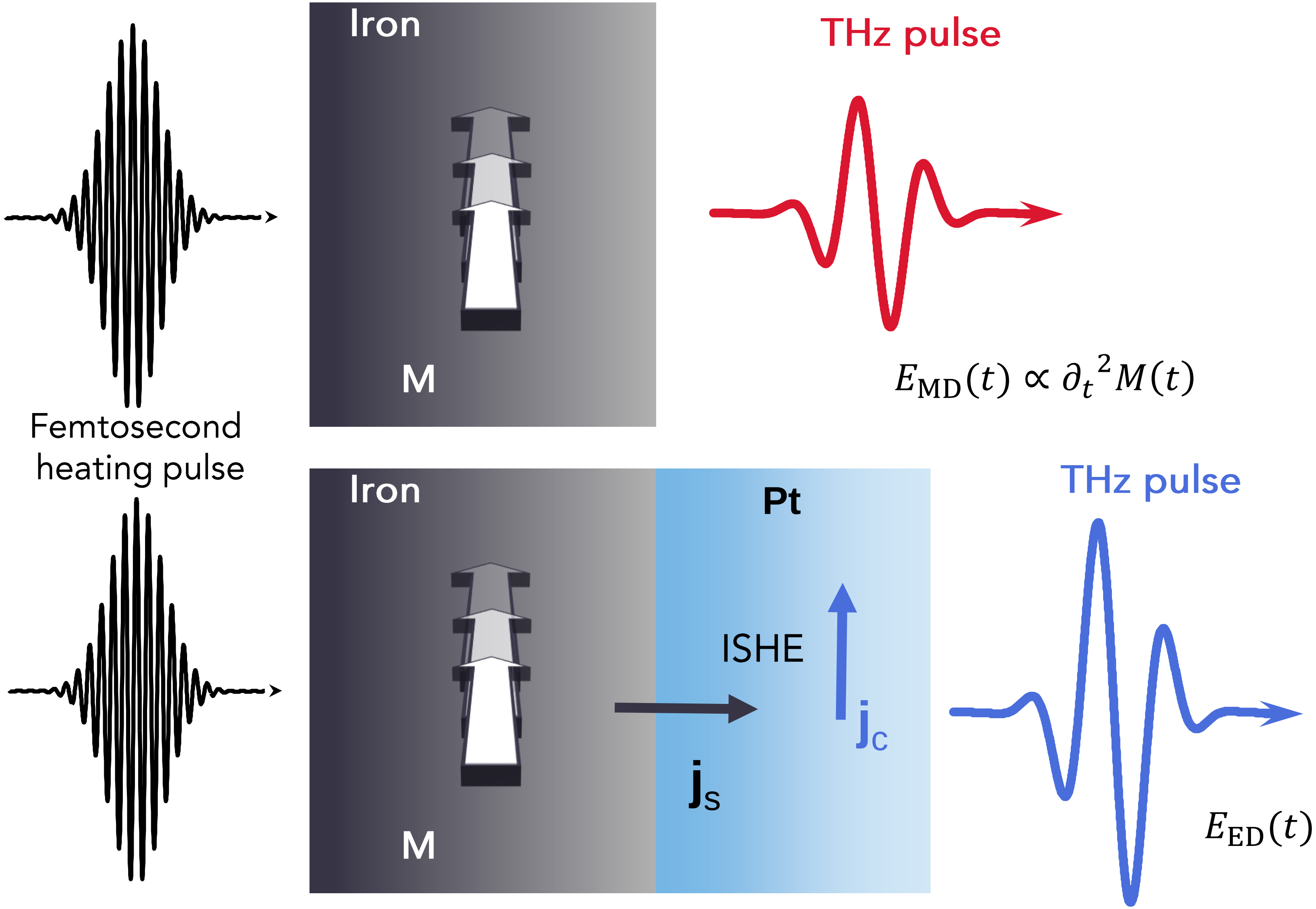
<u>Probe diverse materials:</u>
TIs, Weyl semimetals, superconductors, TMDCs etc.

Recent review: TSS *et al.*, APL 120 (2022)

Detailed understanding?

TSS *et al.*, Nat. Phot. (2016)

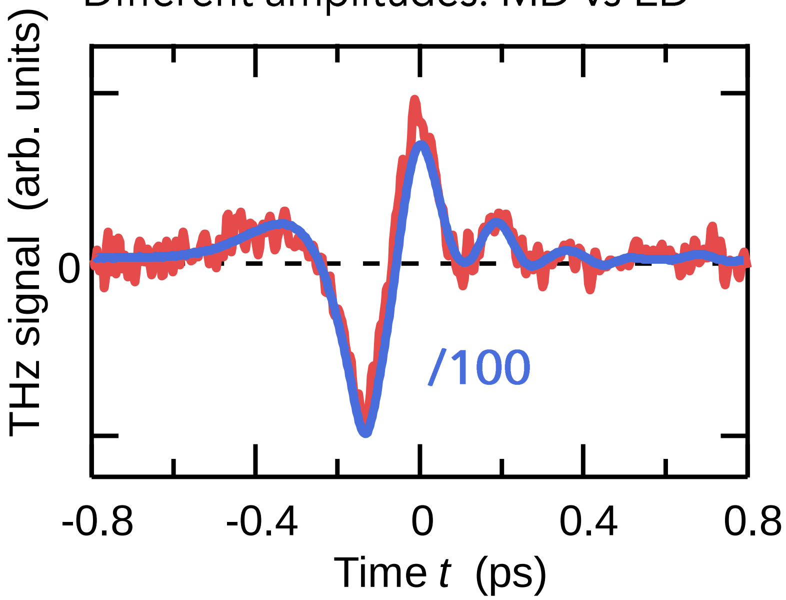
Single vs bilayers



$E_{\mathrm{MD}}(t) \propto \partial_t{}^2 M(t)$

$E_{\mathrm{ED}}(t) \propto \partial_t j_c(t) \propto \partial_t j_s(t)$

Different amplitudes: MD vs ED



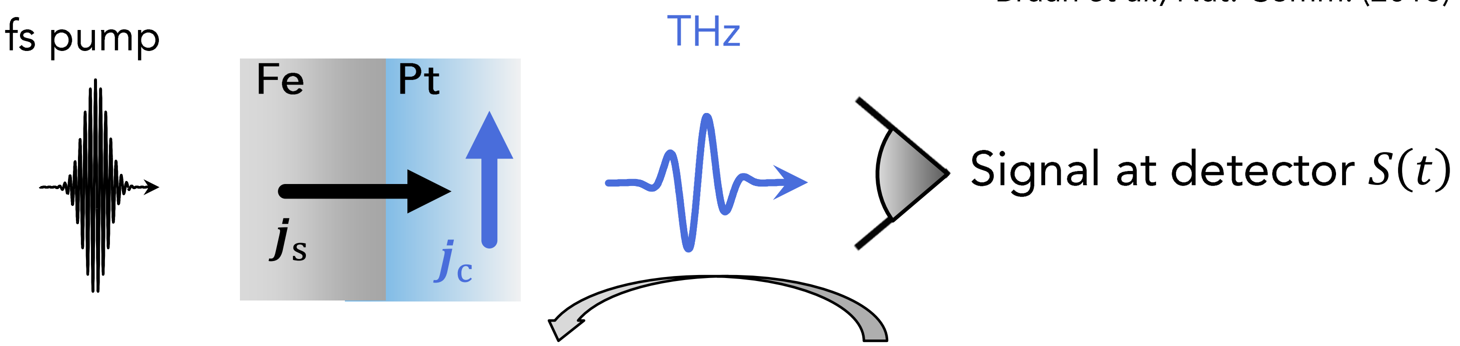
Same dynamics: transport is a weak perturbation

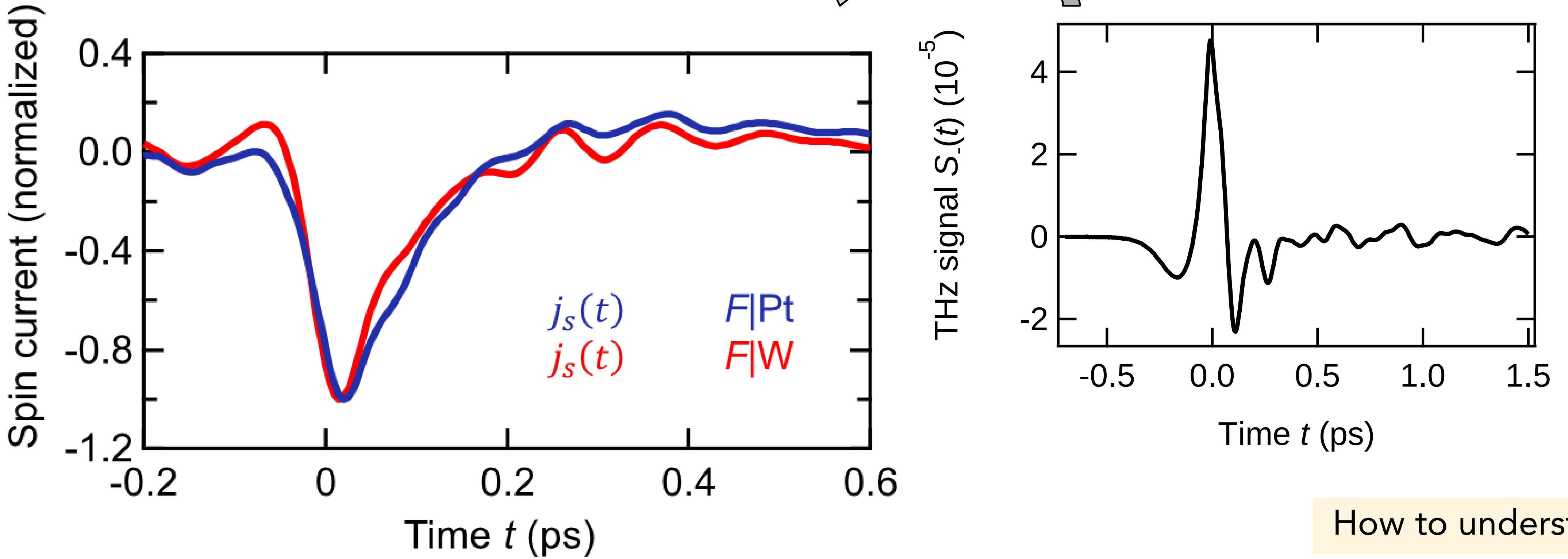
What determines the dynamics?

R. Rouzegar,.., TSS *et al.*, PRB (2022)

Inversion procedure:

Braun *et al.*, Nat. Comm. (2016)

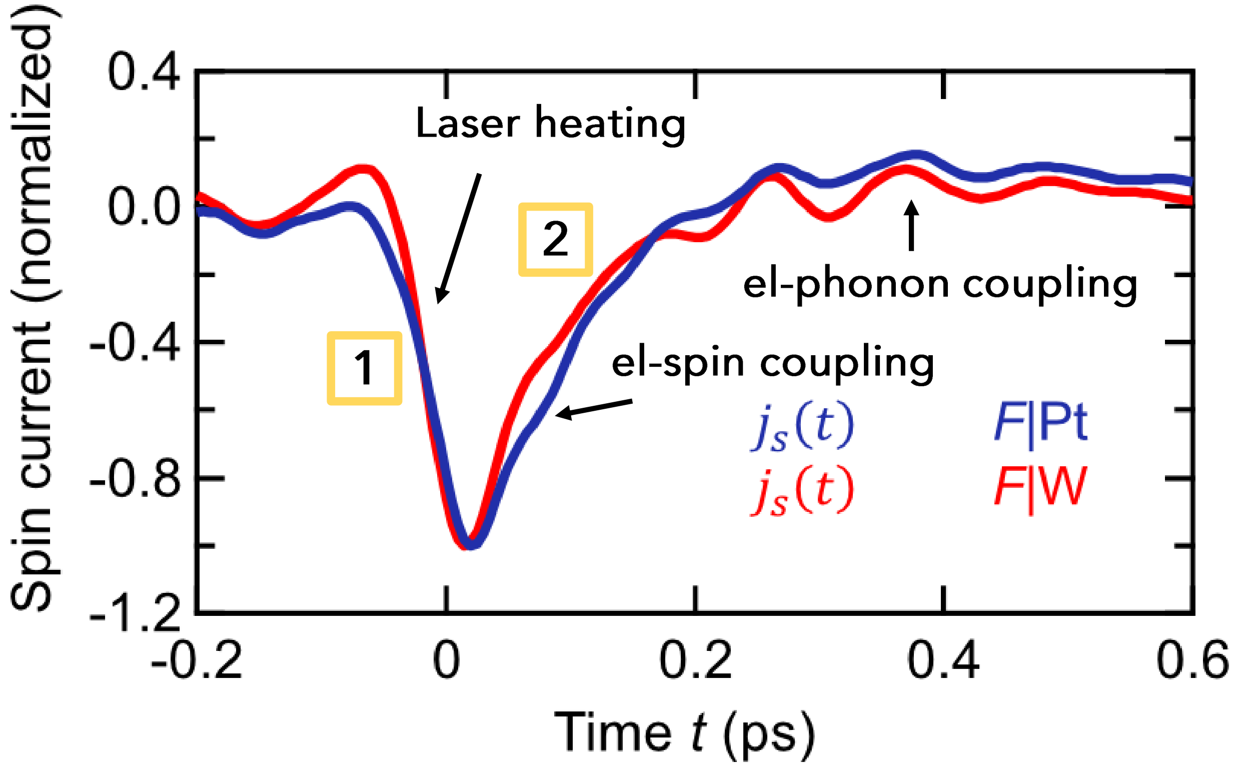


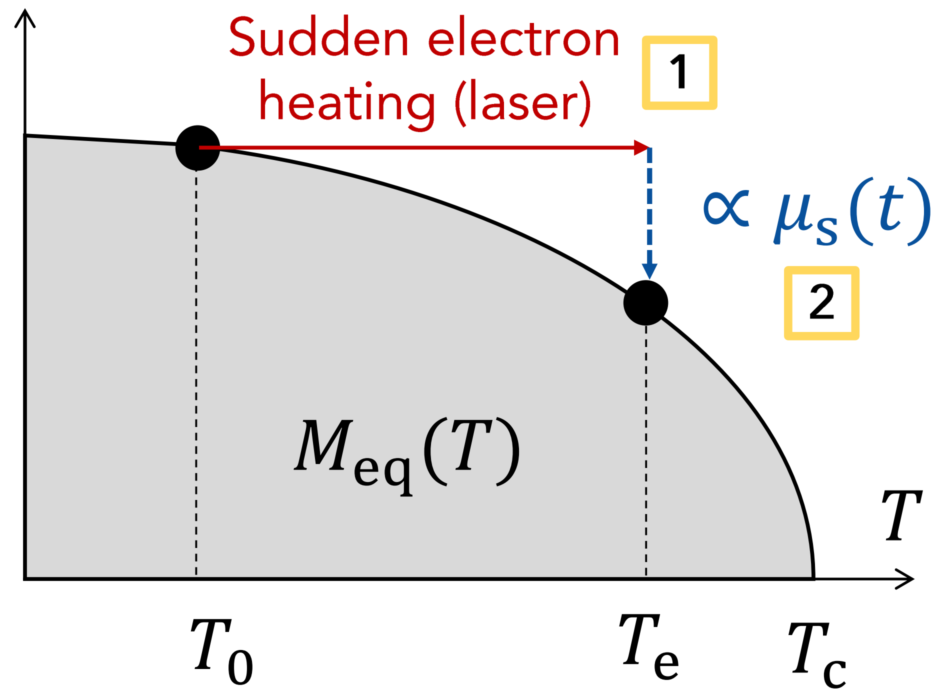


How to understand?

R. Rouzegar,.., TSS *et al.*, PRB (2022)

THz spin current





μ_s is an excess magnetization "Spin voltage"

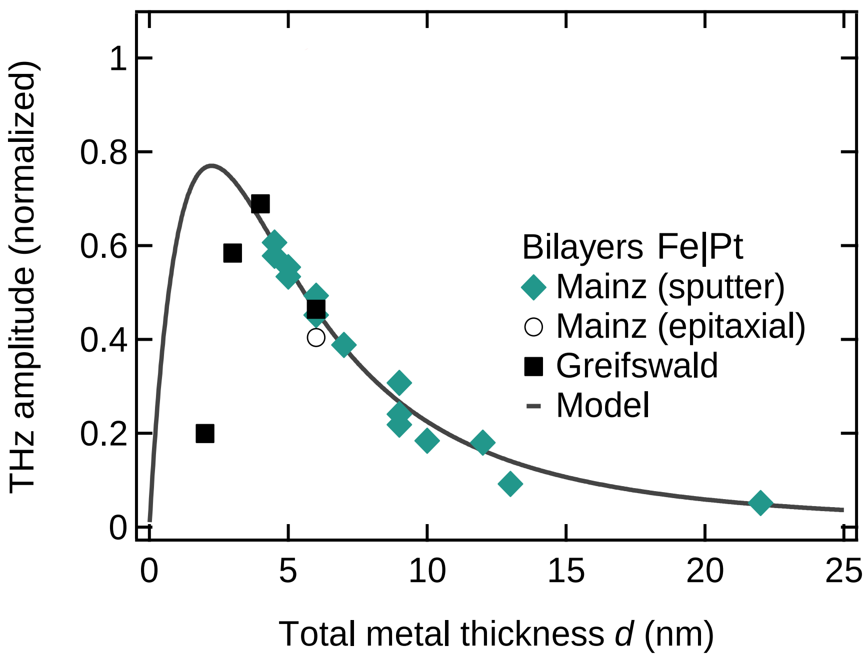
Relaxes locally and via transport

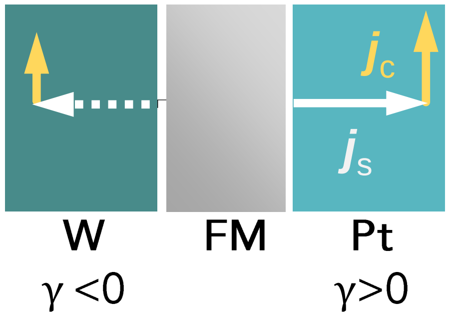
Nice but what good for?
Optimization?

