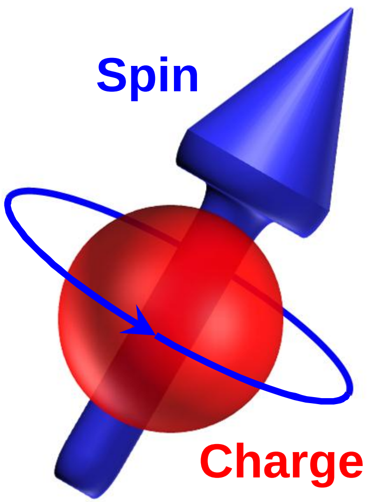
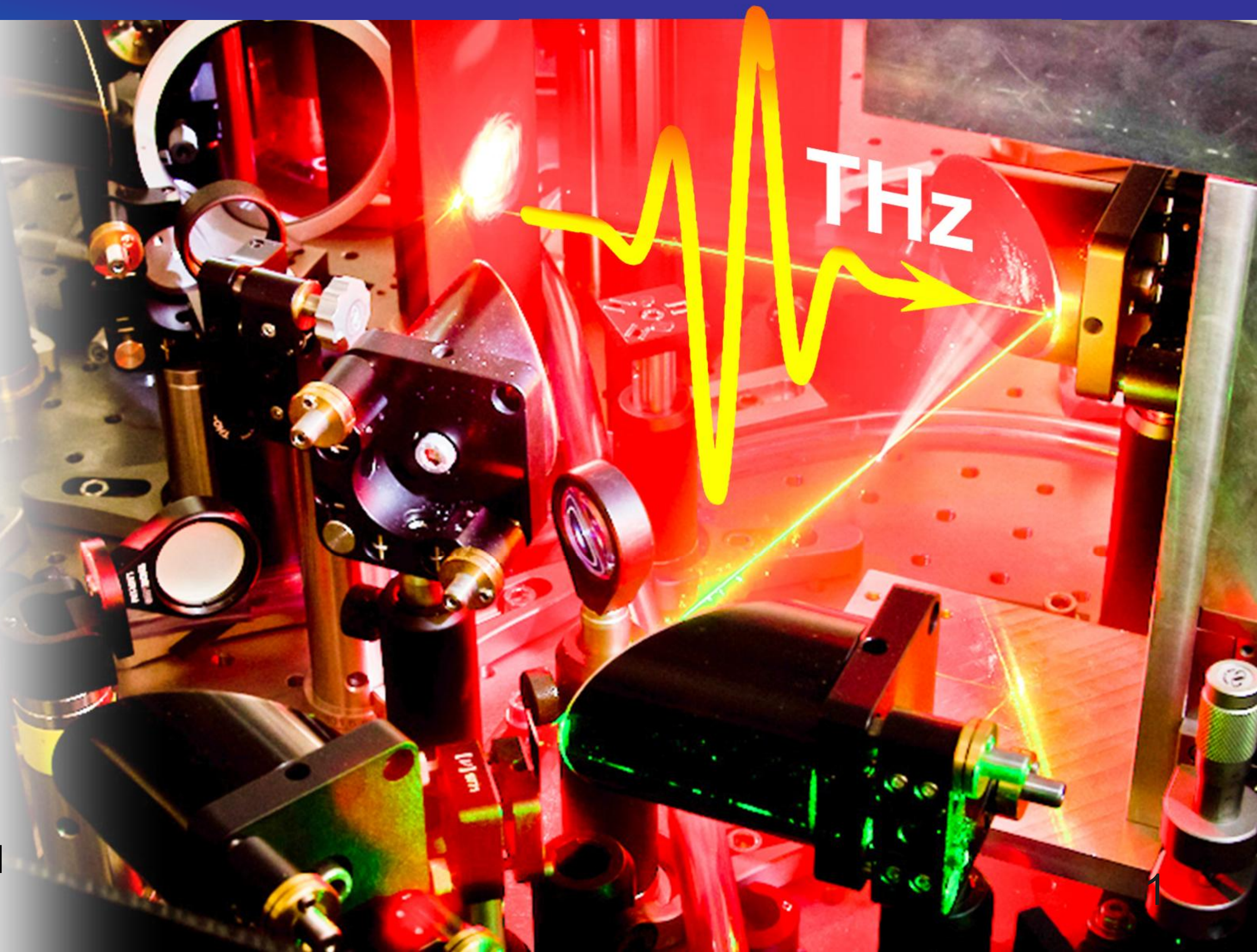
# Terahertz spin transport in magnetic nanostructures

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Fritz-Haber-Institut/MPG

# Acknowledgments

**FUB/FHI THz group, for these results:**
Reza Rouzegar
Oliver Gückstock
Lukáš Nádvorník
Tom Seifert
Martin Wolf

JGU

Samridh Jaiswal
Gerhard Jakob
Martin Jourdan
Mathias Kläui

Marco Battiato
Pablo Maldonado
Peter Oppeneer

UPPSALA UNIVERSITET

ERNST MORITZ ARNDT UNIVERSITÄT GREIFSWALD

Ulrike Martens
Markus Münzenberg

David Reiß
Piet Brouwer

Freie Universität Berlin

MARTIN-LUTHER UNIVERSITÄT HALLE-WITTENBERG

Liane Brandt
Georg Woltersdorf

TECHNISCHE UNIVERSITÄT DRESDEN

Richard Schlitz
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DFG

**CRC / TRR 227**

**Ultrafast Spin Dynamics**

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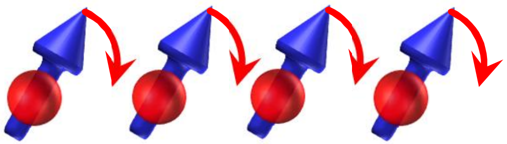
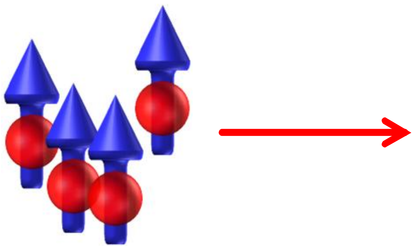
# Spintronics: 3 basic operations

1. **Turn spins around**



→ Torque

2. **Transport spins**

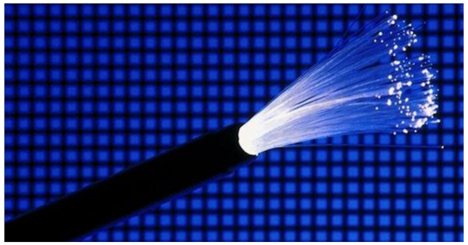


→ Spin current

3. **Detect spin dynamics**

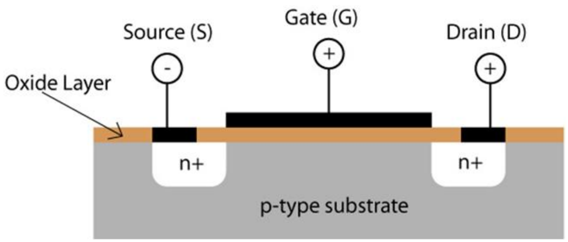
**Goal:** Reach speed of other information carriers: 1 THz = $10^{12}$ Hz

**Photons in fibers:** >10 Tbit/s



Hillerkuss *et al.*, Nature Phot. (2011)

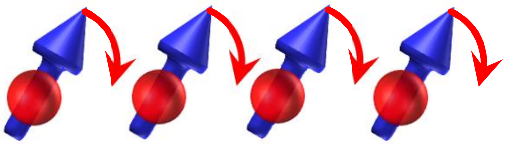
**Electrons in a FET:** ~1 THz cut-off




Del Alamo *et al.*, Nature (2011)

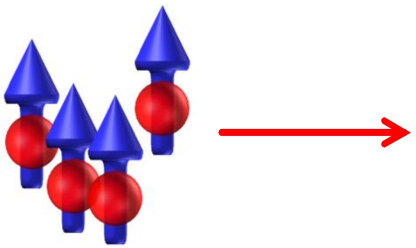
# Spintronics: 3 basic operations

1. **Turn spins around**



→ Torque

2. **Transport spins**



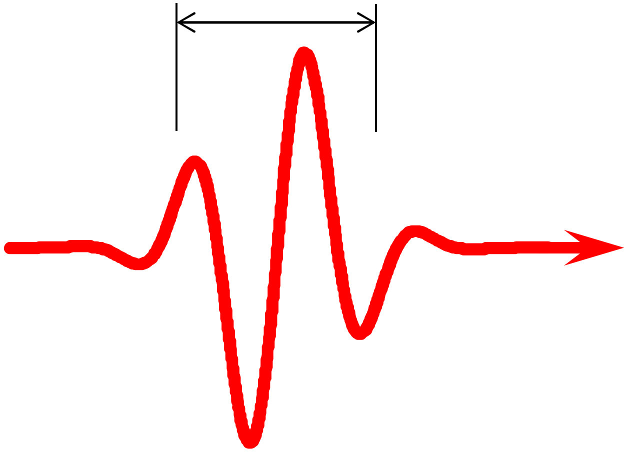
→ Spin current

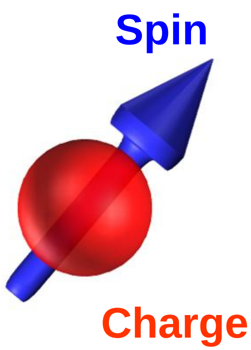
3. **Detect spin dynamics**

**Goal:** Reach speed of other information carriers: 1 THz = $10^{12}$ Hz

**Idea:** Use THz electromagnetic pulses

**1 ps = (1 THz)$^{-1}$ = 300 μm/$c$**



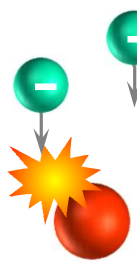



**THz + spin transport = useful?**

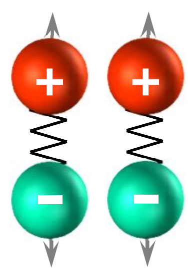
# Why THz spin transport?

1) **Test the speed of spintronic effects, reveal initial steps**
   E.g. spin-Hall, spin-Seebeck and GMR

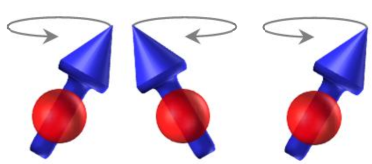
2) **New physics, new methods**
   As THz coincides with many fundamental modes



Intraband transport

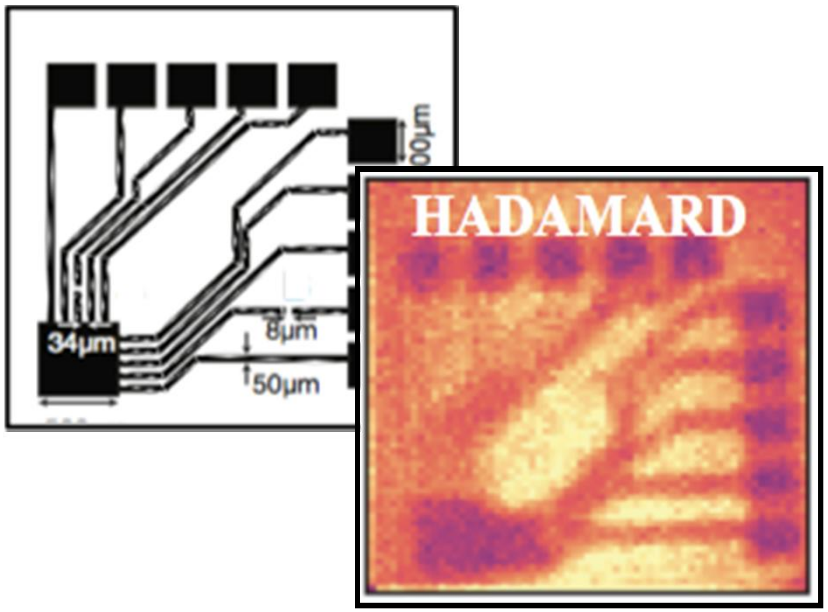


Phonons



Magnons

3) **Reward for THz photonics**
   E.g. THz sources and modulators for spectroscopy and imaging



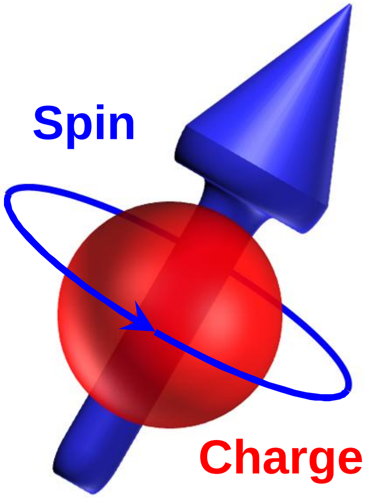

Stantchev *et al.*, Science Adv. (2016)

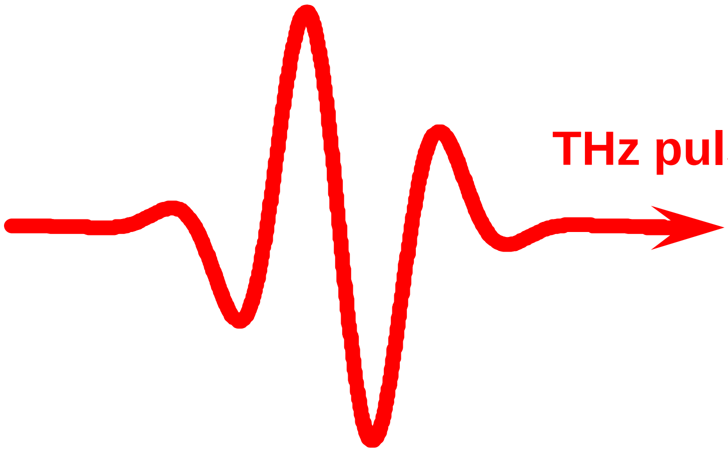
**How to drive spin transport?**

**Important drivers of spin transport**

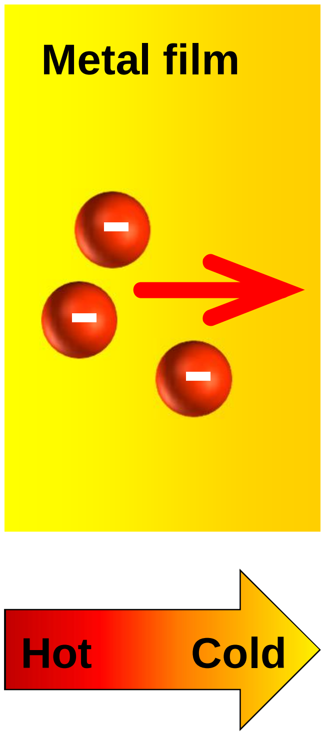
- Electric fields: E.g. spin Hall effect
- Heat: Spin-caloric effects

**How to transfer to the THz range?**



# Heat-driven: The Seebeck effect




Temperature gradient

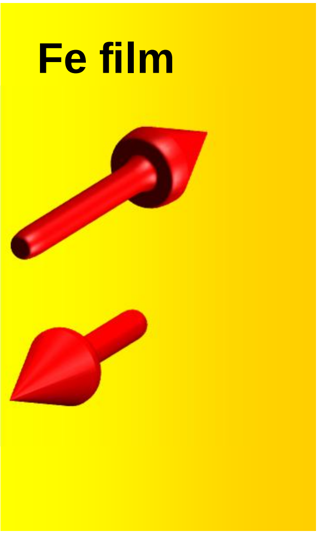
**Thomas Seebeck (1821):**

A temperature gradient drives an electron current

**Uchida and Saitoh (2008):**

In ferromagnets, the Seebeck current is spin-dependent

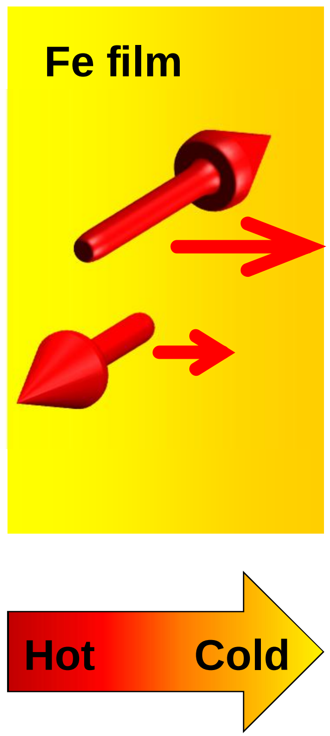
# Spin-dependent Seebeck effect (SDSE)




**↑ and ↓ electrons have very different transport properties**

Uchida, Saitoh *et al*., Nature (2008)
Bauer, Saitoh, Wees, Nature Mat. (2013)

# Spin-dependent Seebeck effect (SDSE)




Temperature gradient

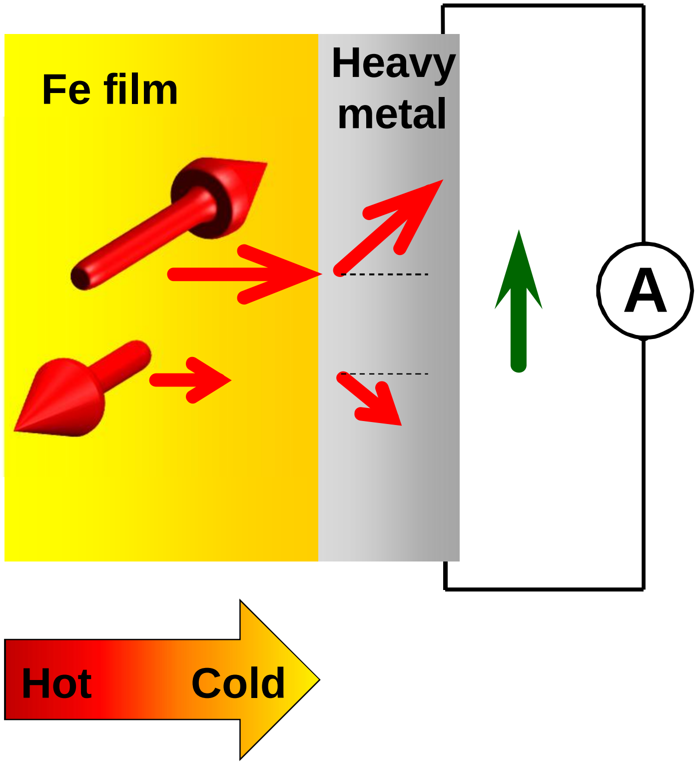
**$\uparrow$ and $\downarrow$ electrons have very different transport properties**

Uchida, Saitoh *et al.*, Nature (2008)
Bauer, Saitoh, Wees, Nature Mat. (2013)

$\Rightarrow$ Spin-polarized current

**Detection with the inverse spin Hall effect**

# Inverse spin Hall effect (ISHE)




Temperature gradient

Spin-orbit coupling deflects electrons

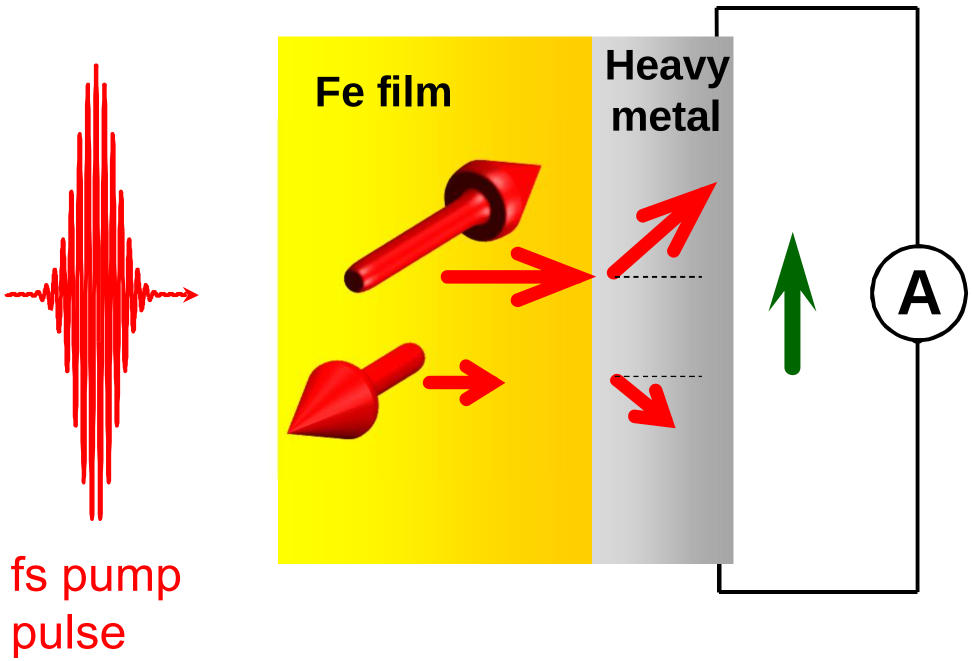
⇒ Transverse charge current

⇒ Spin-to-charge (S2C) conversion

Saitoh *et al.*, APL (2006)

**How can we induce an imbalance as fast as possible?**

# Inverse spin Hall effect (ISHE)




**Ultrafast spin transport**

Malinowski *et al.*, Nature Phys. (2008)
Melnikov, Bovensiepen *et al.*, PRL (2011)
Rudolf *et al.*, Nat. Comm. (2012)
Eschenlohr *et al.*, Nature Mat. (2013)
Turgut *et al.*, PRL (2013)
Schellekens *et al.*, Nat. Comm. (2014)
Ganichev *et al.*, Nature (2002)