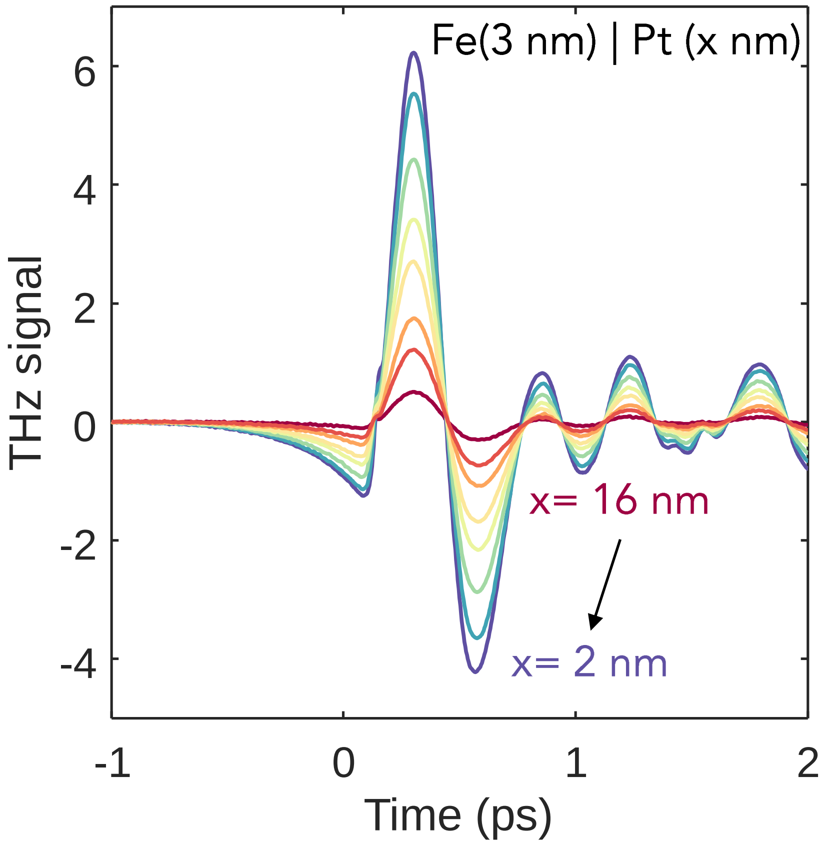
Choi, Cahill *et al.*, Nat. Comm. (2014)
Mueller, Rethfeld, PRB (2014).
R. Rouzegar,.., TSS *et al.*, PRB (2022)
Gupta, Oppeneer *et al.*, PRB (2023)

Optimized spintronic THz source

Thickness optimization

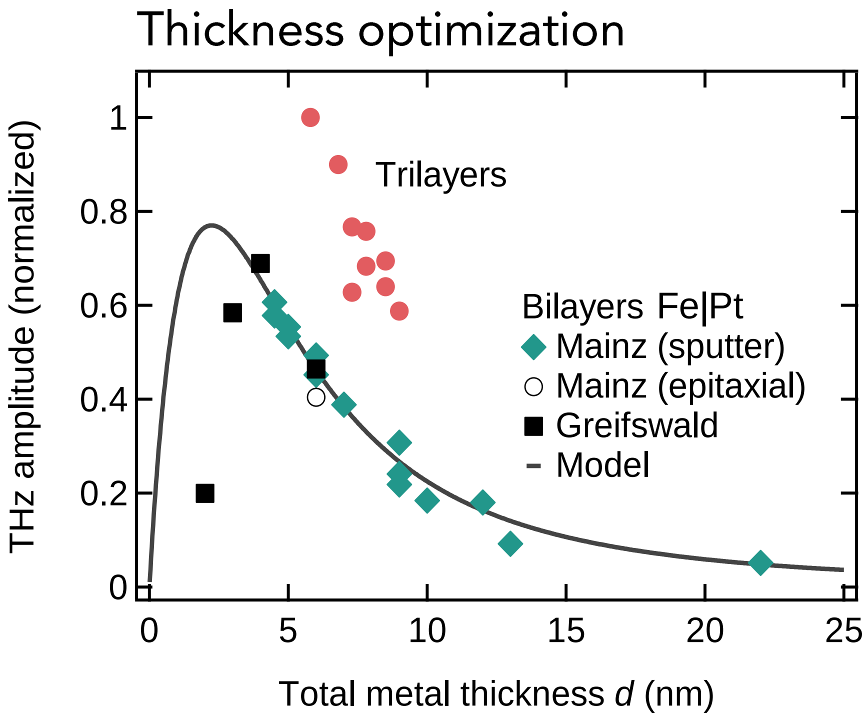


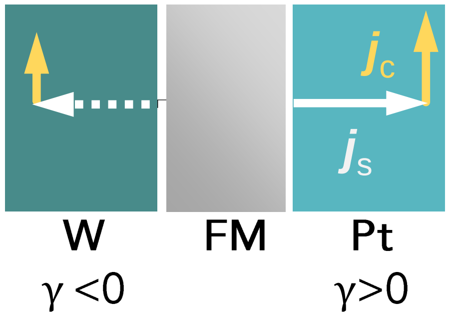


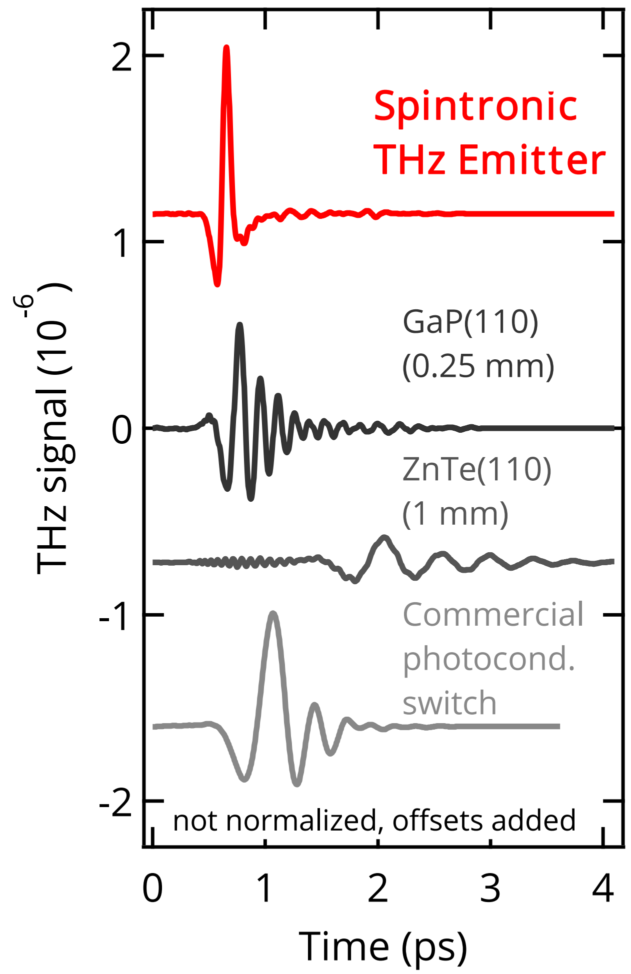


TSS *et al.*, Nat. Photon. (2016)

Optimized spintronic THz source



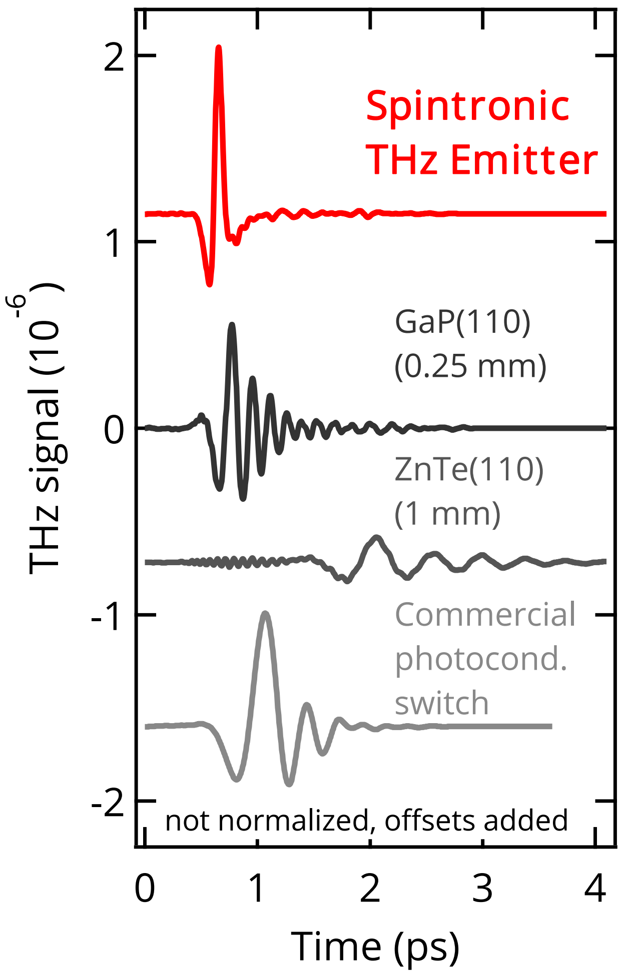




Outperforming standard THz sources

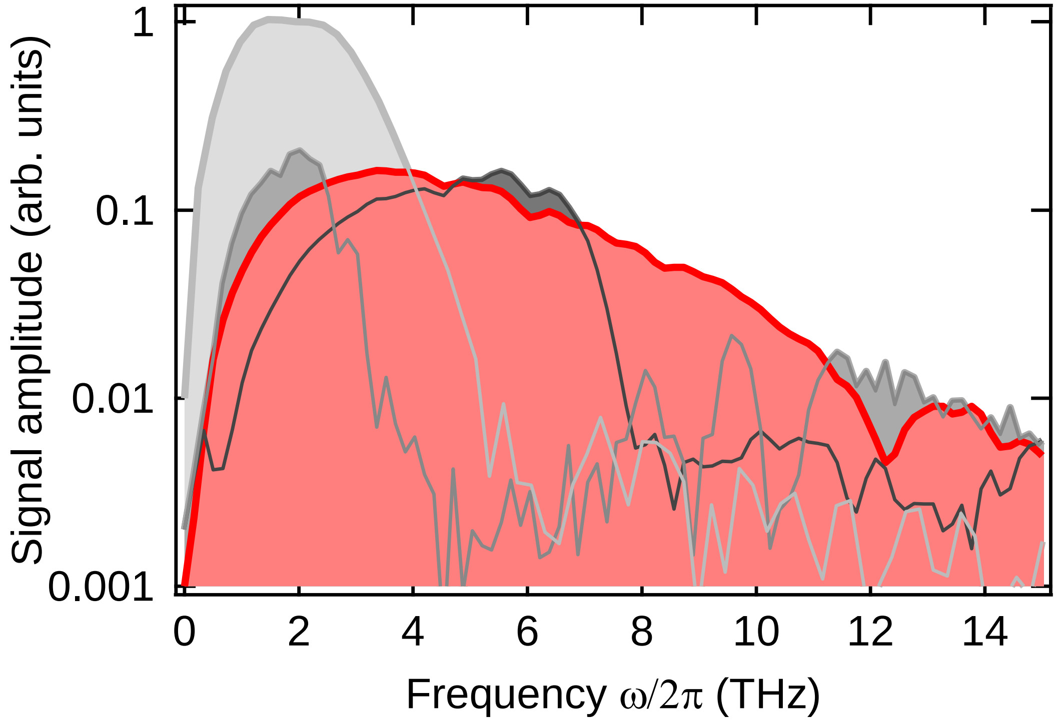
TSS *et al.*, Nat. Photon. (2016)

Optimized spintronic THz source



Fourier transform

Broadband: 1-40 THz



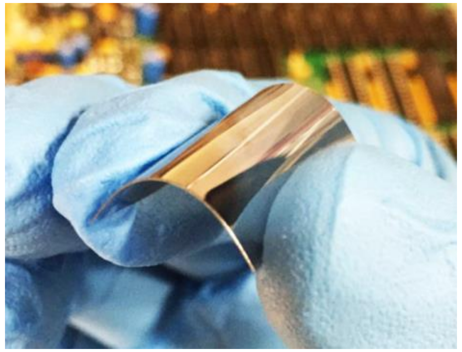
Applications?

TSS *et al.*, Nat. Photon. (2016)

Broadband spintronic THz emitter

Flexible/ curved substrates

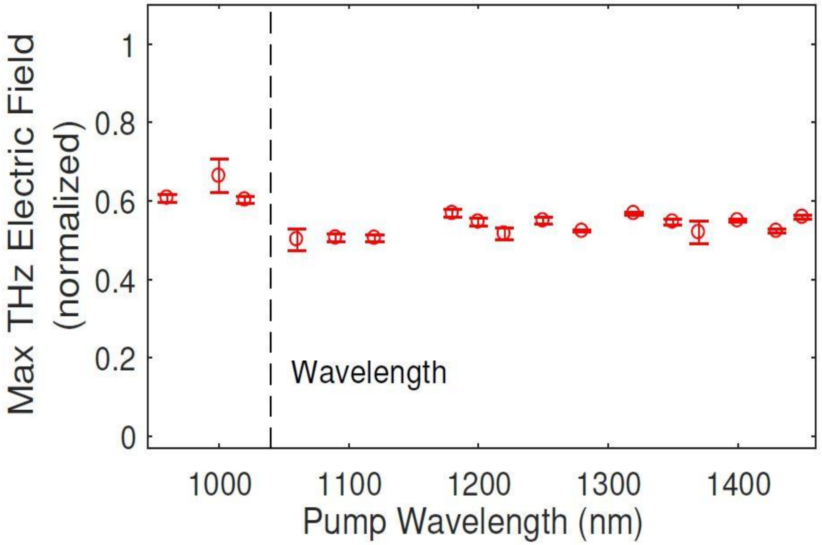
Wu, Yang *et al.*, Adv. Mat. (2016)



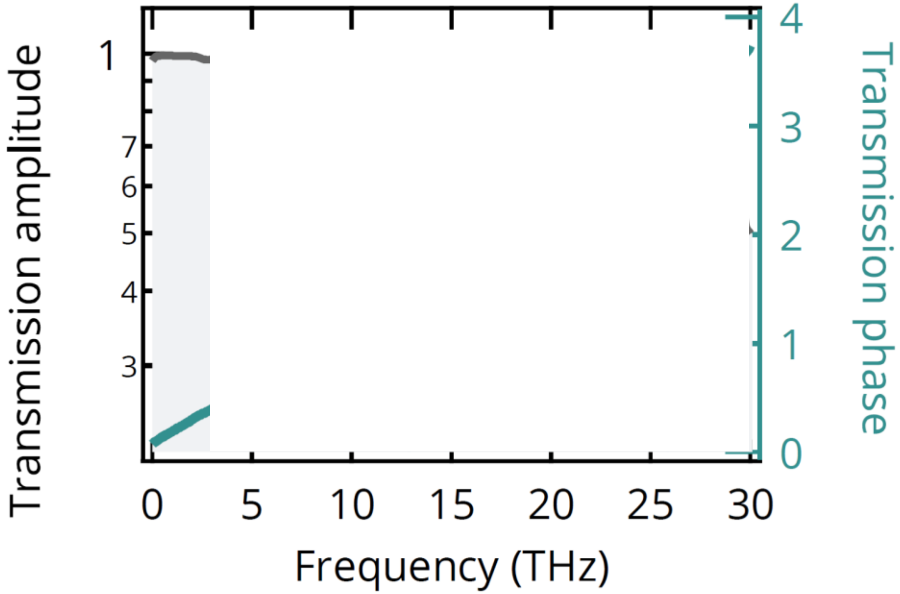
Insensitive to pump wavelength

Herapath, Hornett, TSS *et al.* APL (2019)

Papaioannou *et al.*, IEEE Trans. (2018)

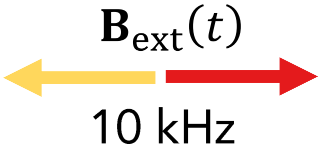


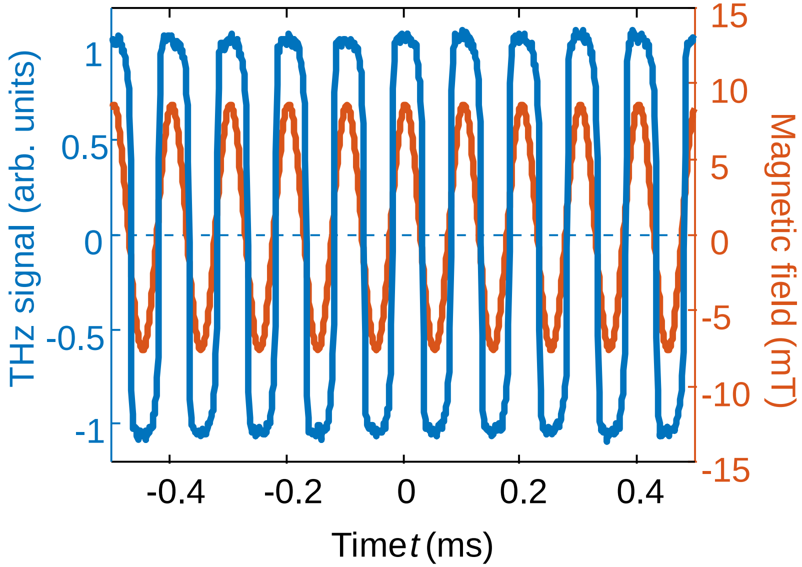
THz transmission (8 µm Teflon)