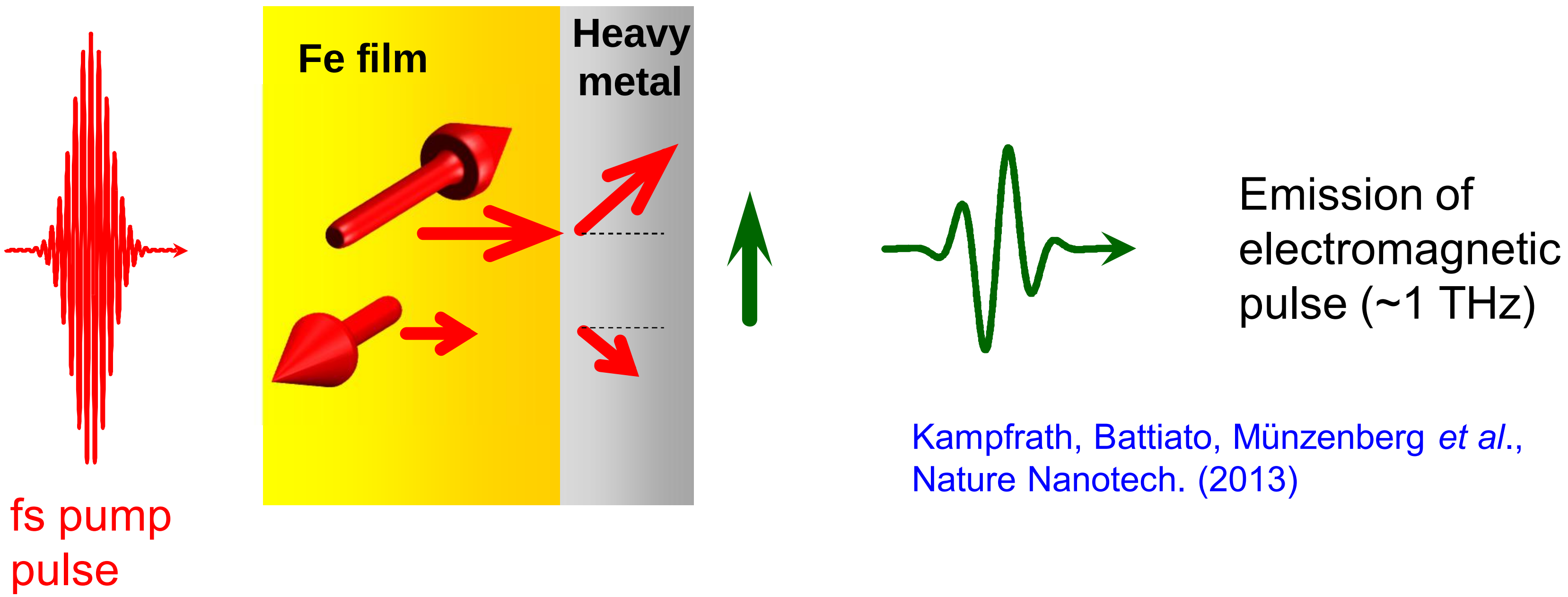
**Goal:**

Measure quasi-electrically

**Technical challenge:**

- Electric detection has cutoff at $<$ 50 GHz
- But expect bandwidth $>$ 10 THz

# Inverse spin Hall effect (ISHE)



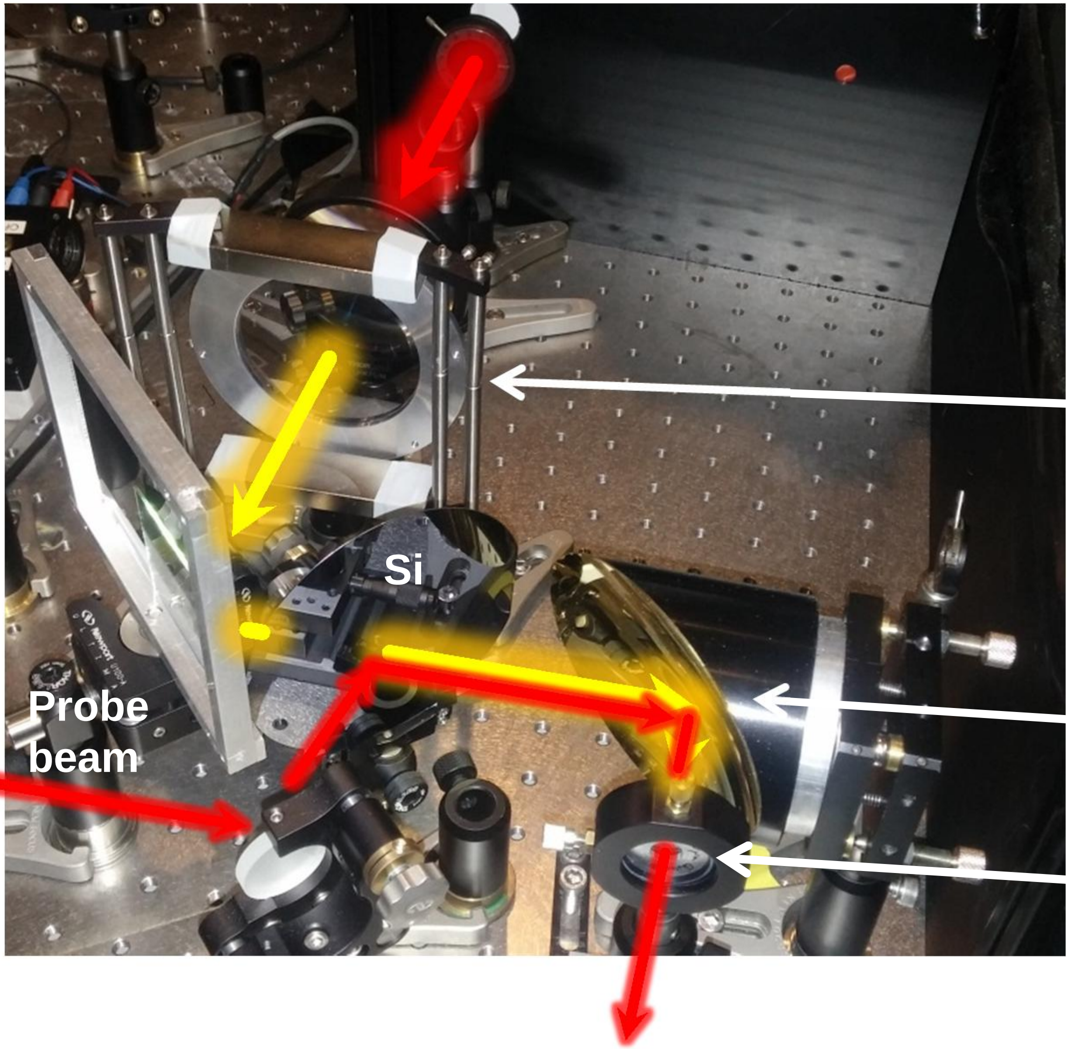

Kampfrath, Battiato, Münzenberg *et al.*, Nature Nanotech. (2013)

$\Rightarrow$ Measure THz emission from photoexcited FM|NM bilayers

**Samples:** Polycrystalline films (from Kläui, Münzenberg, Woltersdorf labs)

**Pump pulses:** From Ti:sapphire oscillator (10 fs, 800 nm, 2.5 nJ, 80 MHz)

# Simple THz emission setup in lab




**Optical pump beam**

**Spintronic sample**

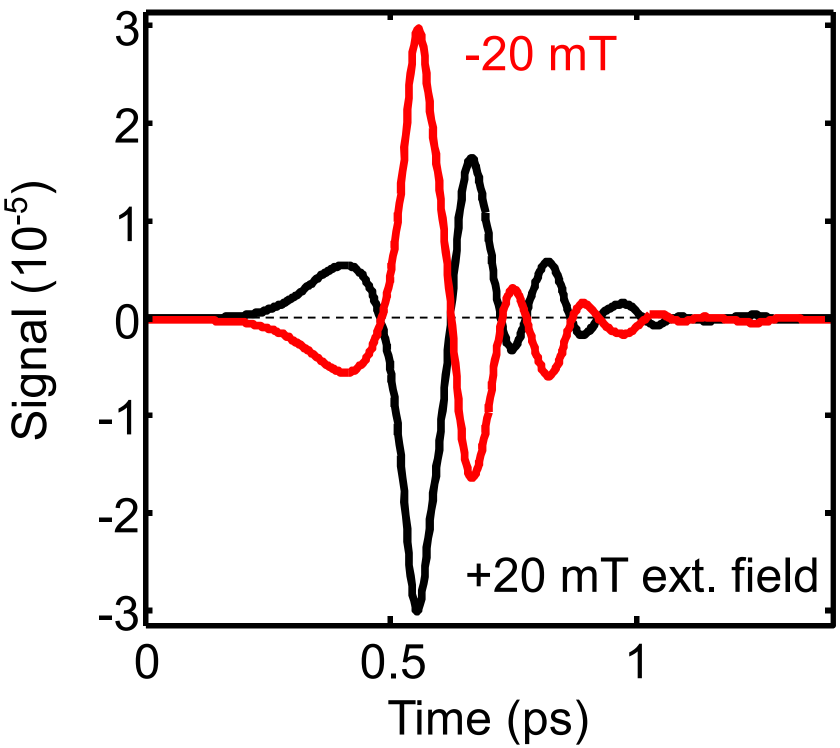
**Parabolic mirror**

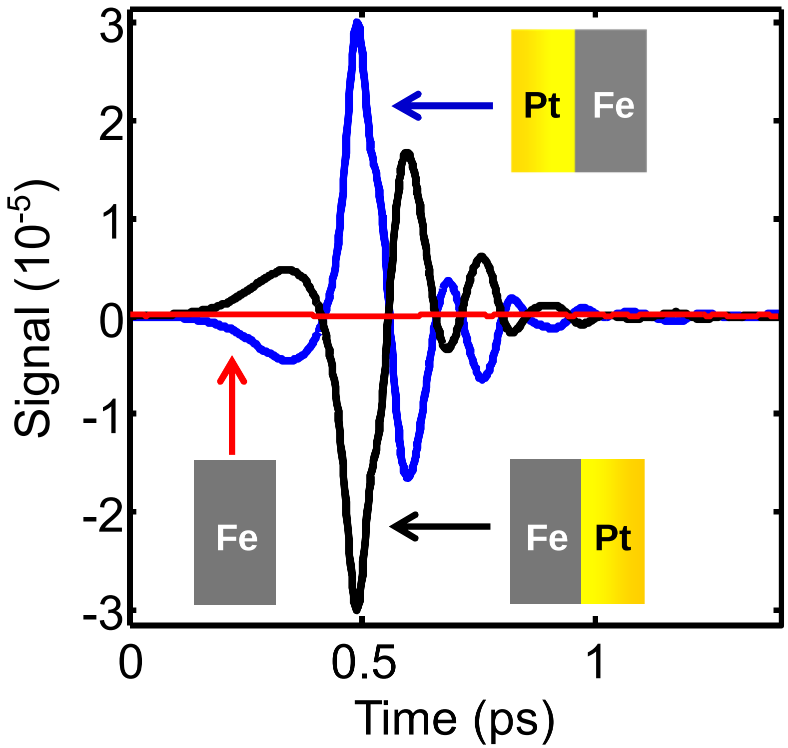
**Electro-optic crystal for sampling of the THz electric field**