Easy polarization modulation

Gueckstock,.. TSS *et al.*, Optica (2021)

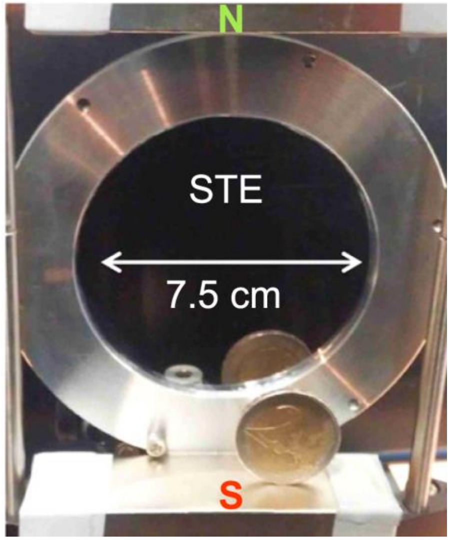


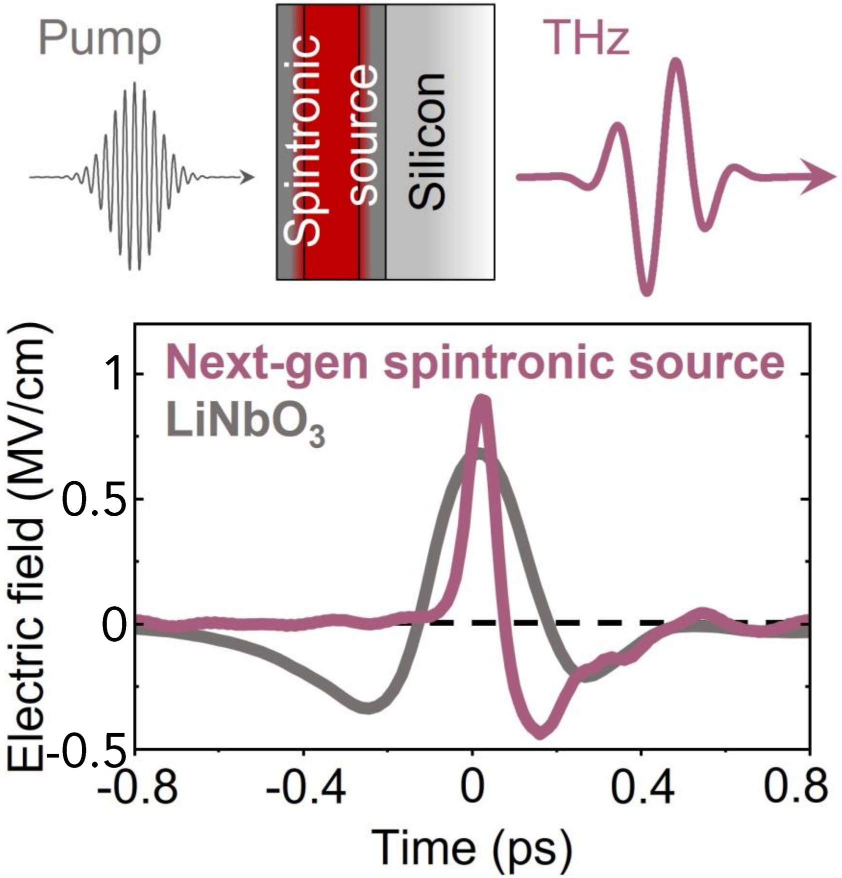


Broadband spintronic THz emitter

Upscaling yields high THz field strength (1 MV/cm)

TSS, *et al.*, APL (2017)





Peak currents in metals (10^6 S/m): 10^{10} A/cm^2

Enables nonlinear THz spectroscopy

TeraSpinTec

Ultrabroadband Spintronic THz emitters

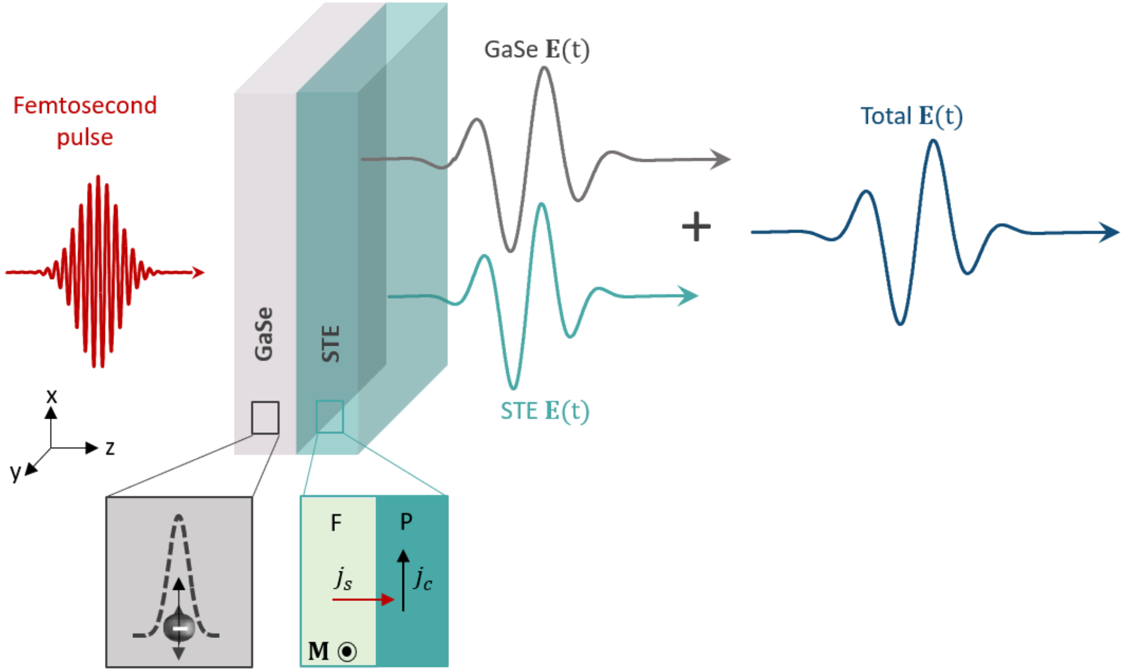
teraspintec.com

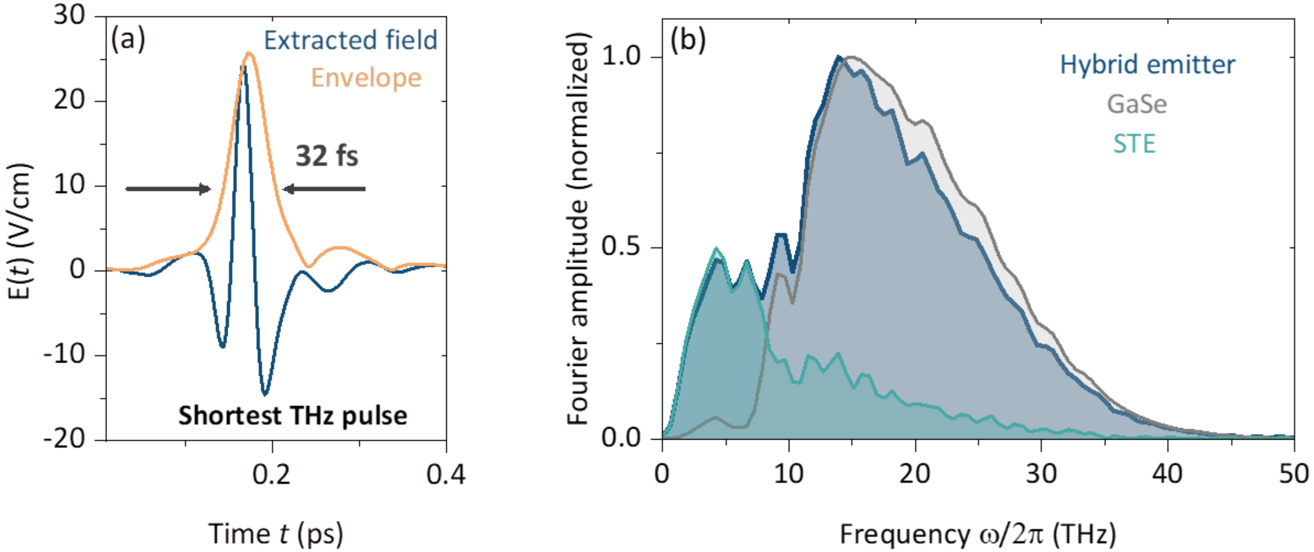
2021: Commercially available spintronic THz emitters

Rouzegar,..TSS, *et al.*, PRAppl (2023)

Hybrid emitter

Alostaz, TSS, *et al.*, arXiv (2023): arXiv:2310.12012



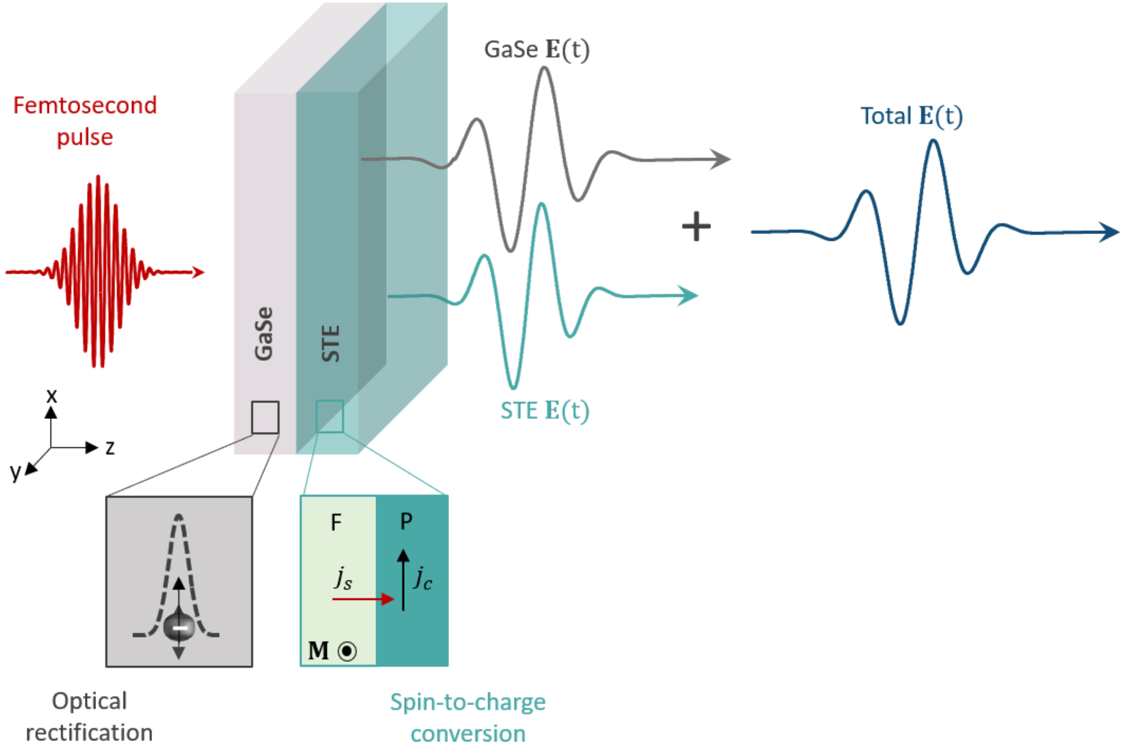


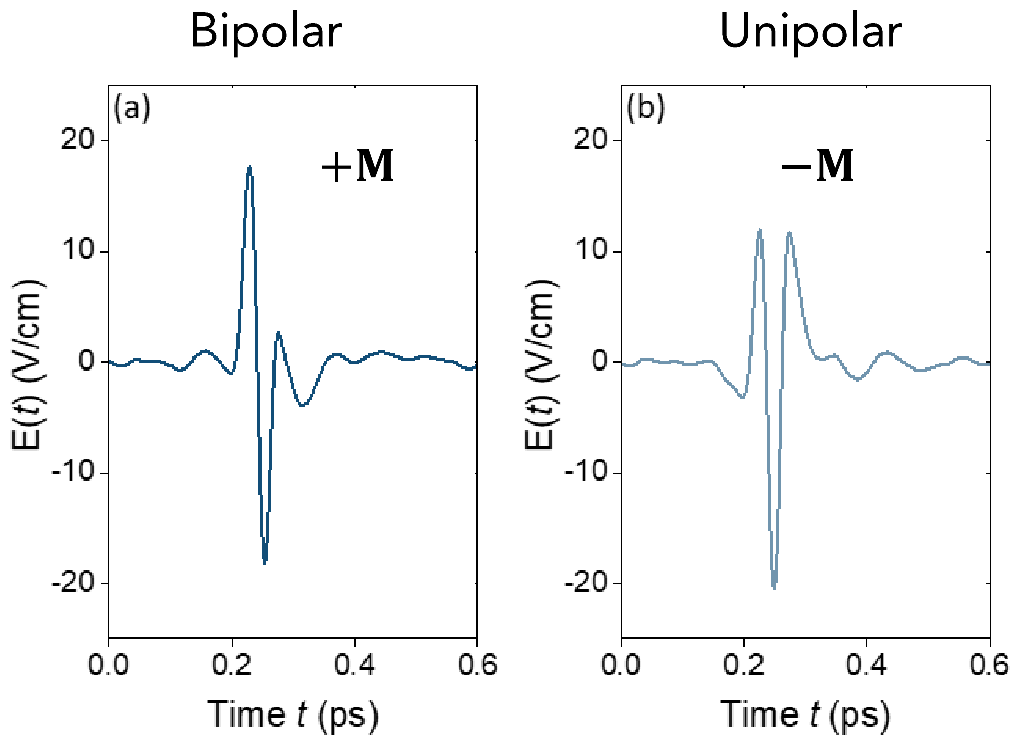
Combining advantages of STE and GaSe to yield optimized THz (probe) pulses

Towards the ideal broadband THz emitter

Hybrid emitter

Alostaz, TSS, *et al.*, arXiv (2023): arXiv:2310.12012

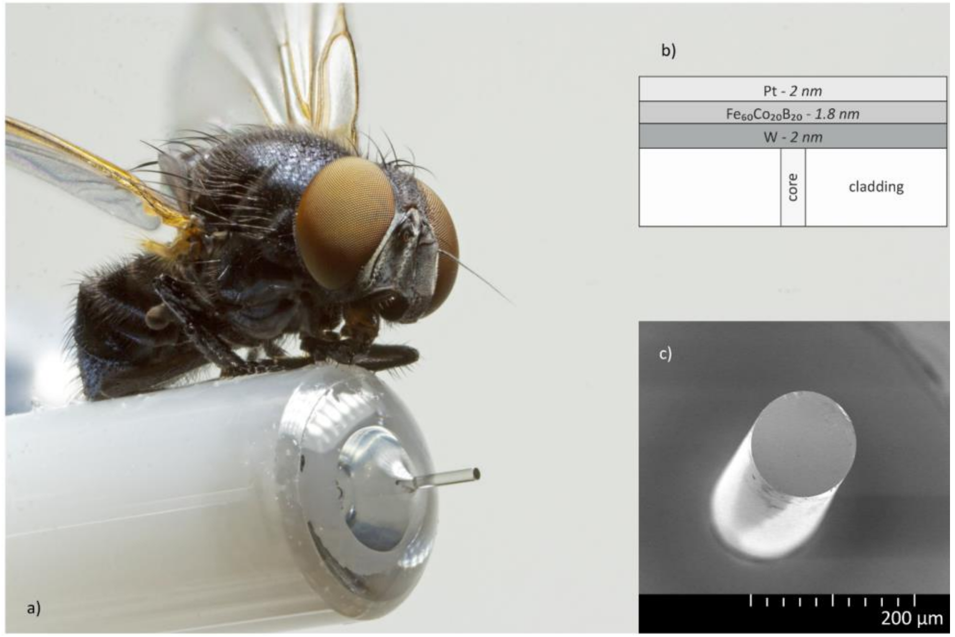




Tune THz pulse shape by magnetic field

Going small

THz microscopy: Fiber-coated spintronic emitter



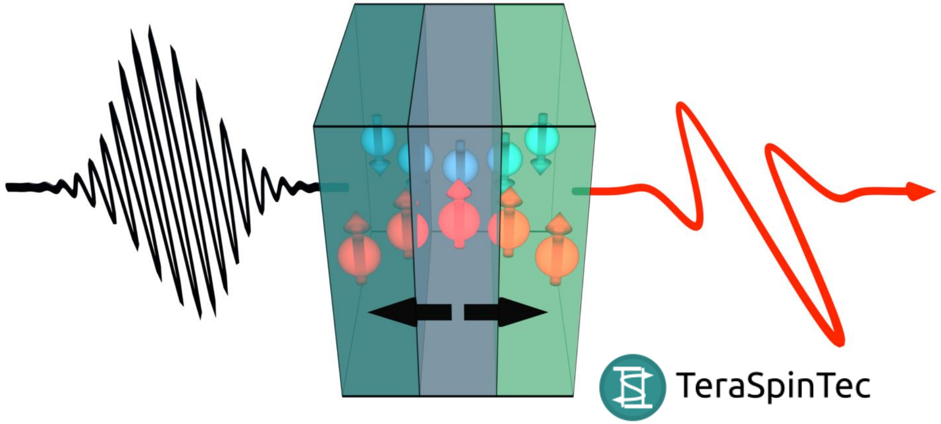
Paries,TSS,von Freymann *et al.*, Opt. Expr. (2023)

S conclusions

New insights

- Inv. spin Hall effect operative at THz rates
- Ultrafast demagnetization and terahertz spin transport driven by the same force: Spin voltage
- Optimization: efficient spintronic THz sources

Spintronic Terahertz Emitters



Recent review: TSS et al., APL 120 (2022)

How about detection?