**To detection of probe ellipticity**

**Typical signals**

# Typical THz waveforms from Fe|Pt bilayers




**Further findings**

- Signal ∝ pump power
- THz electric field ⊥ sample magnetization

Consistent with scenario spin transfer + ISHE

**Need more evidence for the spin Hall scenario**

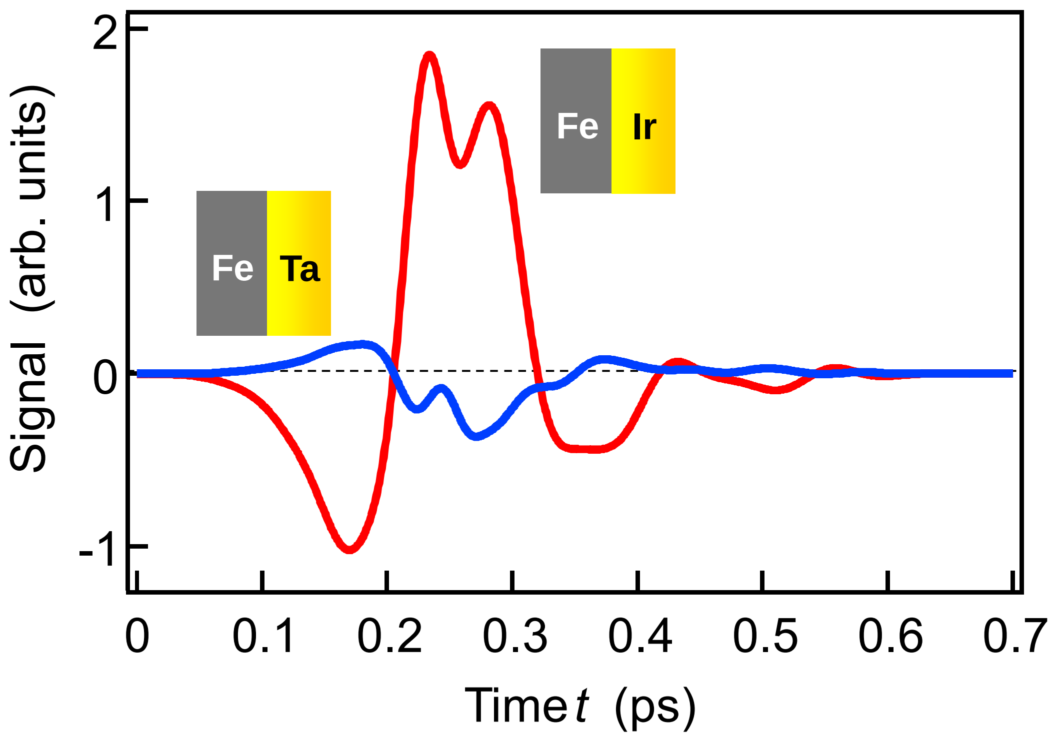
# Ultrafast inverse spin Hall effect

**Idea:**

Vary nonmagnetic cap layer

**Ta vs Ir:**

Opposite spin Hall angles, Ir larger




The inverse spin Hall effect is still operative at THz frequencies

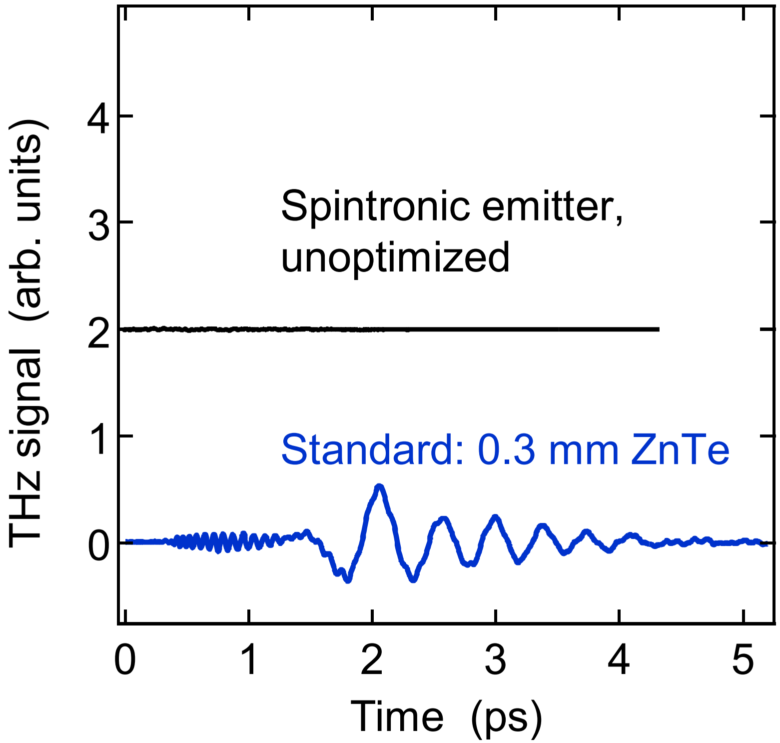
Kampfrath, Battiato, Oppeneer, Freimuth, Mokrousov, Radu, Wolf, Münzenberg *et al.*, Nature Nanotech. (2013)

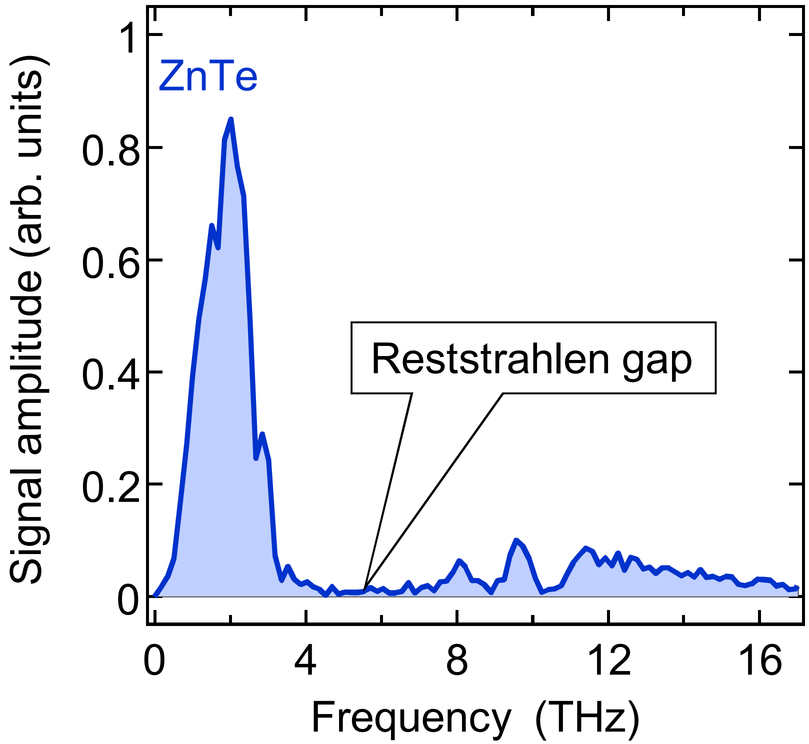
**Interesting applications:**

1) Rapid material characterization regarding spin-to-charge conversion
2) Measurement of spin-current dynamics
3) Generation of THz electromagnetic pulses

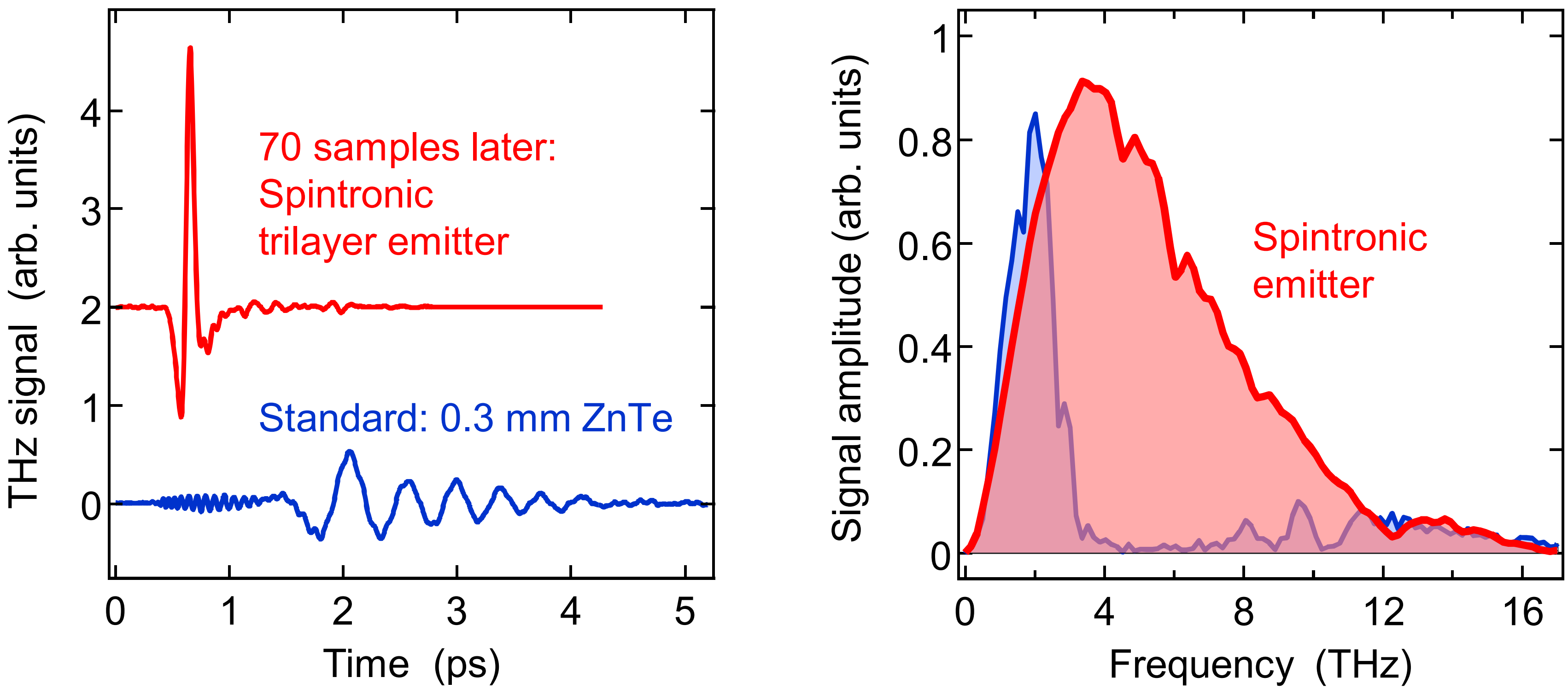
**Compare to state-of-the-art emitter**

# Spintronic THz emitter



# Spintronic THz emitter




More broadband, efficient and cheaper than standard emitters like ZnTe

Seifert, Radu, Kläui, TK *et al.*, Nature Photon. (2016)

Yang, Wu, Qi *et al.*, Adv. Opt. Mat. (2016)
Wu, Yang *et al.*, Adv. Mat. (2017)
Torosyan, Beigang, Papaioannou *et al.*, Sci. Rep. (2018)