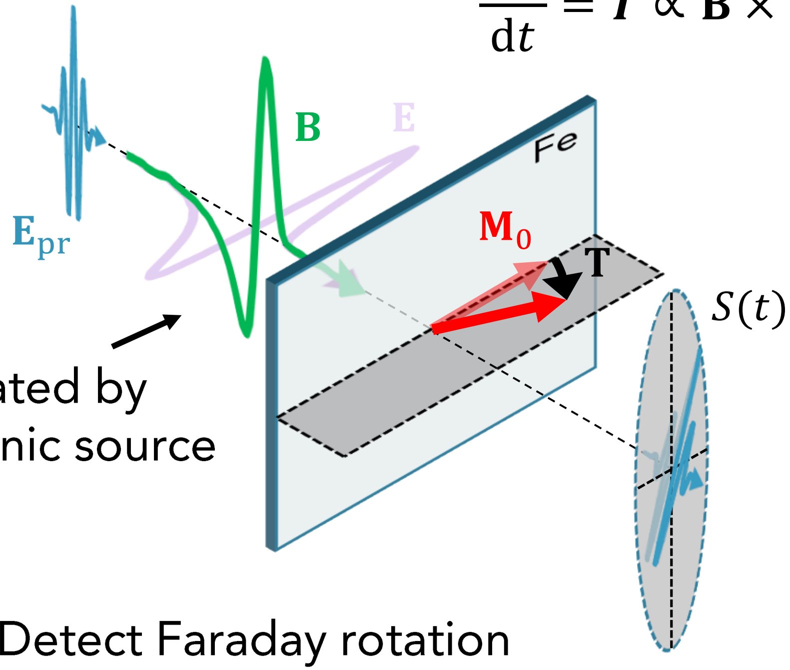
TSS *et al.*, Nat. Photon. 10 (2016)

R. Rouzegar,.., TSS *et al.*, PRB (2023)

Broadband spintronic THz detection

Zeeman torque

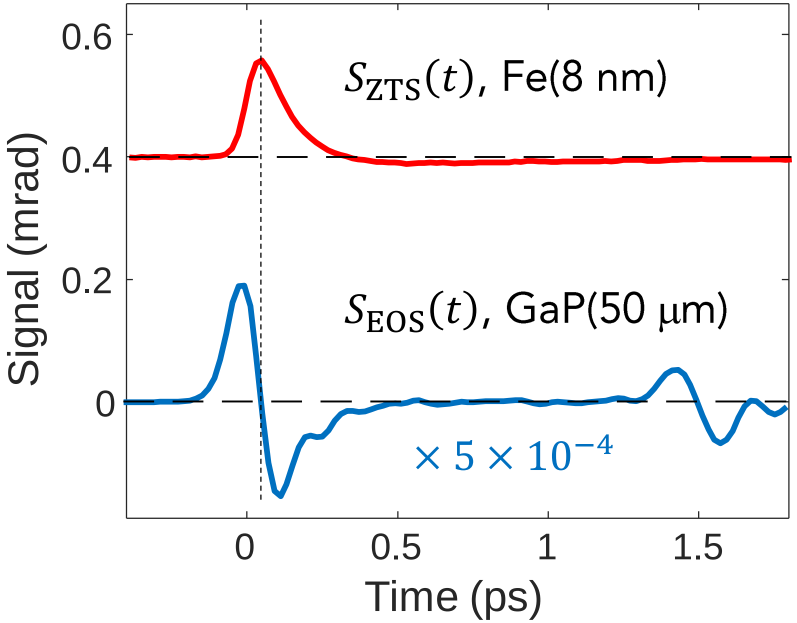
$$\frac{\mathrm{d}\mathbf{M}}{\mathrm{d}t} = \boldsymbol{T} \propto \mathbf{B} \times \mathbf{M_0}$$



Generated by spintronic source

Detect Faraday rotation

$$S(t) \propto \Delta\mathbf{M} \propto \int \mathrm{d}t\ \ \mathbf{B} \times \mathbf{M_0}$$

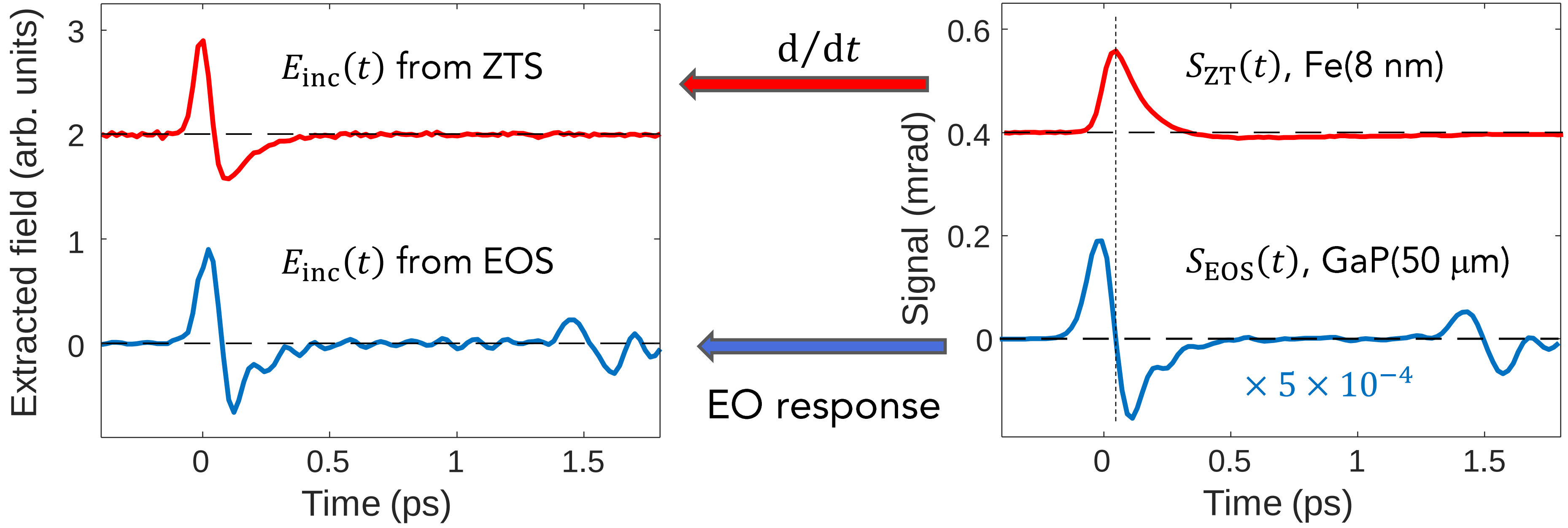


Extract THz fields

Chekhov,TSS *et al.*, PR Appl. 20 (2023)

Vicario et al., *Nat. Photonics* (2013)
Bonetti et al., *PRL* (2016)
Shalaby et al., *PRB* (2018)

Zeeman torque



- Extracted fields agree well
- Cleaner Zeeman-torque signal, yet reduced amplitude

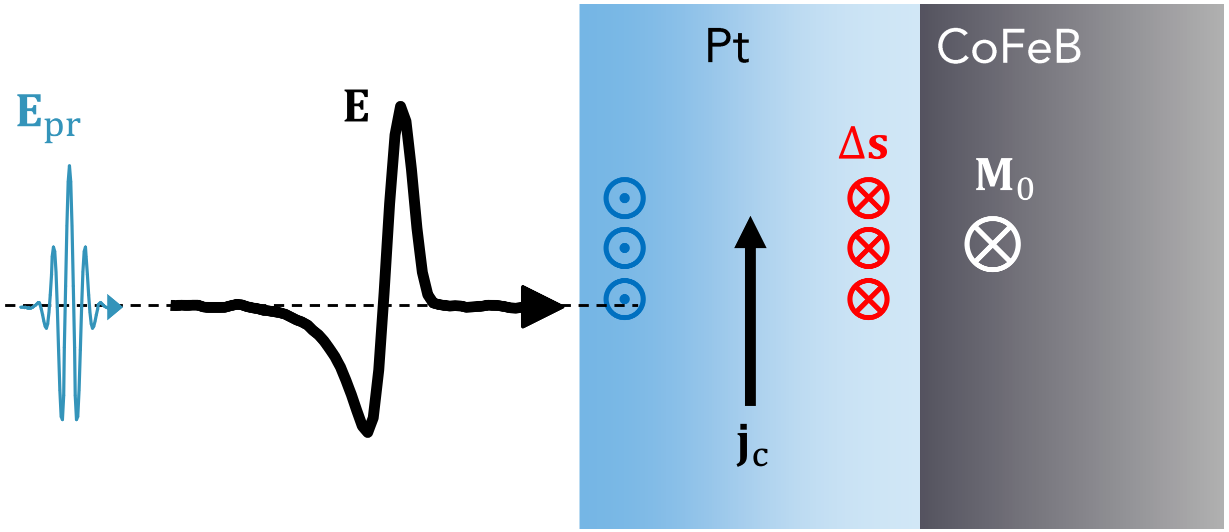
Chekhov,TSS *et al.*, PR Appl. 20 (2023)

Direct spintronic emitter inversion?

Vicario et al., *Nat. Photonics* (2013)
Bonetti et al., *PRL* (2016)
Shalaby et al., *PRB* (2018)

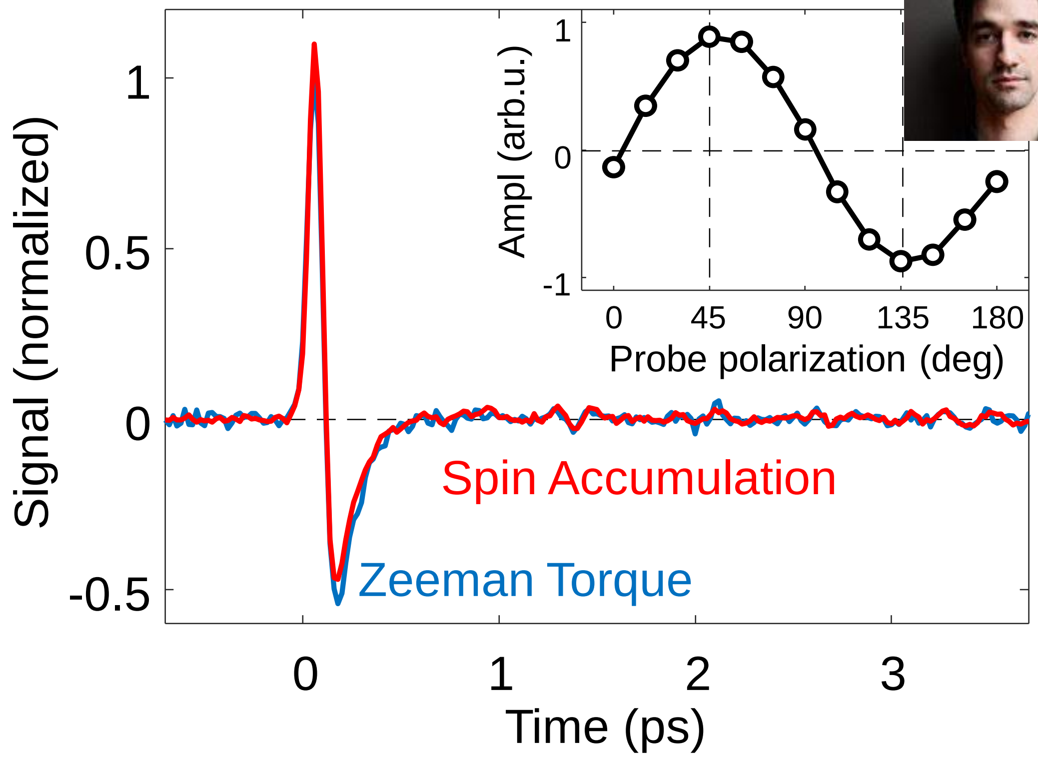
Broadband spintronic THz detection

Spin accumulation



Probe in-plane magnetization change via magnetic linear birefringence

$$S(t) \propto \Delta \boldsymbol{s} \cdot \mathbf{M}_0$$



Response is linear in $\mathbf{M}_0$ and THz field

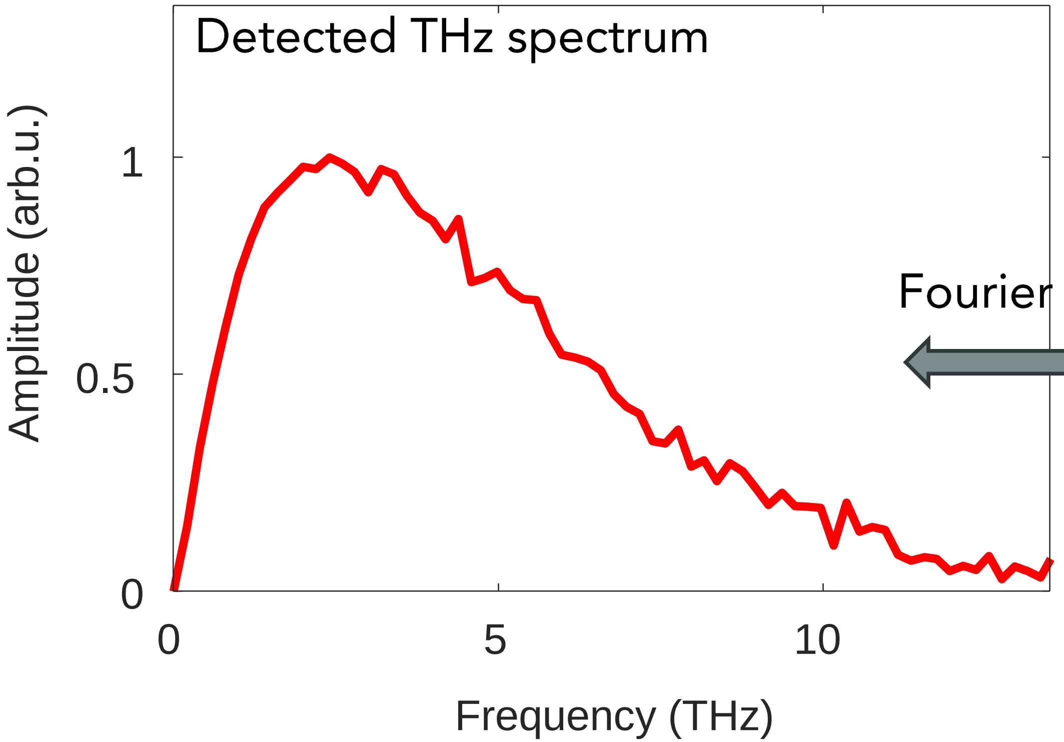
Direct access to strong-field THz transients

Salikhov, Kovalev *et al.*, Nat. Physics (2023)

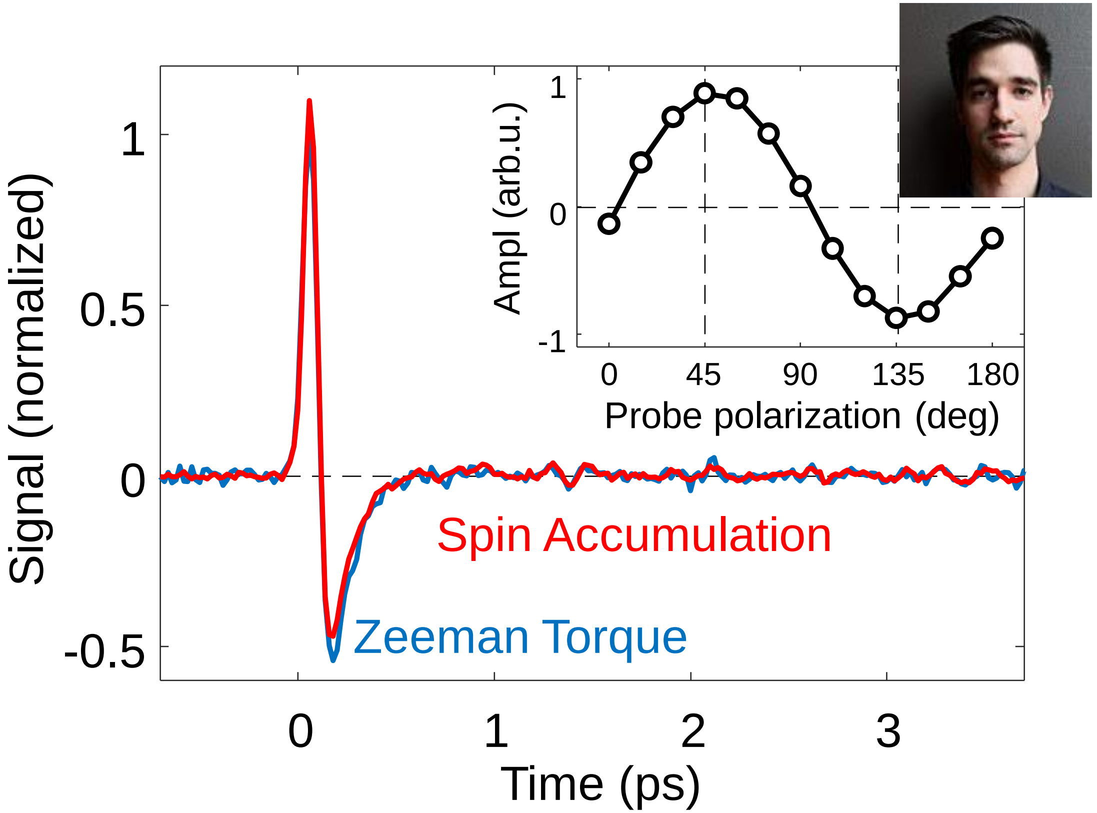
Chekhov, *et al.*, in preparation

Broadband spintronic THz detection

Spin accumulation



Fourier transform



Spintronic THz detection
bandwidth 1-12 THz

Salikhov, Kovalev *et al.*, Nat. Physics (2023)

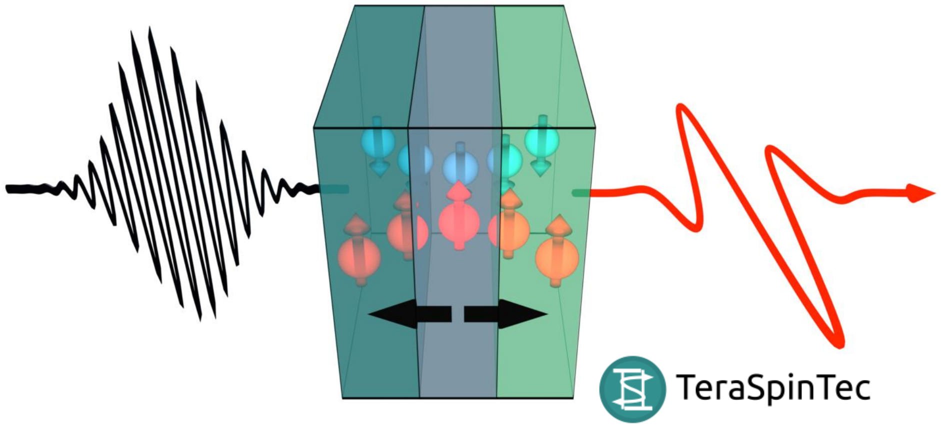
Chekhov, *et al.*, in preparation

S conclusions

New insights

- Inv. spin Hall effect operative at THz rates
- Iron is an excellent terahertz *S*-current source
- Optimization: efficient spintronic THz sources
- Inversion allows for broadband spin-based detection

Spintronic Terahertz Emitters



Recent review: TSS et al., APL 120 (2022)

TSS *et al.*, Nat. Photon. 10 (2016)

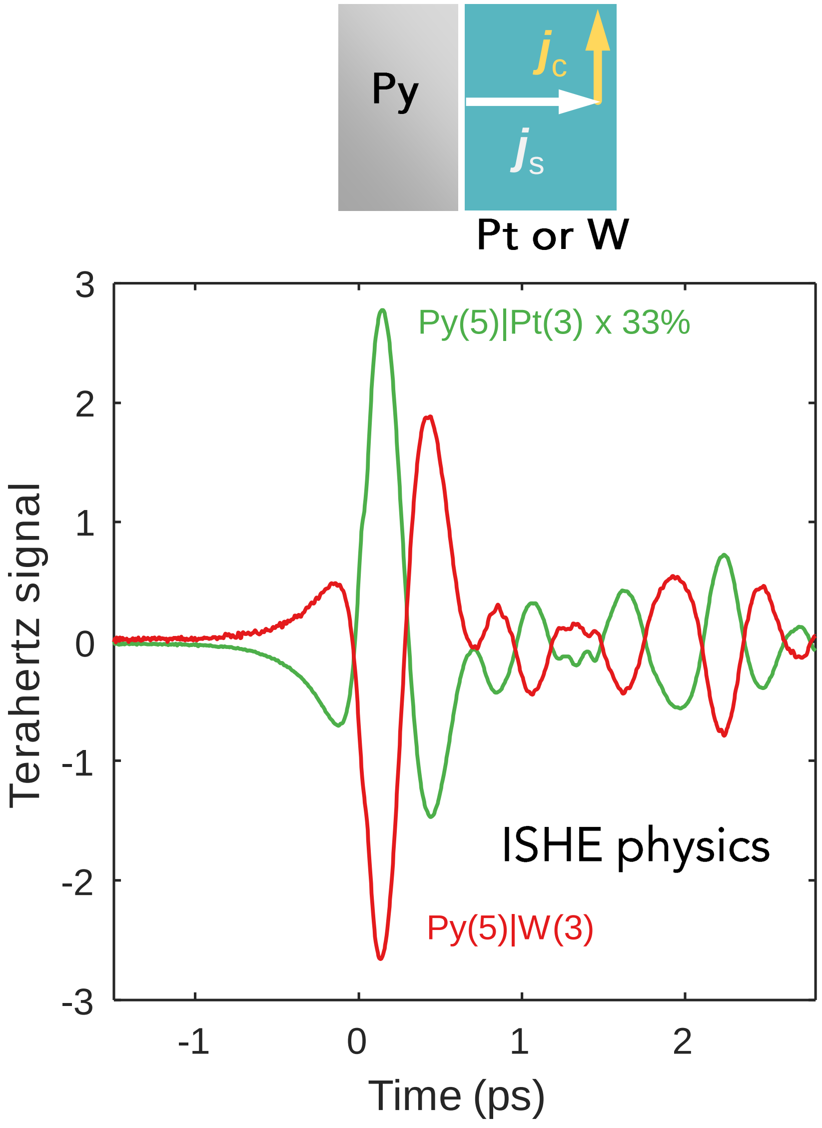
R. Rouzegar,.., TSS *et al.*, PRB (2023)

Chekhov, TSS *et al.*, PR Appl. 20 (2023)

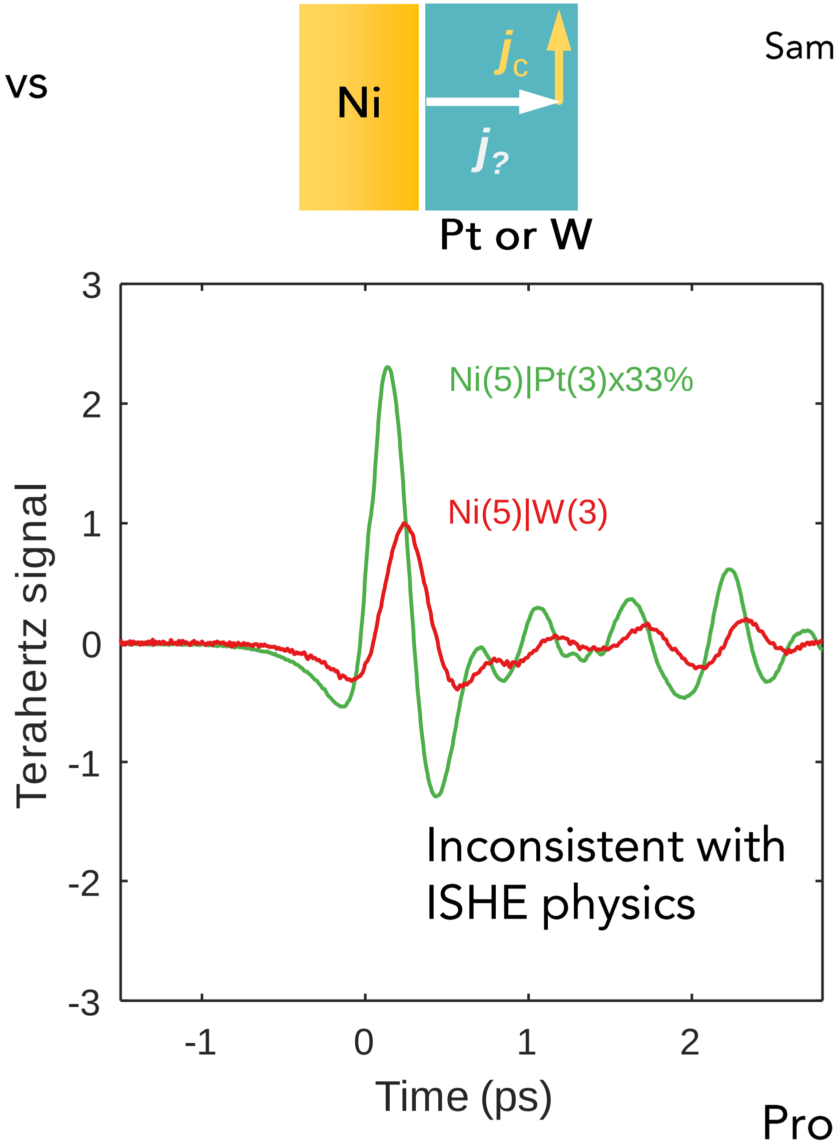
A puzzling experiment..

Surprising experiment

Samples: H. Hayashi, K. Ando



vs



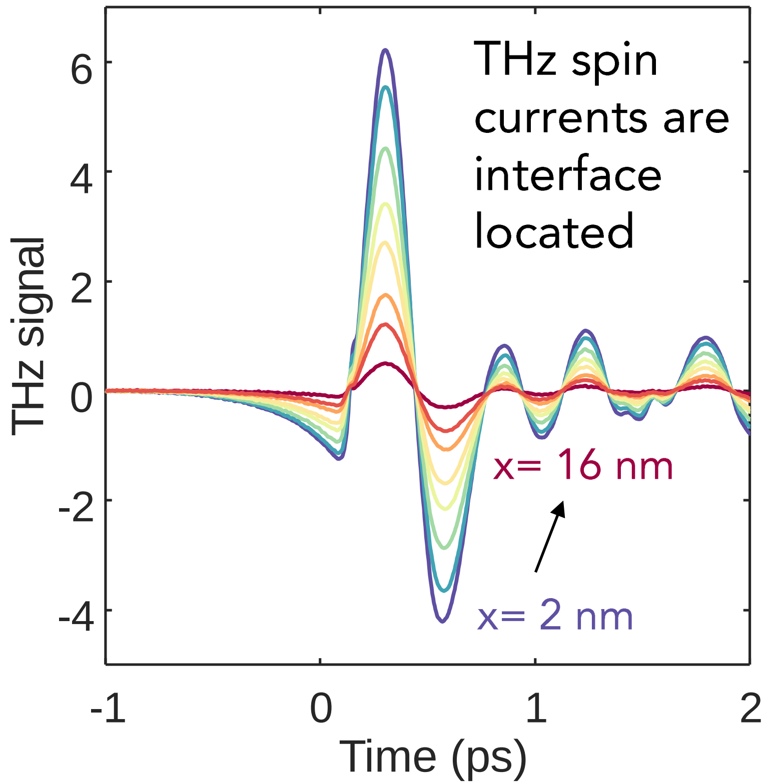
1. Sign change
2. Delayed
3. Slower

Propagation effect?

Py= $Ni_{80}Fe_{20}$
All signals are odd in sample magnetization

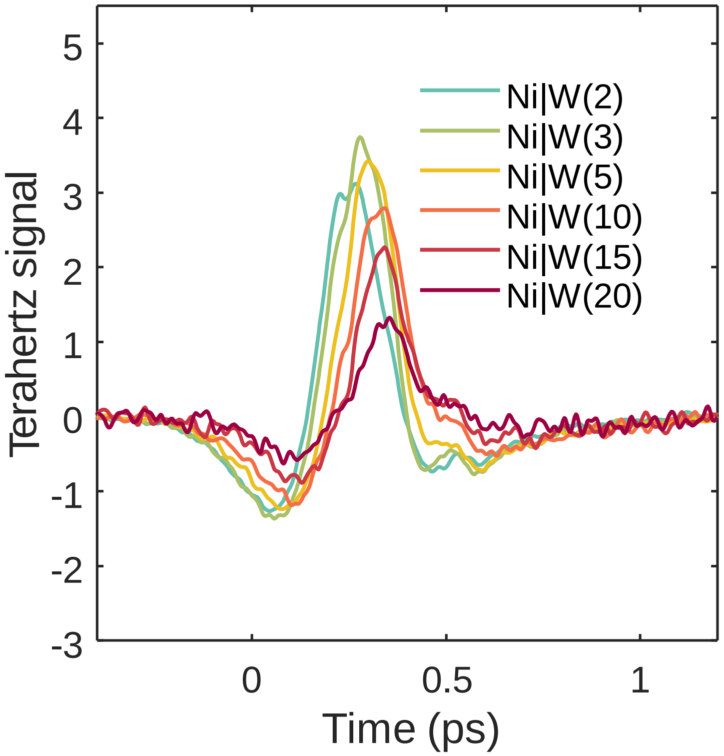
Impact of W Thickness

Fe | Pt (x nm)



Prototypical for spin currents

Ni | W (x nm)

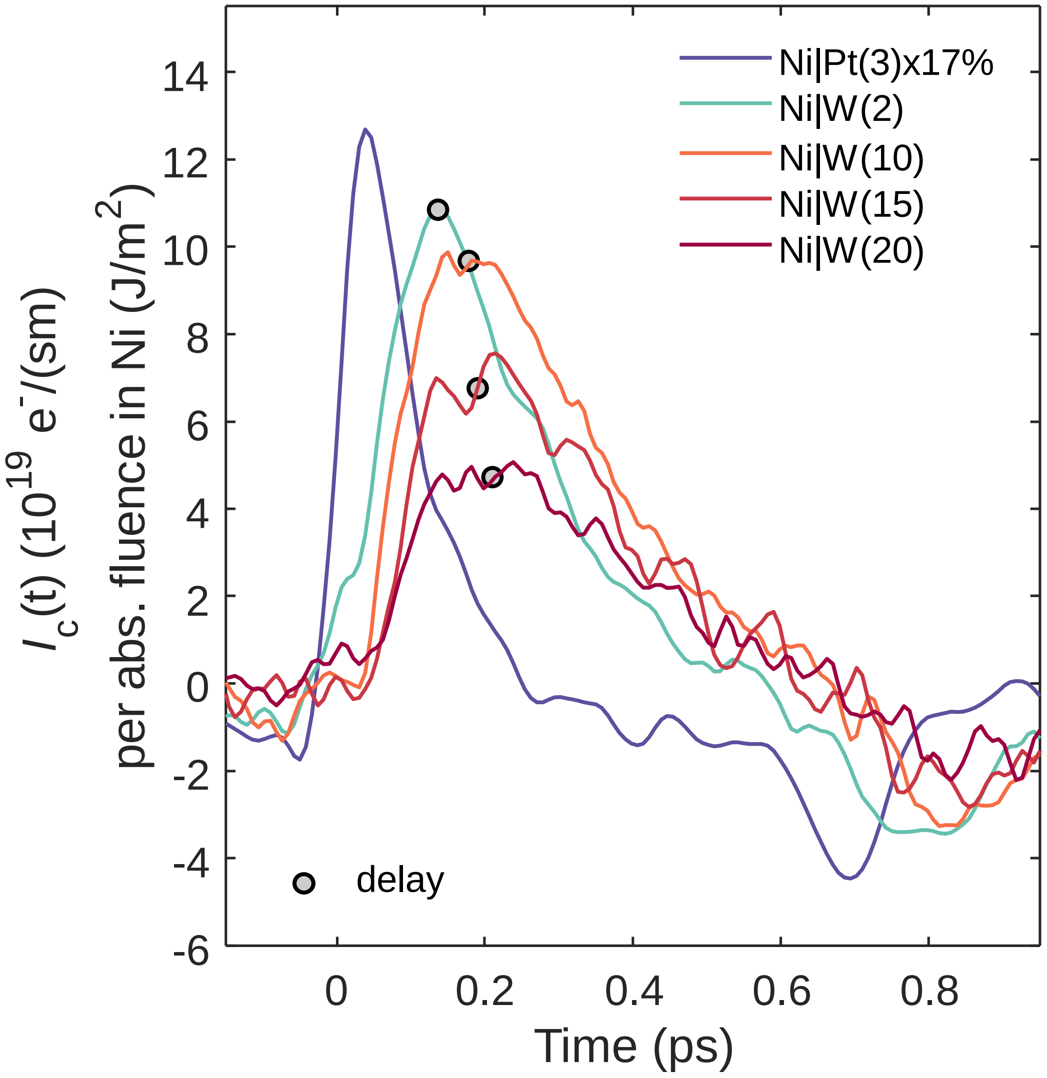


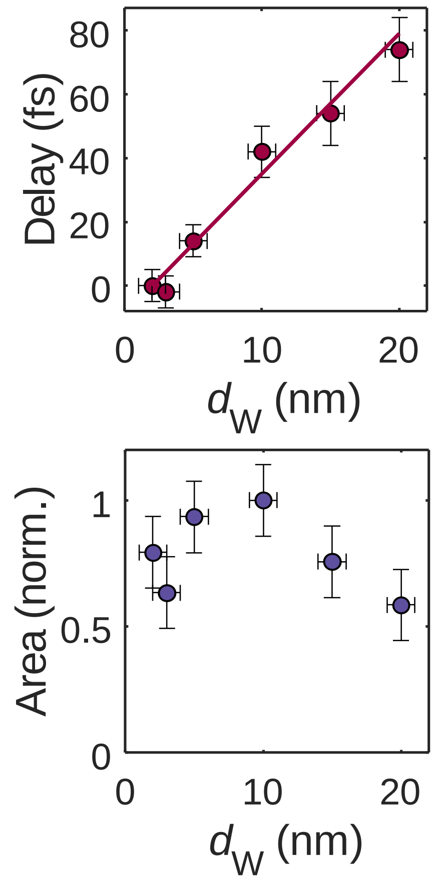
Totally untypical for spin currents

Look at THz currents vs time

TSS, *et al.*, Nature Nano. (2023)
Xu, Fert, Zhao *et al.* ArXiv (2022)
Wang, Jinag *et al.*, npj Quant. Mat. (2023)

Ni|W THz Currents





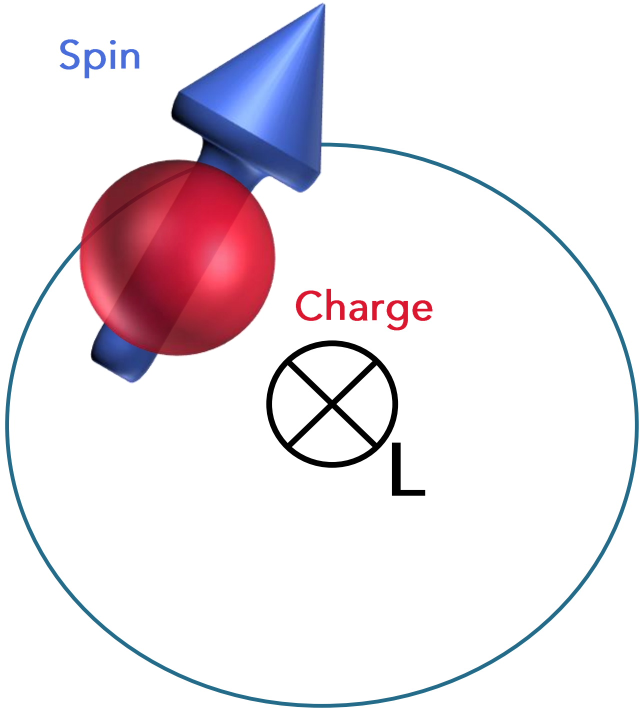
Suggests propagation over 10s of nm

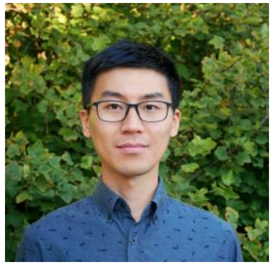
What can propagate that far?