**A quick look in the lab again…**

# Spintronic THz emitter in the lab


Movie by Oliver Gückstock and Lukáš Nádvorník

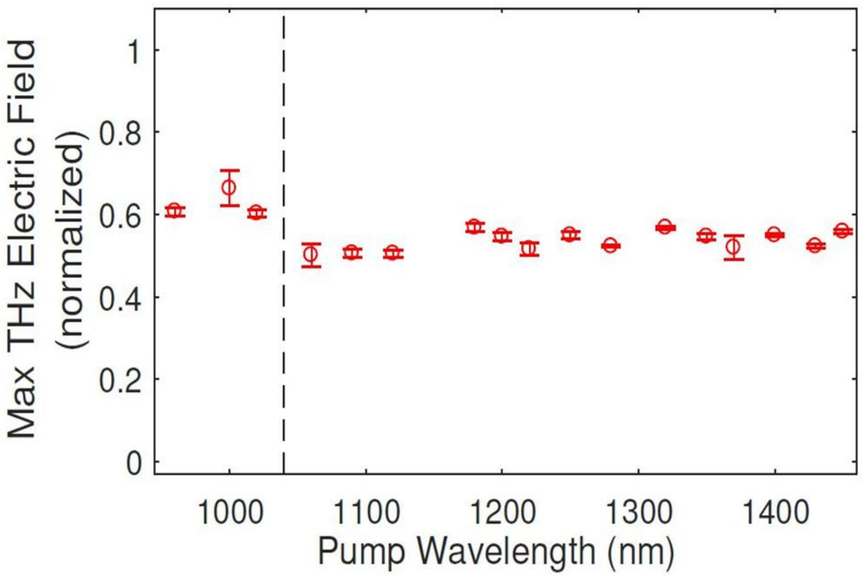
**Magnetism and thin metal enable interesting features…**

# More features of spintronic THz emitters

**Emitter is insensitive to pump wavelength**

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

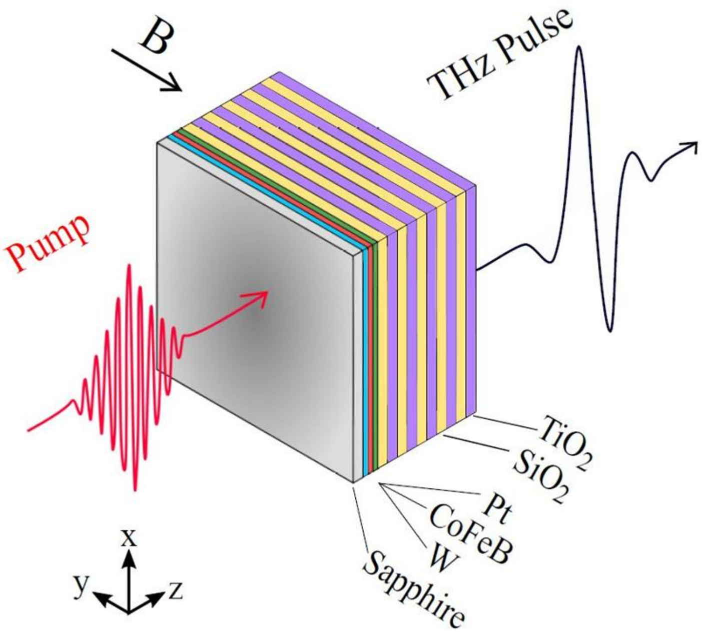
Herapath, Seifert, TK, Hendry *et al*., APL (2019)




**Cavity pushes pump absorption from ≈ 50% to ≈ 100%**

Feng *et al*., Adv. Opt. Mat (2018)

Herapath, Seifert, TK, Hendry *et al*., APL (2019)

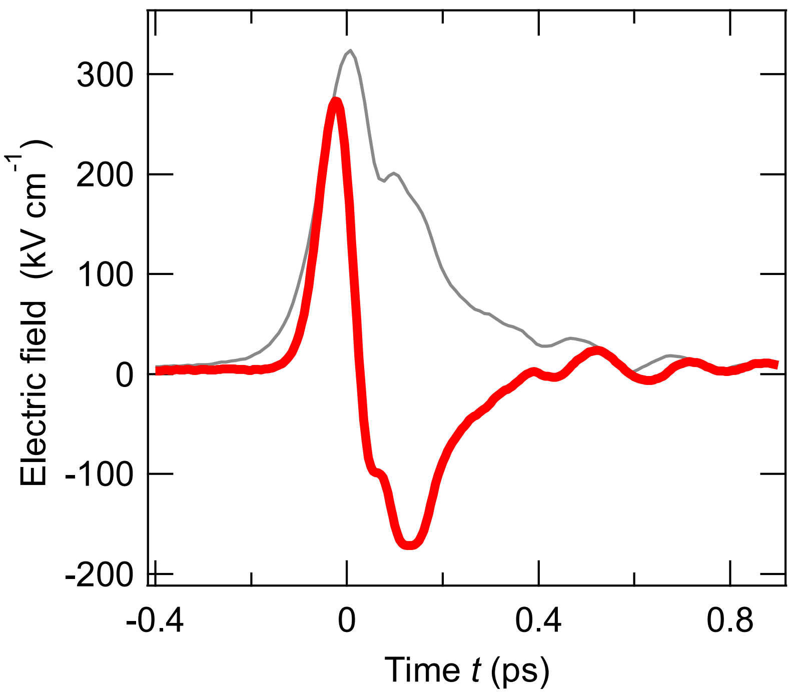



**Larger emitters are easy and cheap to fabricate**

# More features of spintronic THz emitters

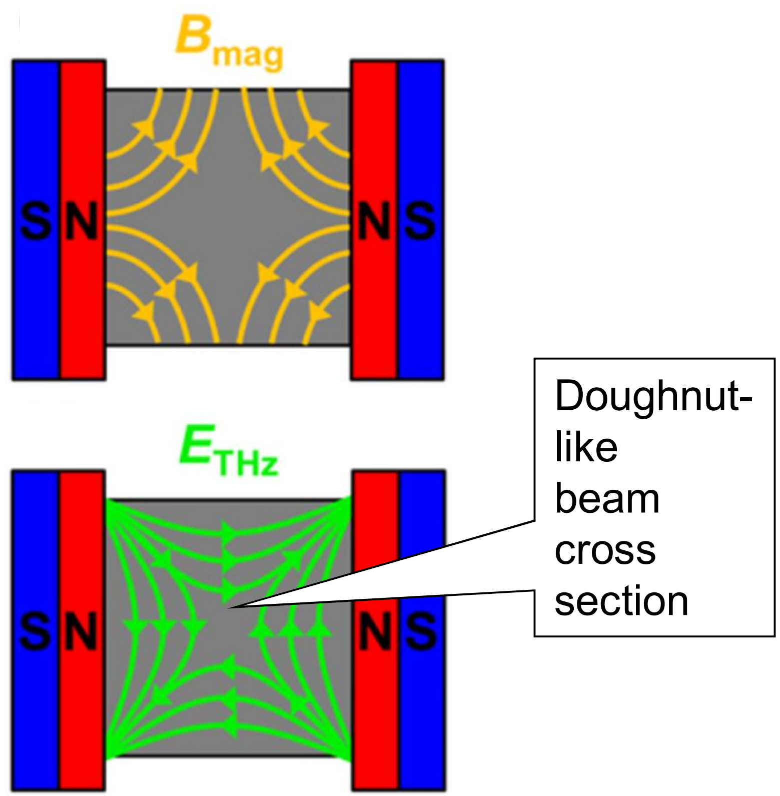
**Upscaling to ~5 cm diameter yields 0.3 MV/cm peak field**

Seifert, Kläui, TK *et al.*, APL (2017)
Fülöp, Tzortzakis, TK, Adv. Opt. Mat. (2020)




**Tailor the magnetization landscape to generate structured THz beams**

Hibberd, Graham *et al.*, APL (2019)

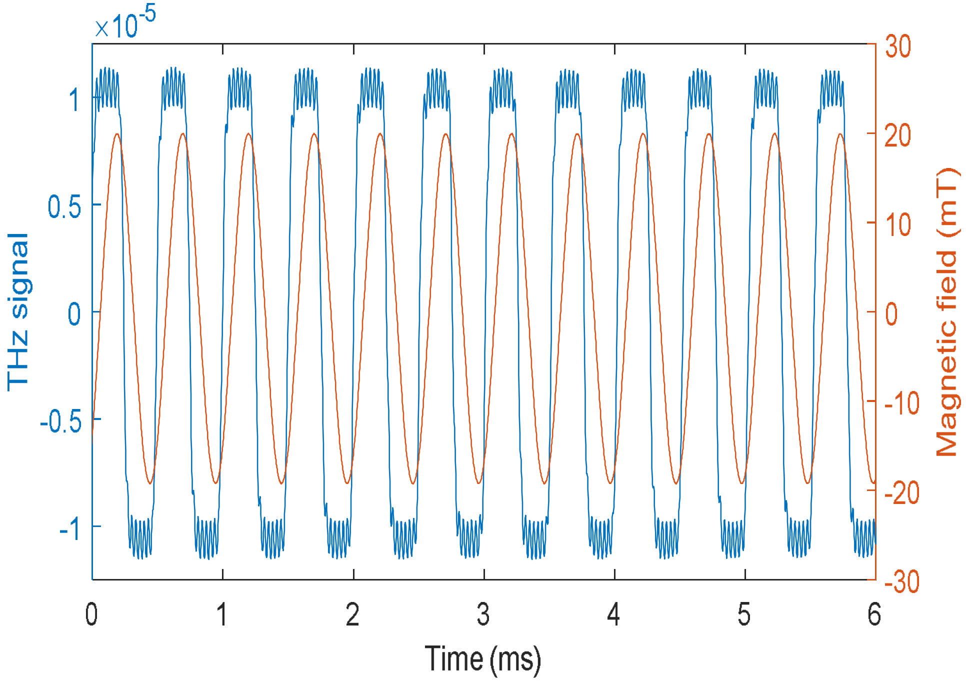



**The magnetization can also be temporally modulated**

# More features of spintronic THz emitters

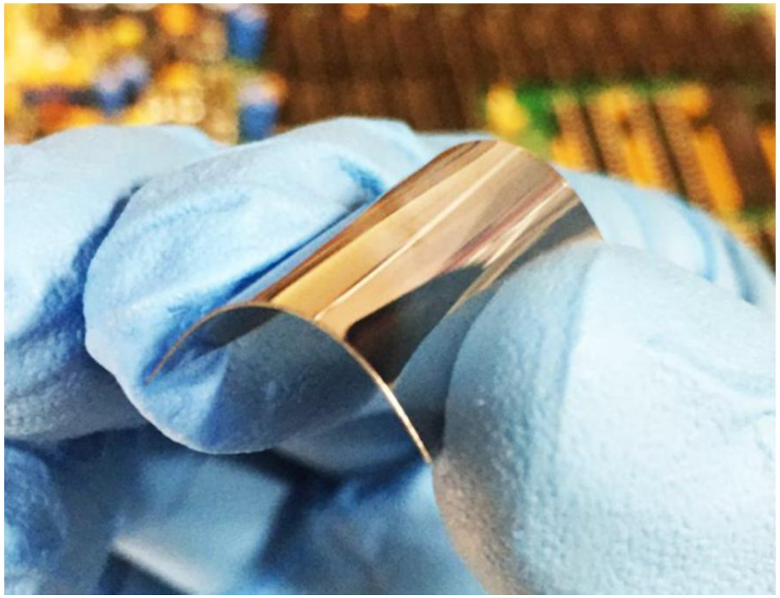
**Polarity modulation of THz field up to 20 kHz**