Anomalous thickness dependence of SOTs in same samples:
Hayashi, Ando *et al.*, Comm. Phys. (2023)

TSS, *et al.*, Nature Nano. (2023)

Orbital angular momentum $\mathbf{L}$





Review: Go, et al. *Europhysics Letters* 135 (2021): 37001.

Orbitronics: data processing with $\mathbf{L}$

But $\mathbf{L}$ is quenched in a solid, right?

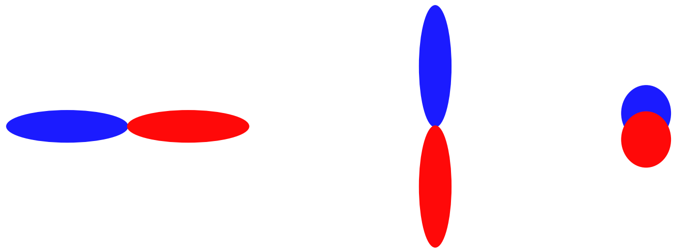
Orbital angular momentum L

P-orbital system

For given $\mathbf{k}$, eigenstate $|\Psi_{n\mathbf{k}}\rangle = e^{i\mathbf{k}\cdot\mathbf{r}}|u_{n\mathbf{k}}\rangle$

„p_x" „p_y" „p_z"

$|u_{n\mathbf{k}}\rangle \propto$



$\langle\mathbf{L}\rangle = 0$

Orbital quenching in the ground state imposed by *PT* symmetry

Superposition state

„$p_x + ip_y$"

$\langle\mathbf{L}\rangle \neq 0$

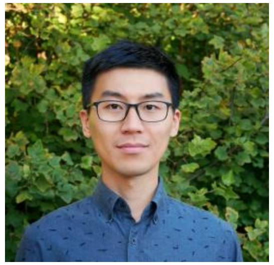
Orbital angular momentum can reappear in nonequilibrium

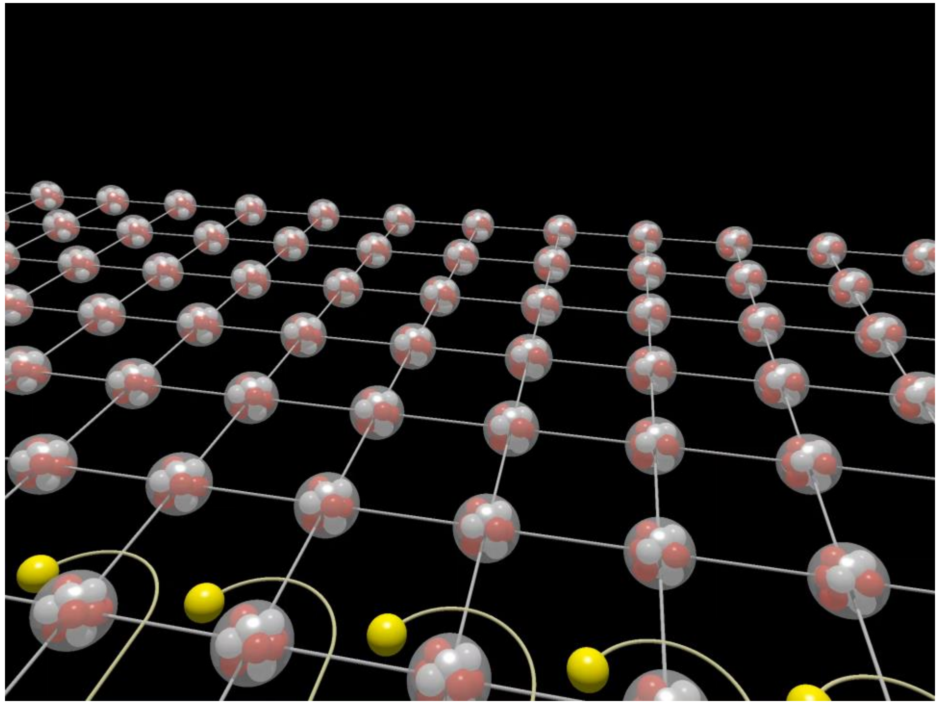
How to imagine an L current?

Go, H.-W. Lee et al. PRL 121 (2018)

Orbital current

Review: Go, et al. *Europhysics Letters* 135 (2021): 37001.

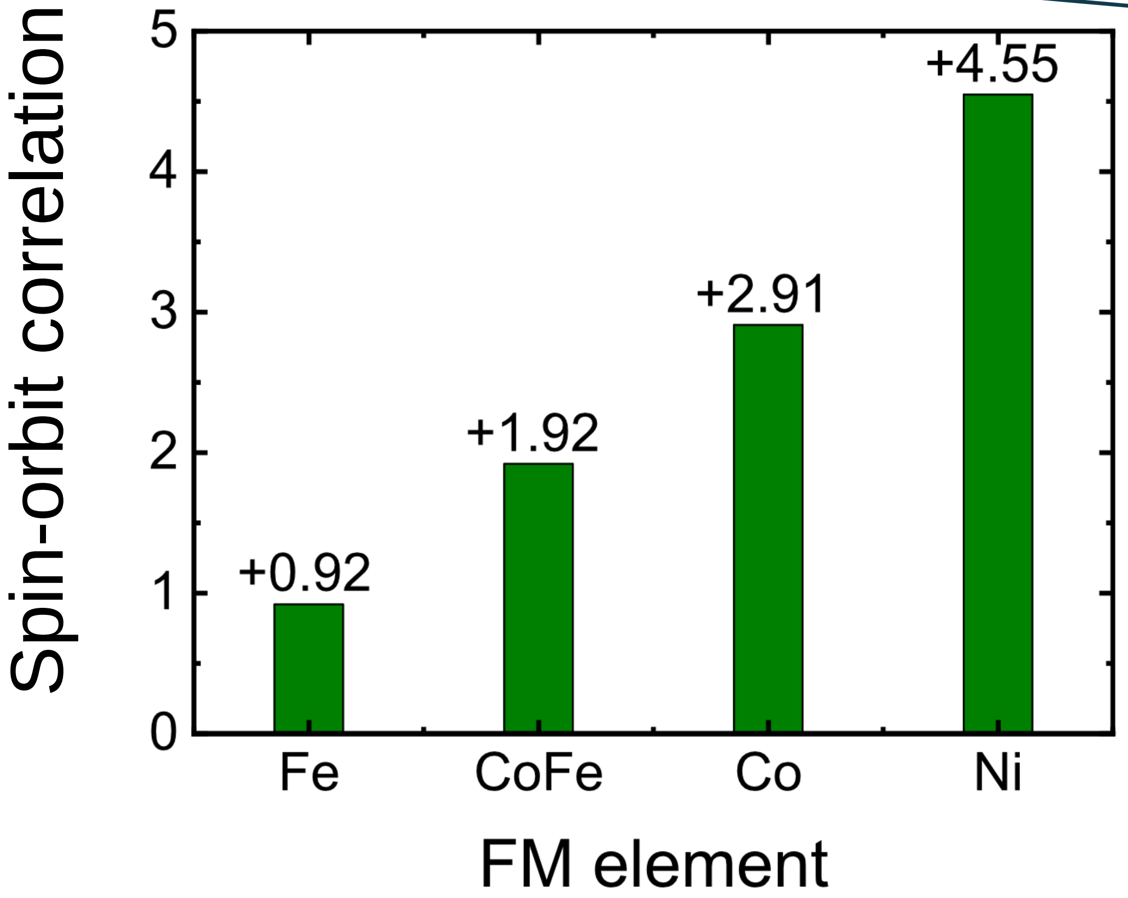


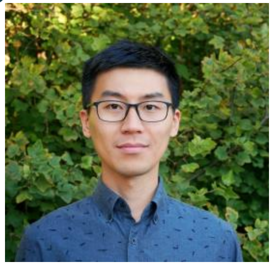


Dongwook, what are good *L* current sources?

Pezo, Manchon et al. PRB 106 (2022)

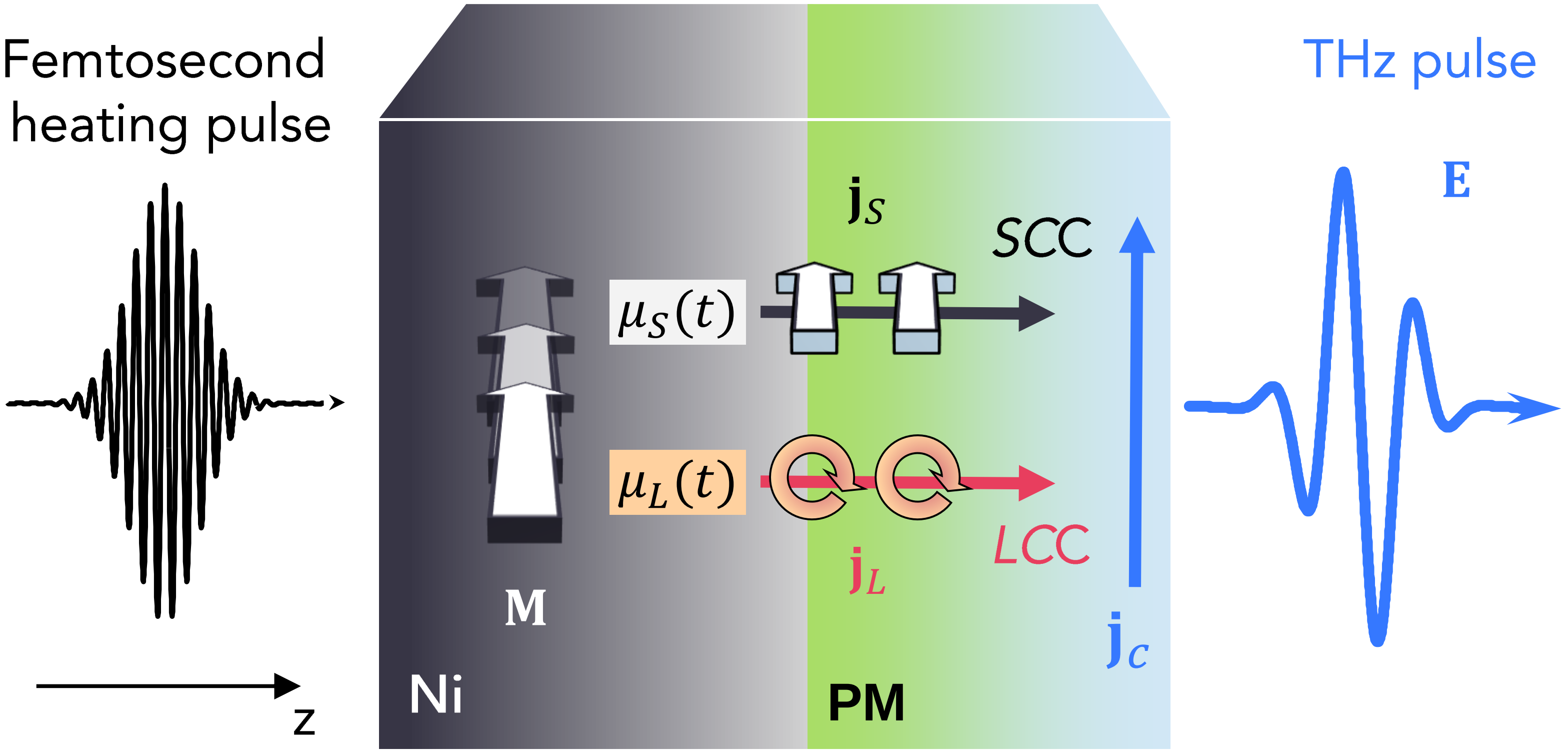
Lee, *et al.*, Nat. Commun. (2021)





Ni is a good *L* source

How to understand the THz experiment?



(i) Generate „driving force"

(ii) Propagation

(iii) Detection

Recent tr-XMCD studies on Ni:

Coupled by spin-orbit interaction

$$\mu_S(t) \propto \mu_L(t)$$

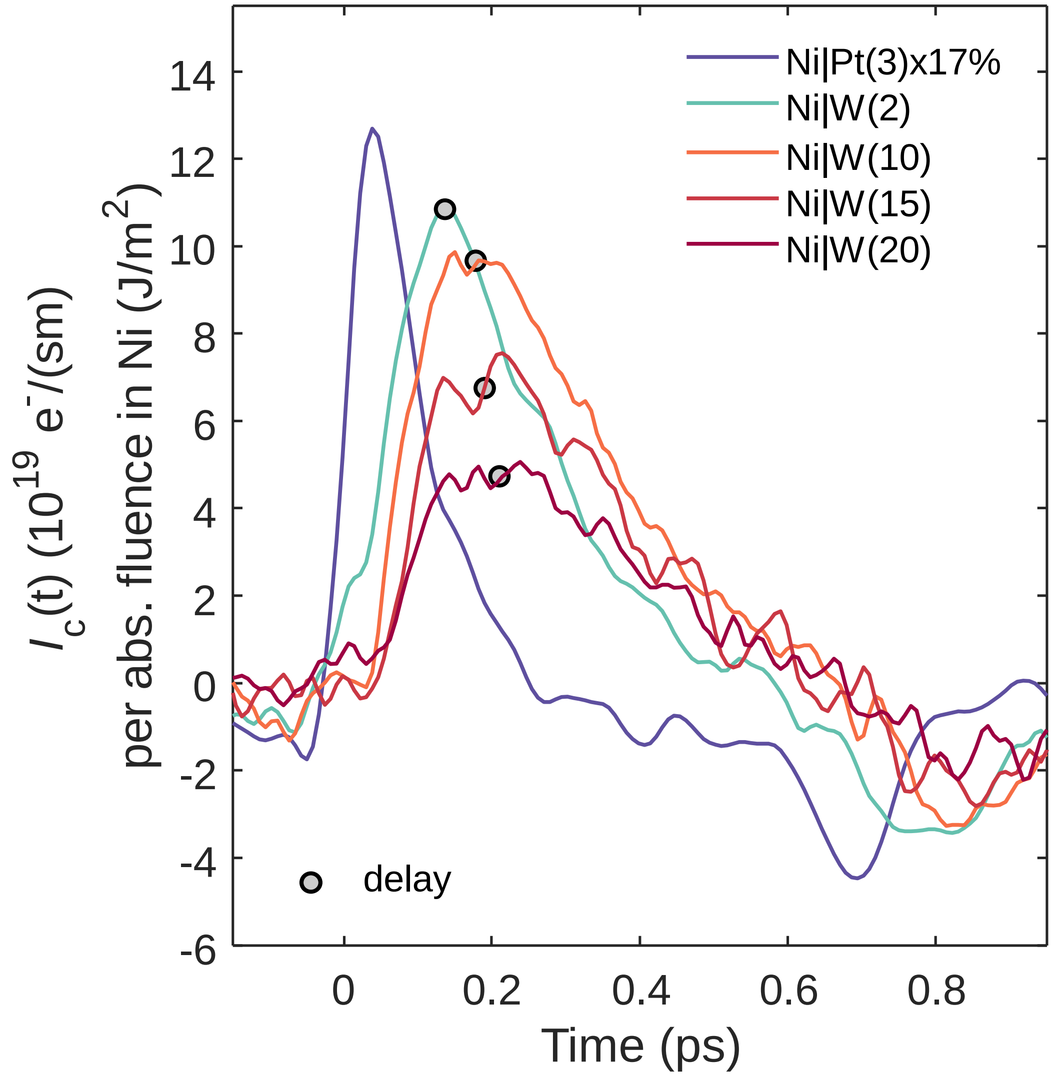
Stamm, *et al.*, PRB (2010)

Observation:
Ni|Pt vs Ni|W different dynamics.

Interpretation:
arises from (ii) and (iii)

TSS, *et al.*, Nature Nano. (2023)

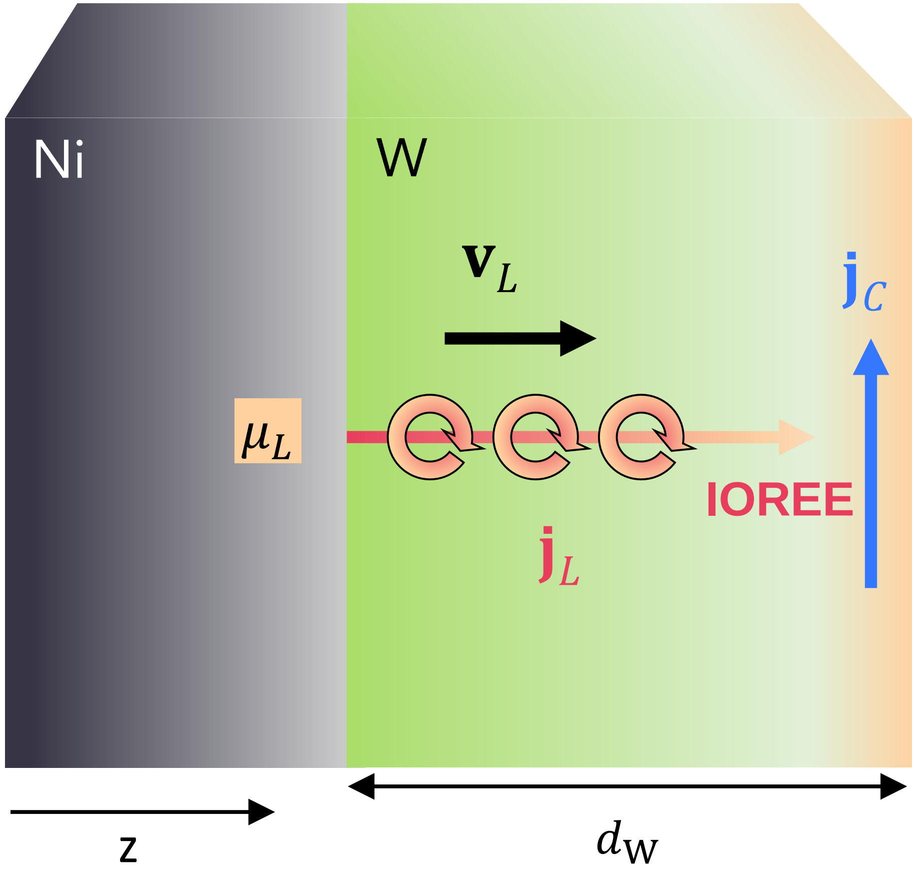
Ni|W THz Currents



1. Delayed
2. Sign change
3. Broadened

What leads to delayed response in Ni|W?

Inv. orbital Rashba-Edelstein effect



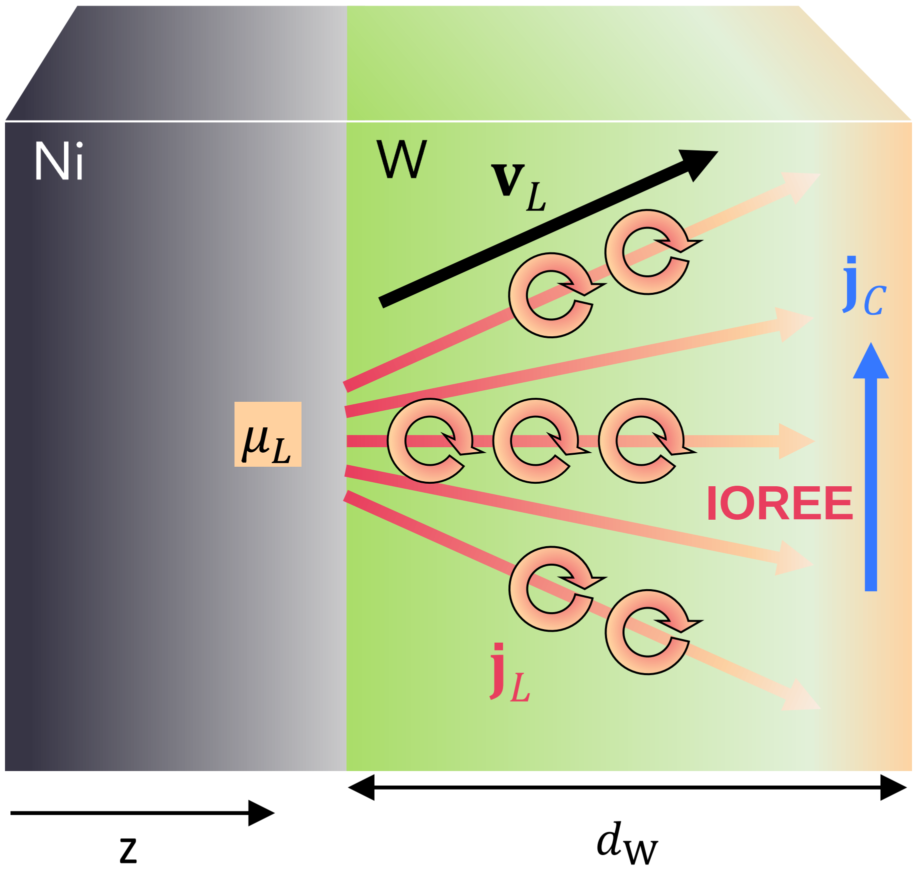
Interpretation

1. Delay: there is some arrival layer

2. Sign change: IOREE in W|SiOx same sign as SHE in Pt
Confirmed by ab-initio calculations

TSS, *et al.*, Nature Nano. (2023)

Inv. orbital Rashba-Edelstein effect



Interpretation

1. **Delay:** there is some arrival layer

2. **Sign change:** IOREE in W|SiOx same sign as SHE in Pt
Confirmed by ab-initio calculations

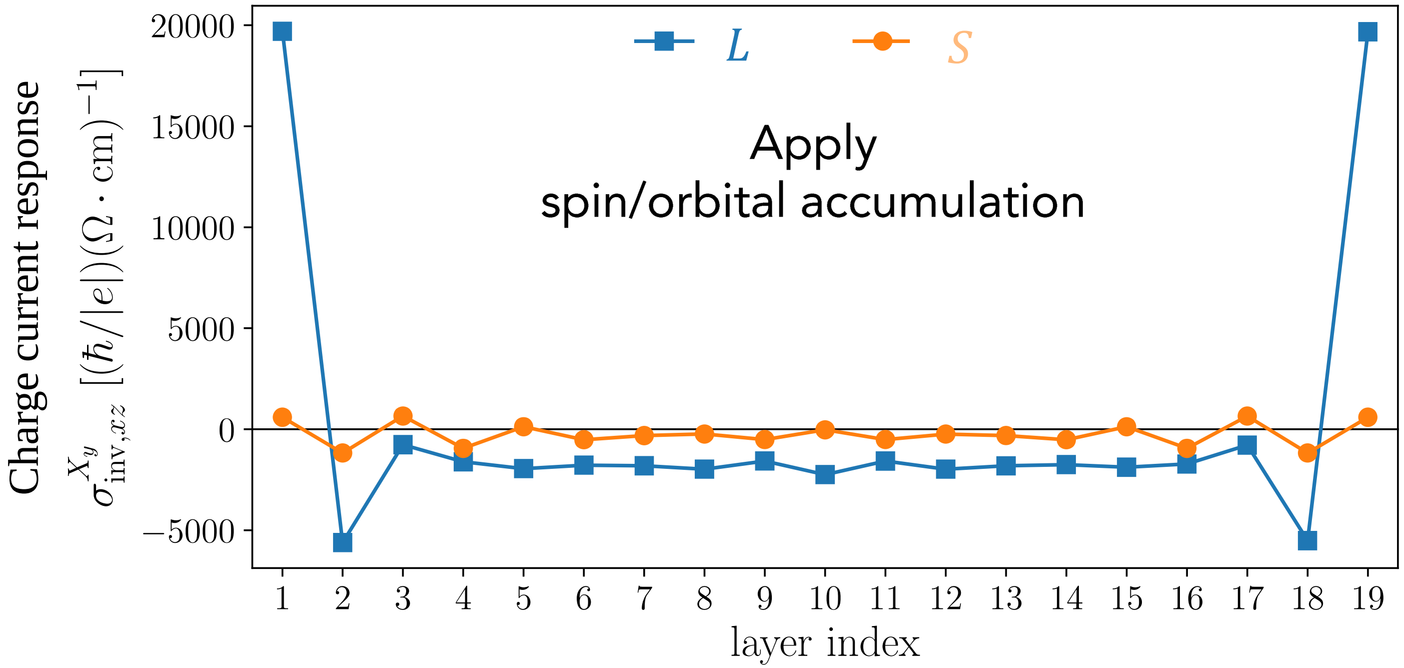
3. **Broadening:** Angular dispersion

4. **Long-range relaxation:** Exponential decay

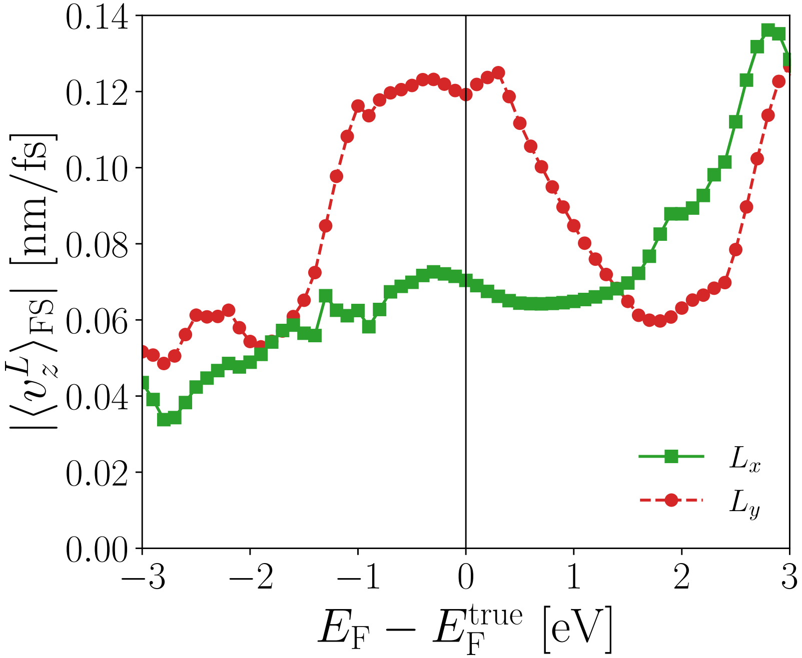
Can theory support this view?

TSS, *et al.*, Nature Nano. (2023)

L/S-to-charge (19 W atoms plus vacuum left/right)



Orbital velocity (bcc W bulk)



Huge inv. orbital Rasha-Edelstein effect

L velocity of 0.1 nm/fs

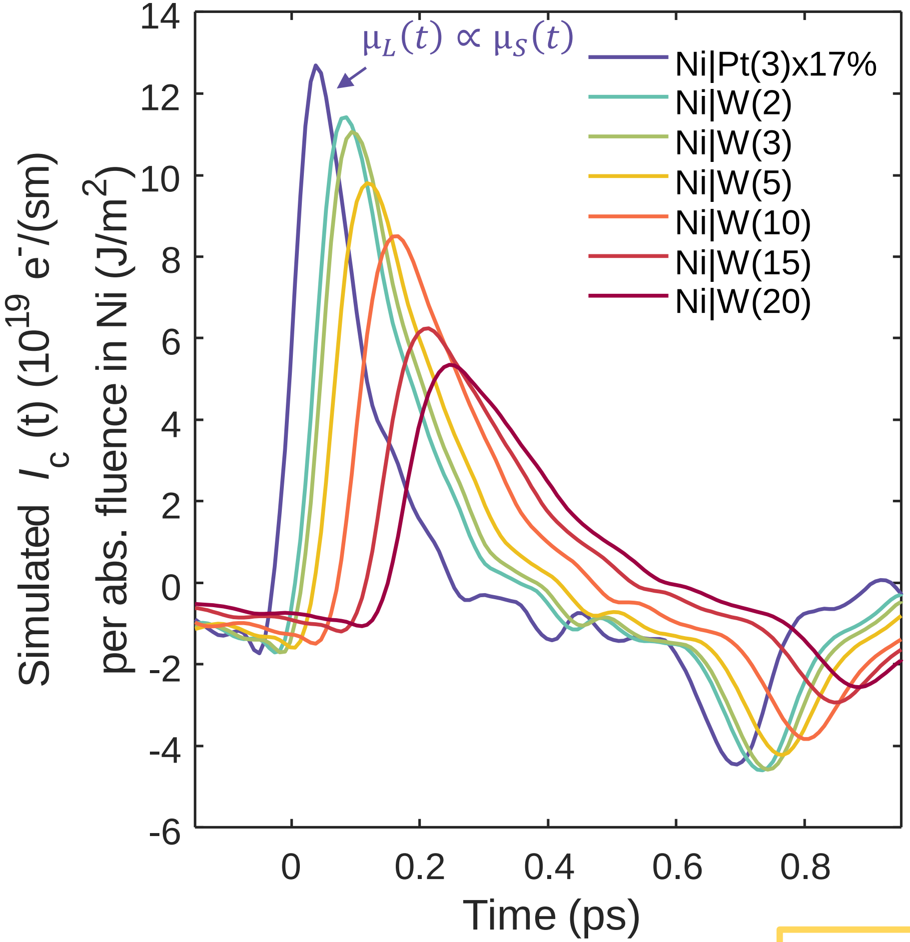
Calculations: Dongwook Go, Frank Freimuth, Yuriy Mokrousov

Model our Ni|W data

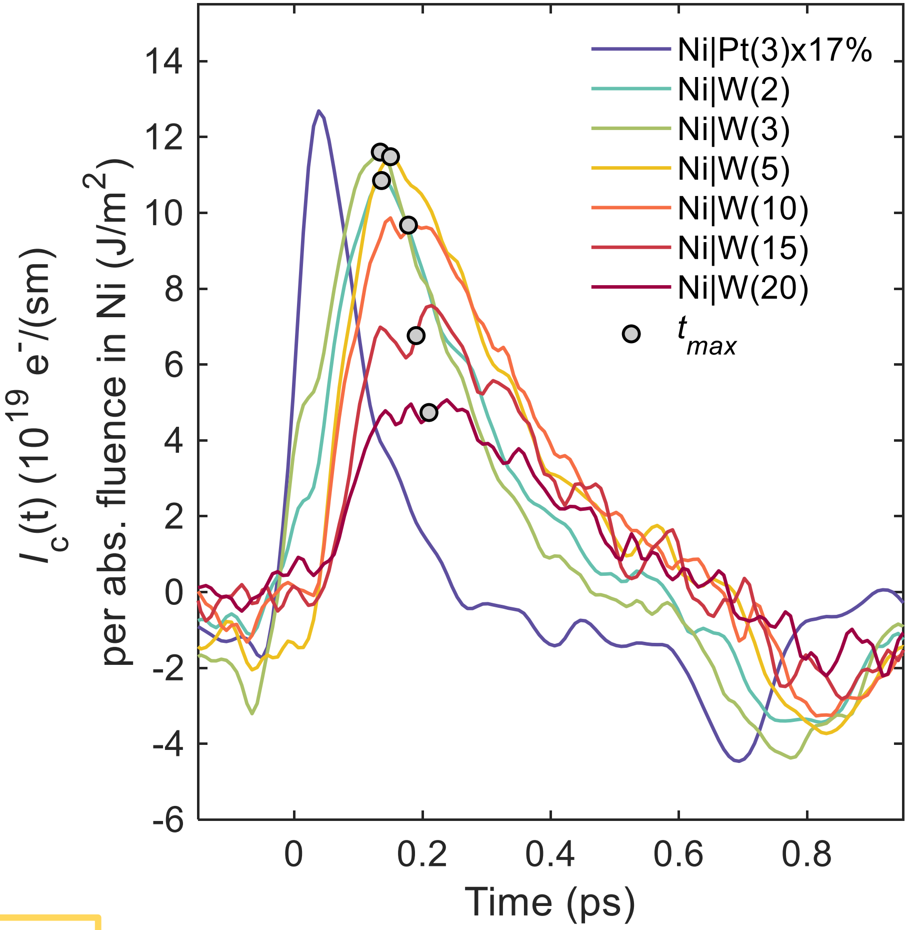
TSS, *et al.*, Nature Nano. (2023)

THz IOREE: experiment vs model

Model



Experiment



L propagation over ≫20 nm

L current lives for about 1ps

Sala *et al.*, *PRL* (2023)

v_L=0.1 nm/fs
$\lambda_{\text{rel}} = 80$ nm

Ti: $\lambda_{\text{rel}} = 80$ nm Choi, Woo et al. Nature (2023)

TSS, *et al.*, Nature Nano. (2023)

Similar time delays observed:
Mishra et al. arXiv:2401.08373 (2024). Xu et al. arXiv:2307.03490 (2023).