Gückstock, Nadvornik, TK *et al.* (2020)




**Bendable substrates**

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

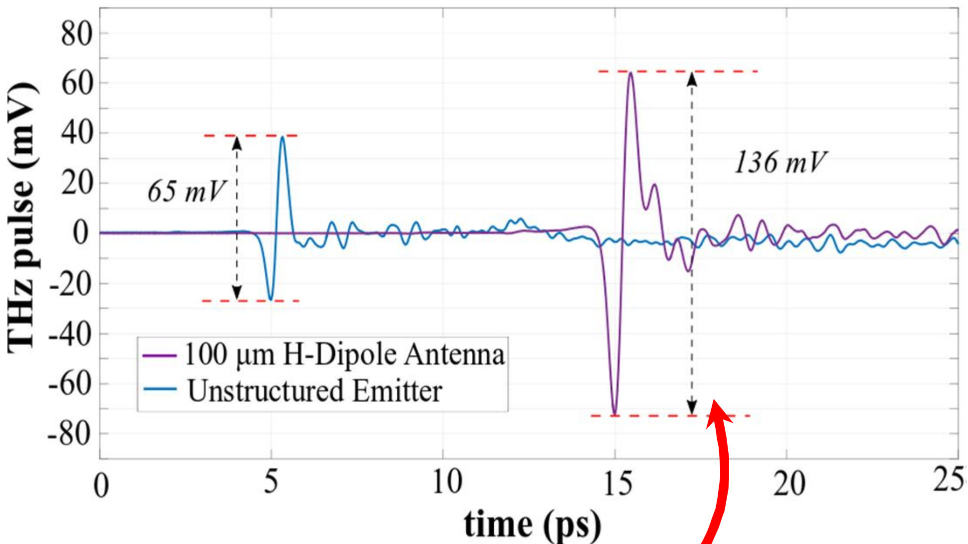


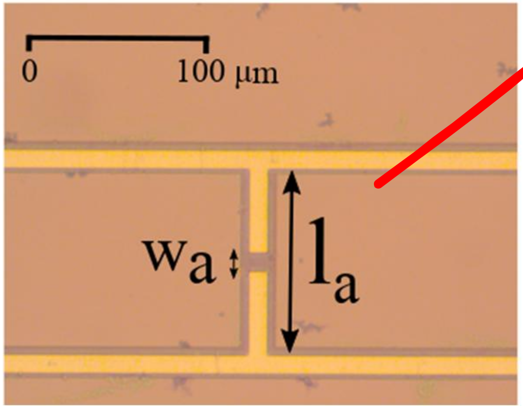
**Metal films can be nanostructured easily**

# More features by nanostructuring

**Antenna-coupled spintronic emitters**

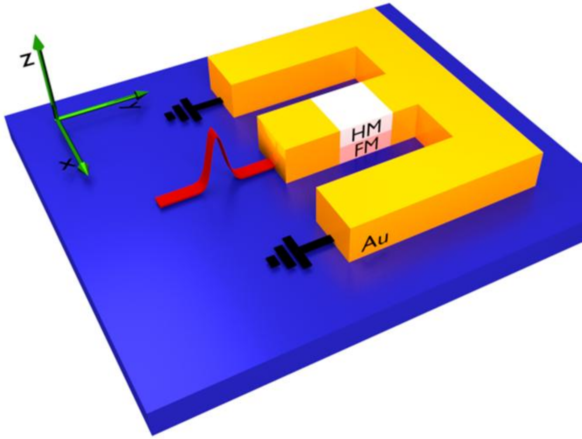
Nandi, Kläui, TK, Preu *et al.*, APL (2019)

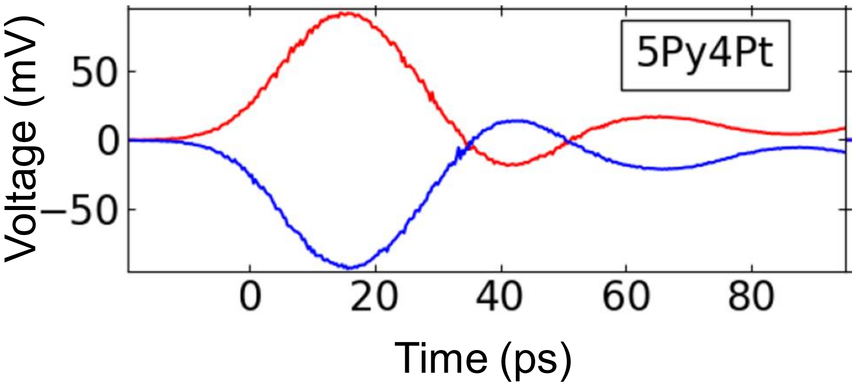



**On-chip THz source**

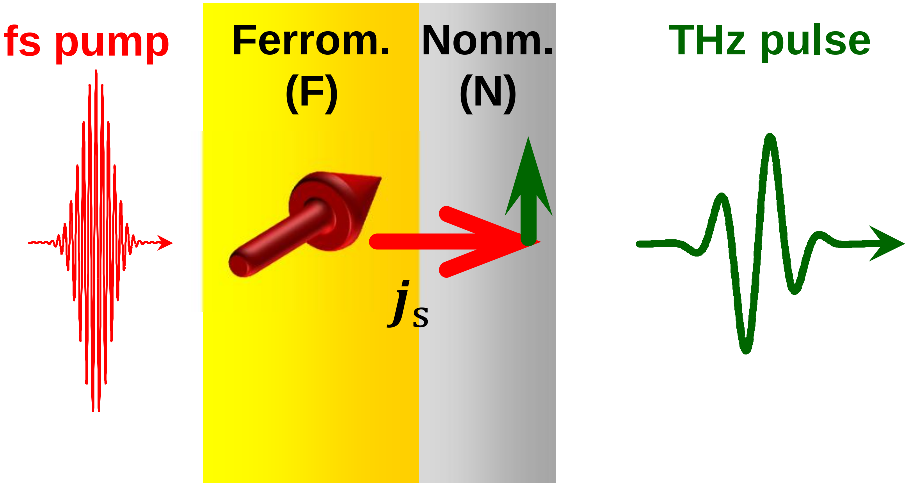
Weber, Hoppe, TK, Woltersdorf *et al.* (2020)




**Challenge:**
**How to further increase the THz field strength?**

# Challenge: More THz amplitude




**Maximize spin-to-charge-current conversion**

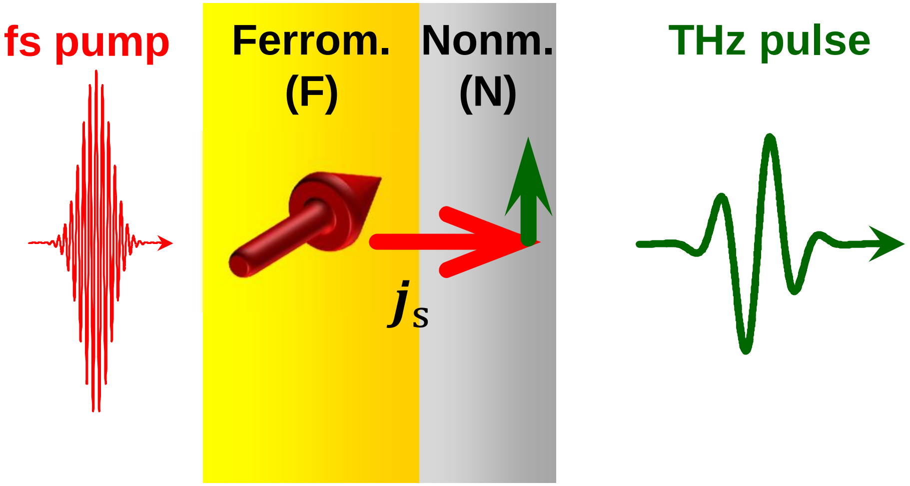
- N should have large ISHE and large spin diffusion length
- Interface engineering

**Maximize spin current for given pump energy**

- Understand how the spin current is induced

**What does the driving THz spin current look like?**

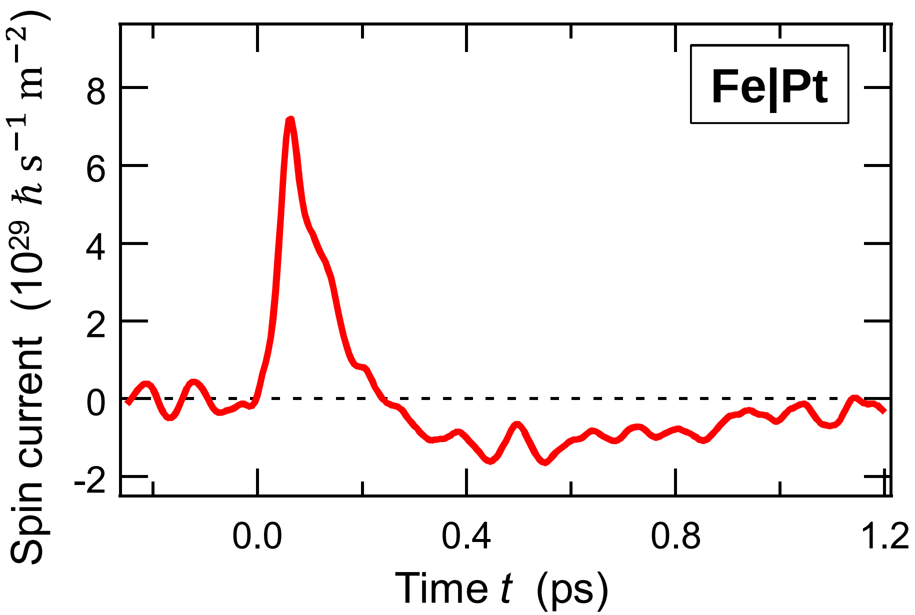
# Dynamics of the spin current




**Approach:** "Calculate back"

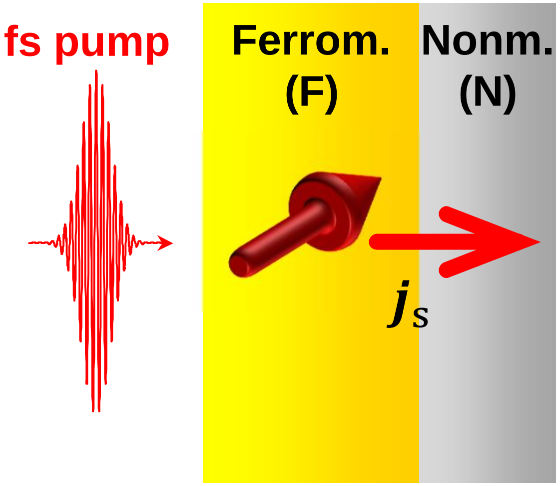
THz field → $j_s(t)$

Seifert, Barker, TK *et al.*,
Nature Commun. (2018)




- Extremely fast bipolar response
- What is the driving force?