S & *L* conclusions

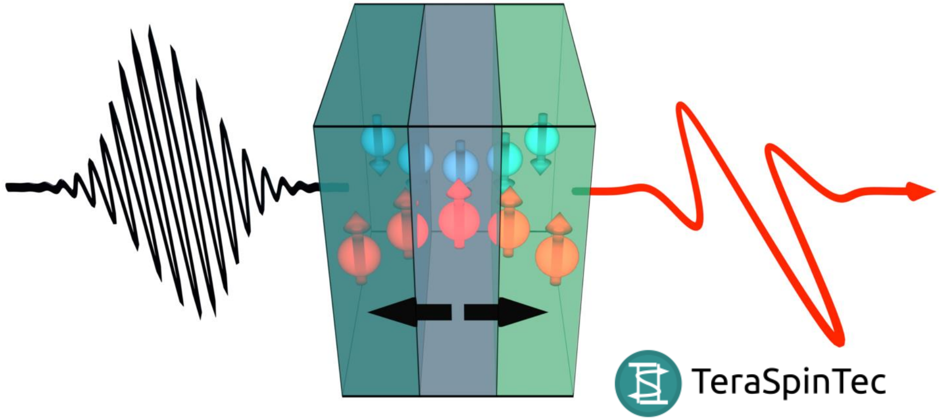
S

New insights

- Spintronic effects operative at THz rates
- Spin voltage drives ultrafast demag and THz spin transport
- Optimization: efficient spintronic THz sources
- Ultrabraodband THz detection through inversion

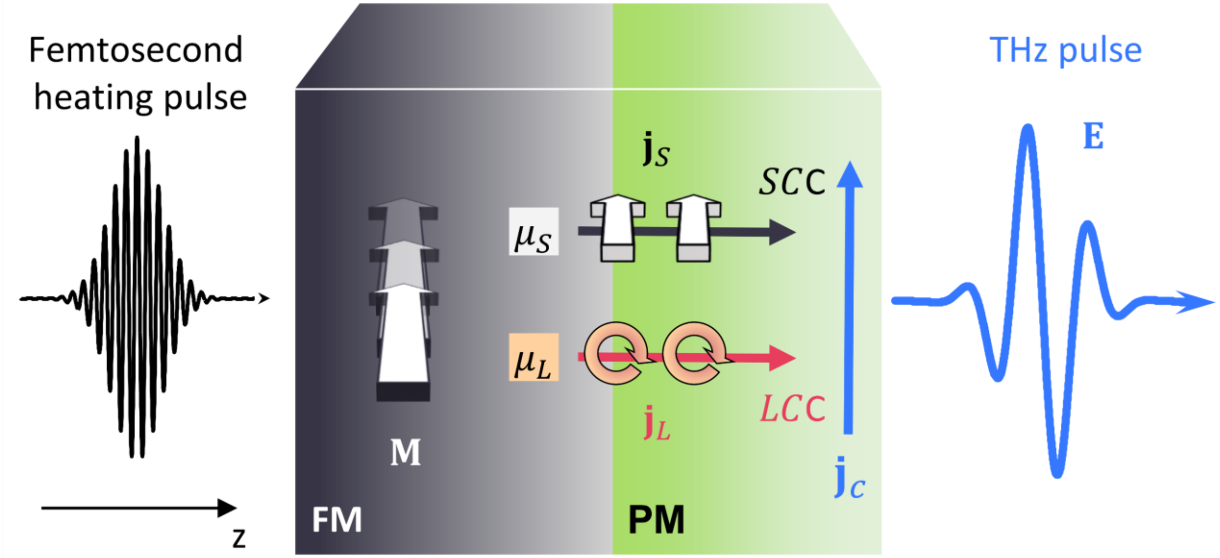
Chekhov, TSS *et al.*, PR Appl. 20 (2023)   R. Rouzegar, TSS *et al.*, PRB 106 (2023)

Spintronic Terahertz Emitters



Recent review: TSS et al., APL 120 (2022)

L



New insights

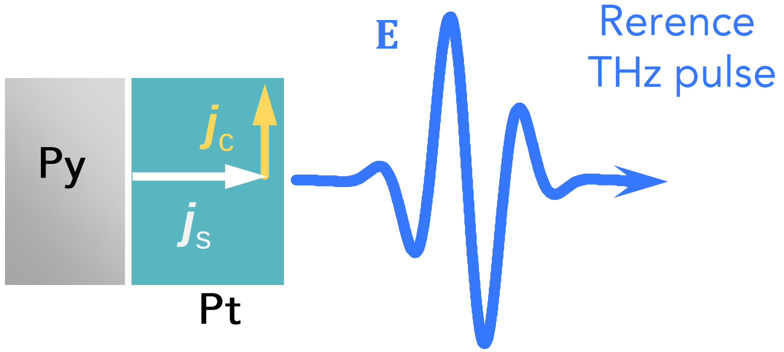
- *L* currents in Ni|W: reach 10's of nm
- *L* currents live for about 1 ps in W
- W|SiOx interface: efficient *L*-to-charge conversion

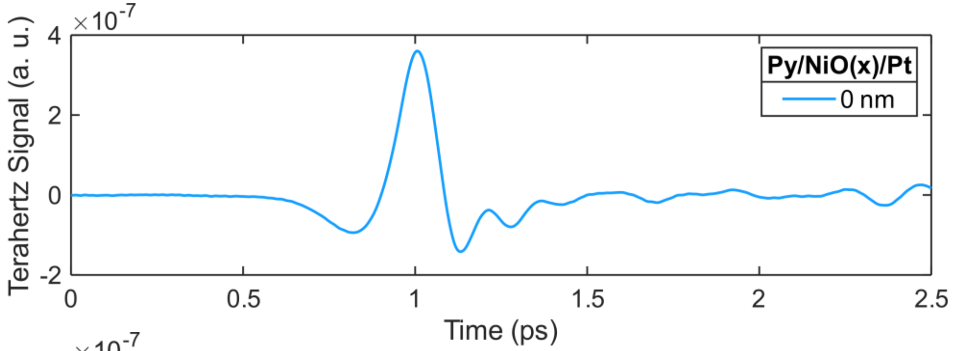
TSS, *et al.*, Nature Nano. (2023), doi.org/10.1038/s41565-023-01470-8

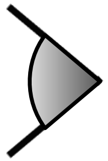
Terahertz emission spectroscopy can sense *S*/*L* current propagation

Outlook: THz spin conductance spectroscopy

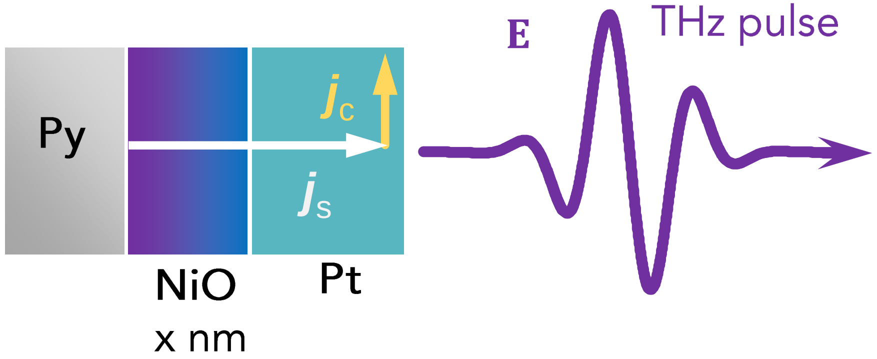
Samples: D. Wu

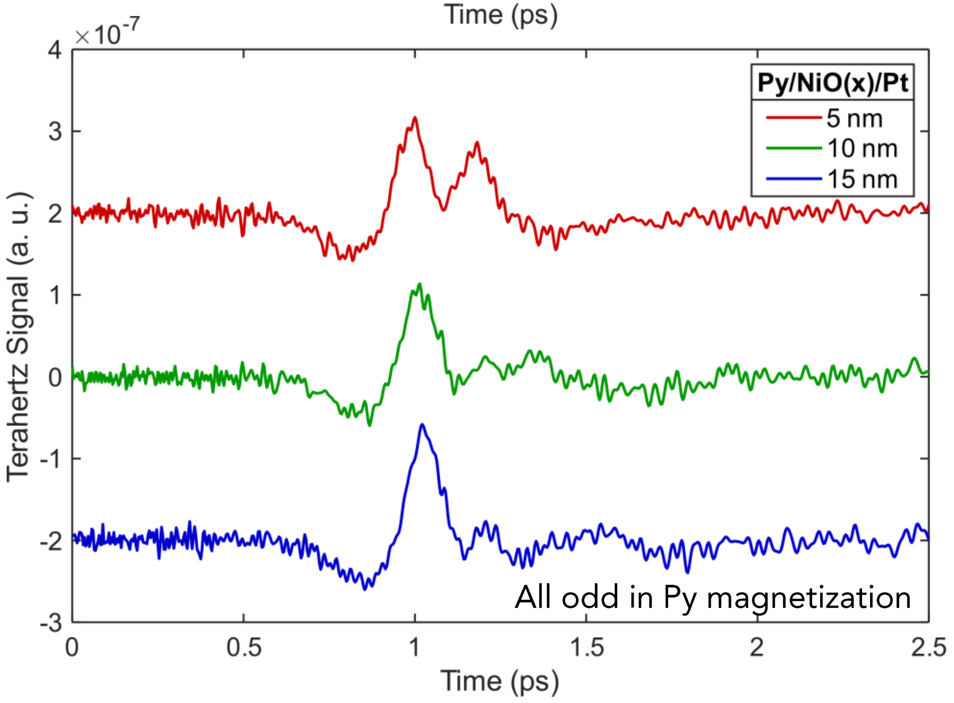


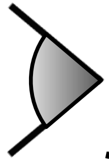




Signal at detector
$S_{\mathrm{ref}}(t)$







Signal at detector
$S(t) = \sigma_{\mathrm{NiO}}(t) * S_{\mathrm{ref}}(t)$

Convolution with NiO spin conductance

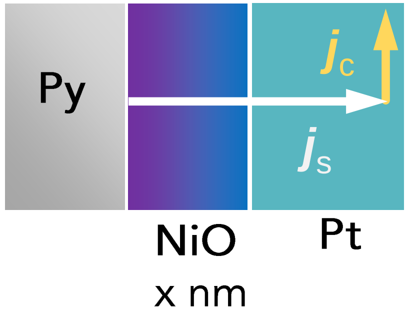
Q. Hongsong, Z. Kaspar

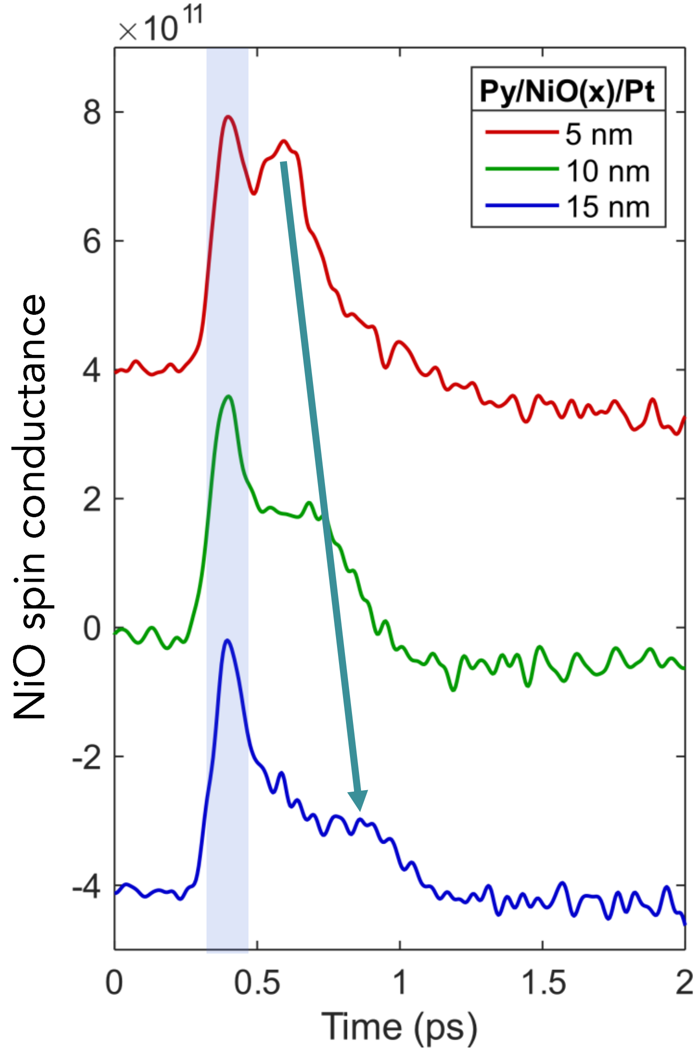

What is the NiO spin conductance?

Outlook: THz spin conductance spectroscopy

Samples: D. Wu

Get NiO spin conductance: Fourier transform & divide $\sigma_{\mathrm{NiO}}(\omega) = S(\omega)/S_{\mathrm{ref}}(\omega)$





Two contributions:

I. Immediate response
   magnetic dipole

II. Time delayed response
   magnon spin current through NiO

Magnon group velocity
30 nm/ps (30 km/s)

Hutchings, Samuelsen, PRB 6 (1972)

Q. Hongsong, Z. Kaspar

Related TES studies: Sasaki *et al.*, APL (2020).; Lee *et al.*, Nature Nano. 16 (2021);

Rouzegar, TSS et al. *arXiv preprint arXiv:2305.09074* (2023).

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Keio University

David Reiß
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HZDR

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Georg Woltersdorf

Felix Paries
Georg von Freymann

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Ultrafast Spin Dynamics

s-Nebula
Novel Spin-Based Building Blocks for Advanced TeraHertz Applications

S

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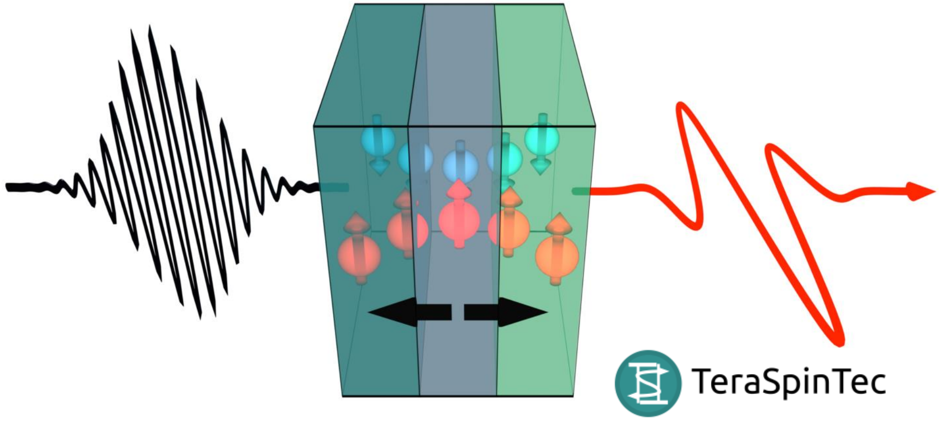
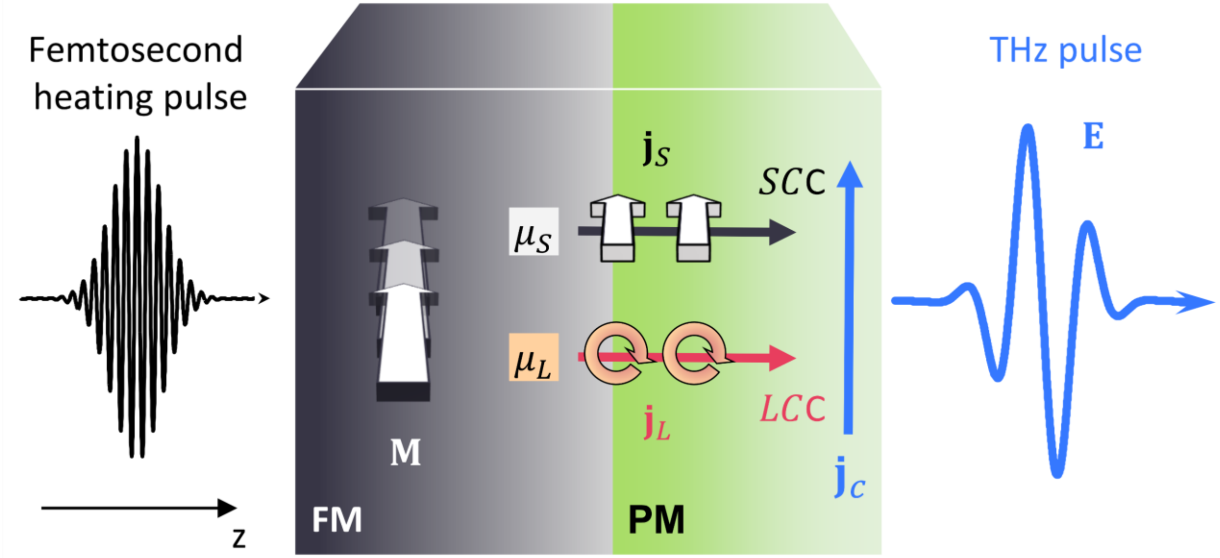


Recent review: TSS et al., APL 120 (2022)

L



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