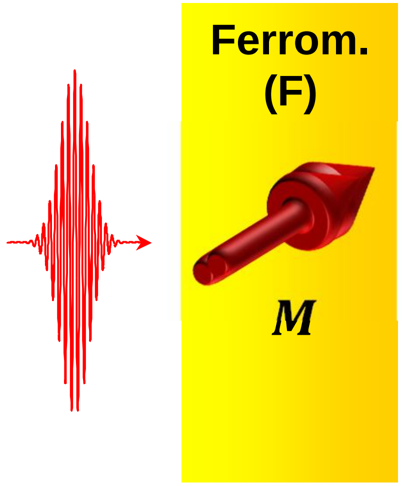
# Transport vs demagnetization




**What is the origin of the ultrafast spin transport?**

**Idea:**

Compare to dynamics of another fundamental process



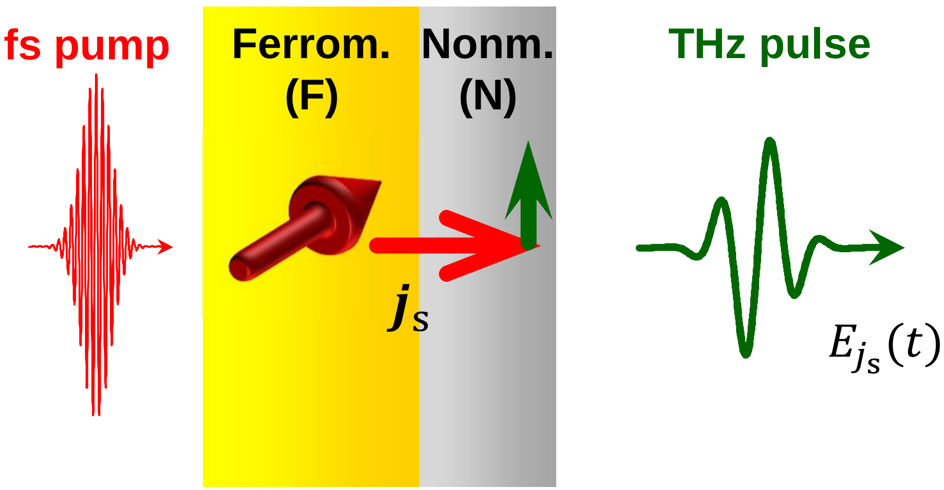

**Ultrafast local magnetization quenching of single ferromagnetic layers**

Beaurepaire *et al*., PRL (1996)

Is there a correlation with ultrafast transport in bilayers?

**Need to measure both processes in one experiment**

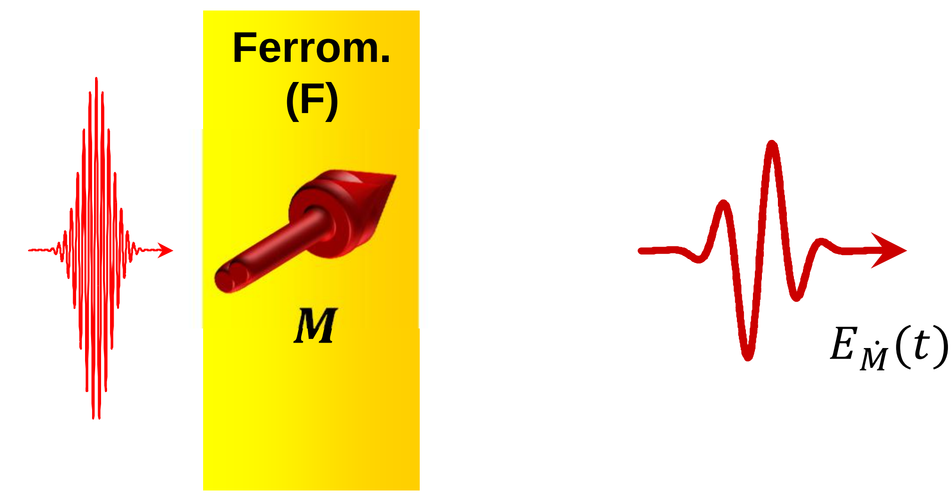
# Transport vs demagnetization




**Spin-to-charge current conversion**
⇒ Electric-dipole (ED) radiation

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

$$E_{j_s} \propto j_s$$



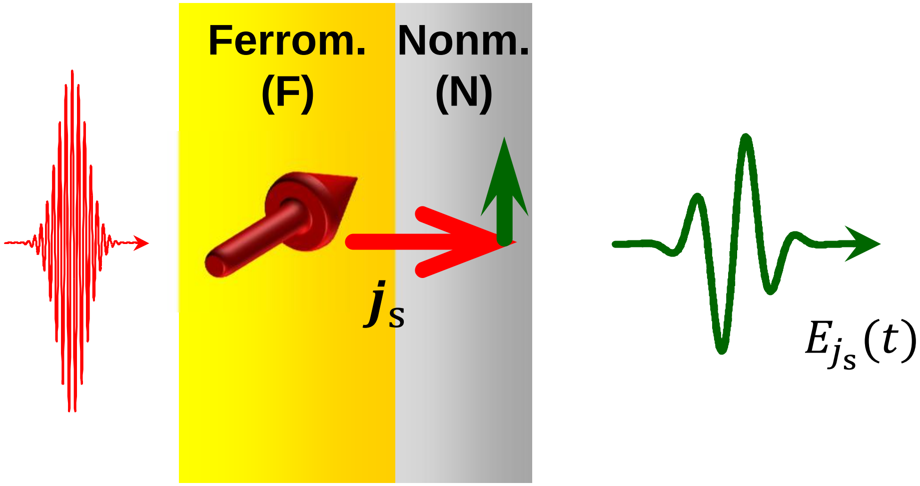

**Ultrafast magnetization quenching**
⇒ Magnetic-dipole (MD) radiation

Beaurepaire *et al.*, APL (2004)

$$E_{\dot{M}} \propto \dot{M} = \partial M/\partial t$$

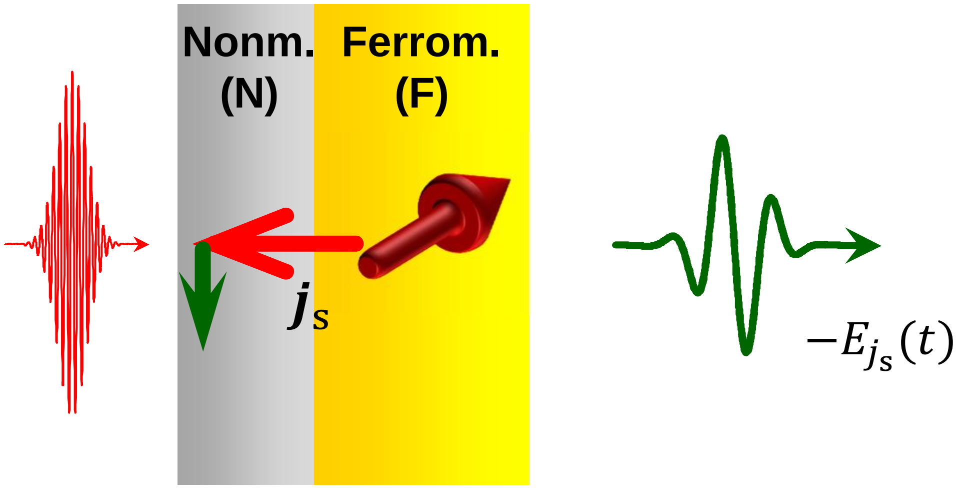
**How to isolate the weak $\dot{M}$ signal?**

# $j_s$ vs $\dot{M}$: Exploit symmetry




**Flip sample about $M$**

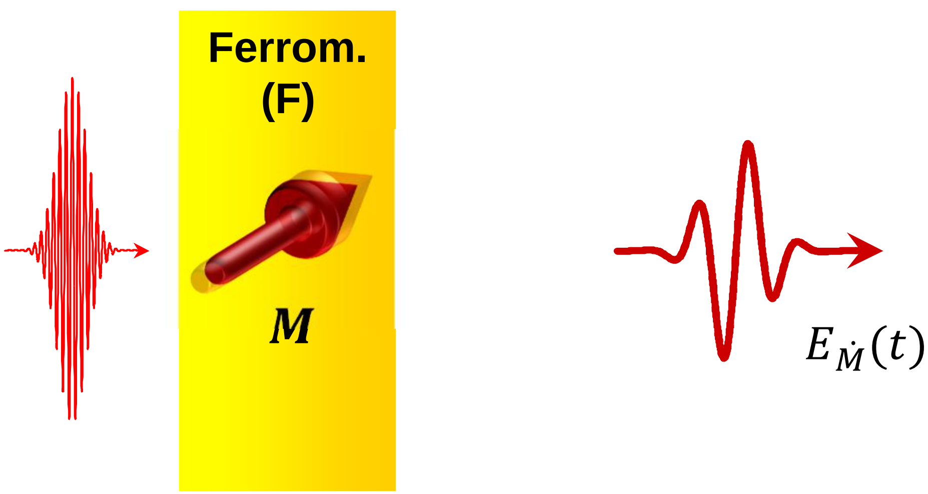
# $j_s$ vs $\dot{M}$: Exploit symmetry




**Flip sample about $M$**

- THz field due to $j_s$ changes sign
- Has "odd" symmetry

Beaurepaire *et al*., Ultrafast Phen. (2004)
Huisman, Kimel *et al*., PRB (2015)

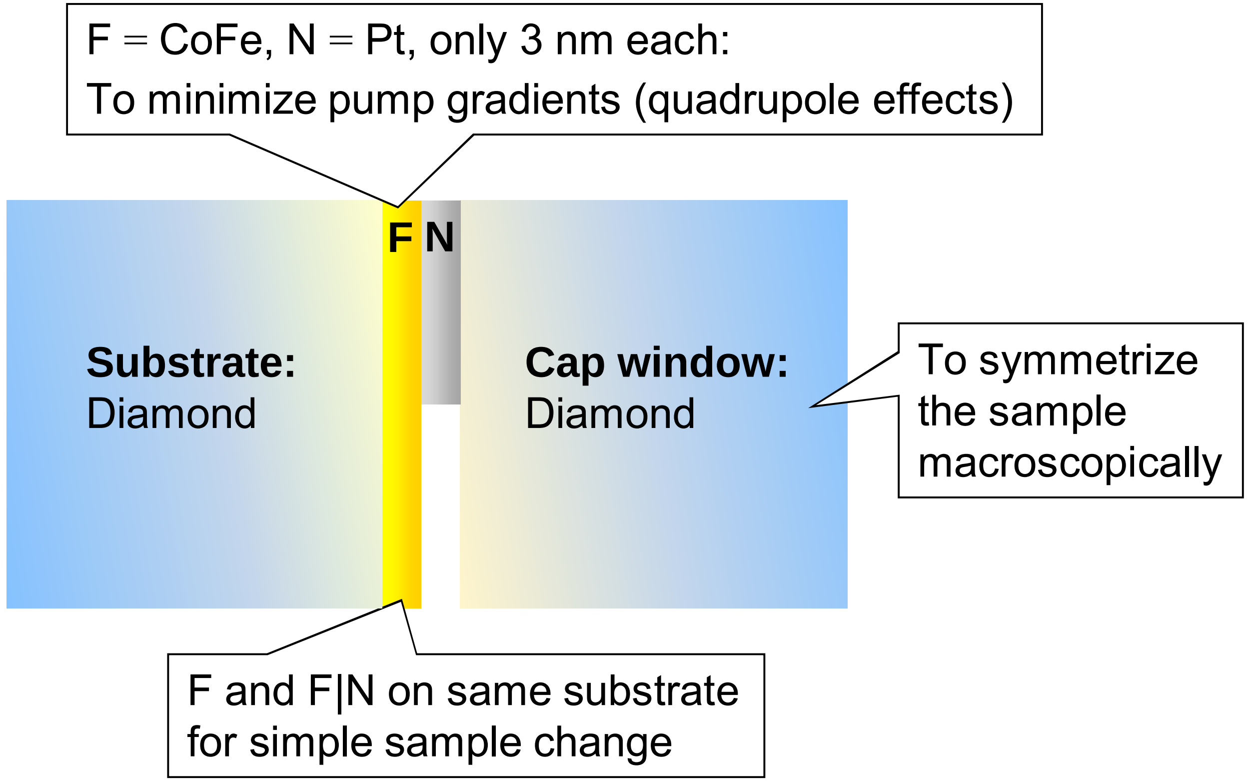



**Flip sample about $M$**

- $\dot{M}$ emission does not change
- Has "even" symmetry
- A potential even signal is hopefully small

**Direct way to measure $j_s$ and $\dot{M}$ in same setup with ~10 fs resolution**

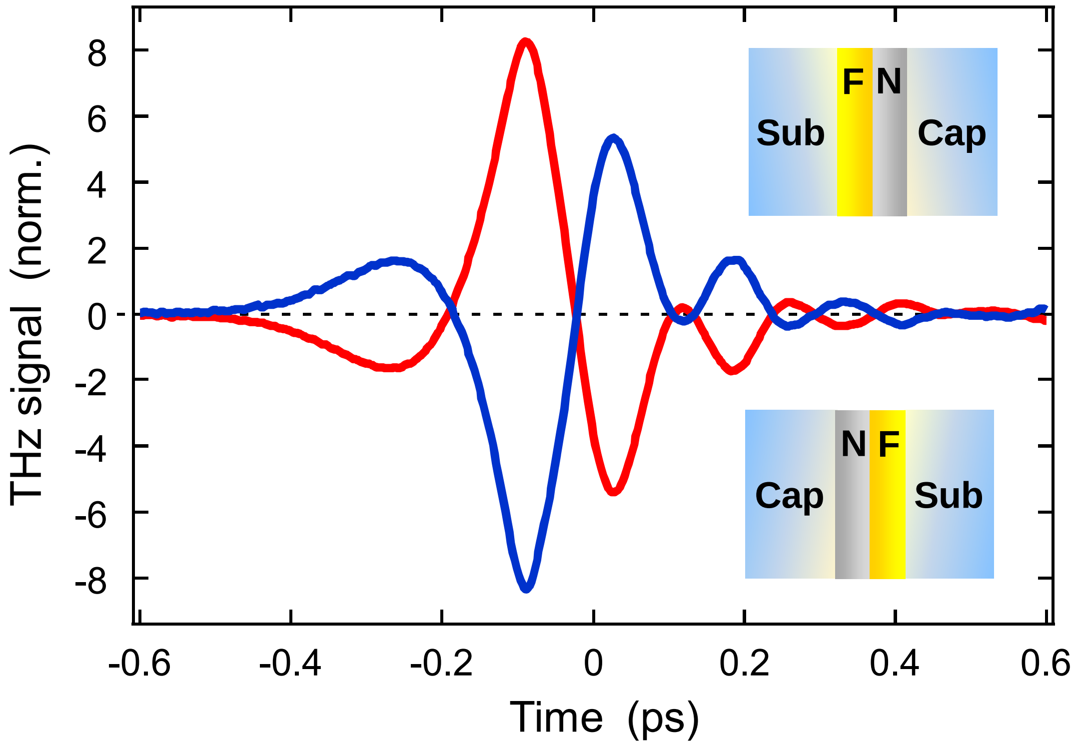
# Sample details




**Pump pulses:** From Ti:sapphire oscillator (10 fs, 800 nm, 1 nJ, 80 MHz)

**Start with F|N stack**

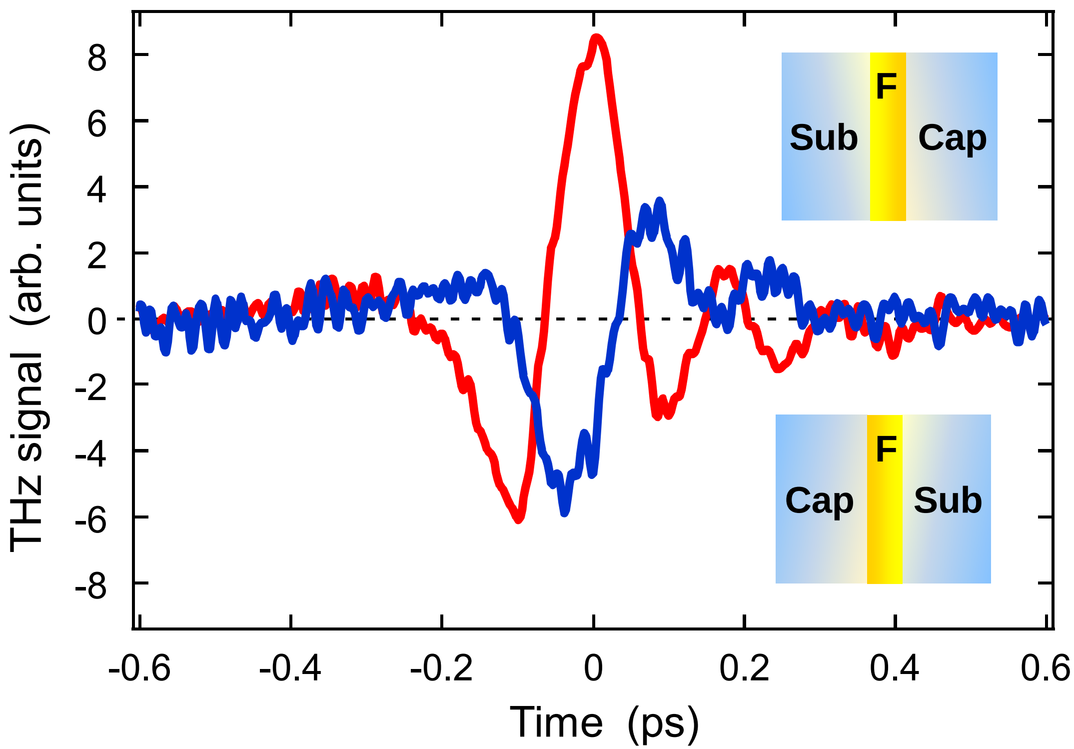
# THz emission from F|N stack




- $\approx$ 100% signal reversal $\Rightarrow$ Even symmetry
- Thus: Emission $E_{j_s}(t) \propto j_s(t)$ is dominant
- Use as reference for proper alignment

**Let's go for the single F layer**

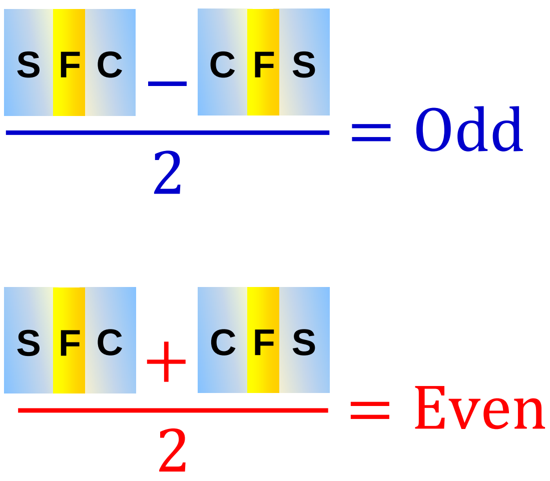
# THz emission from single F layer

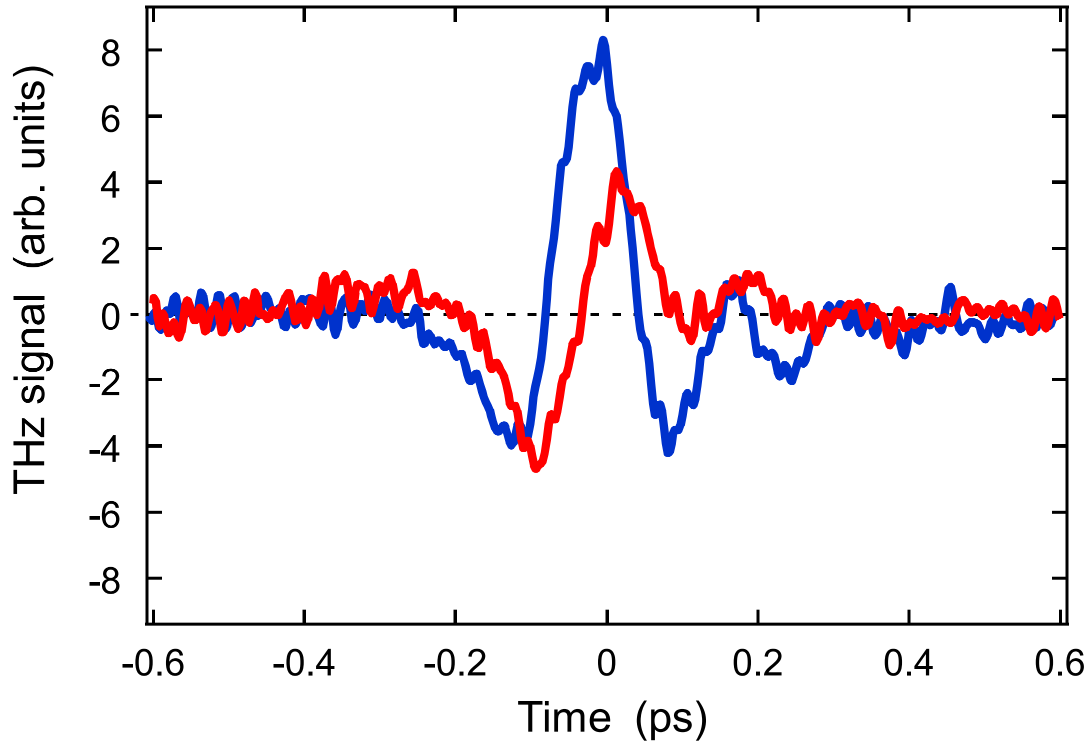



- Signal is ~170 times smaller than from F|N
- Nontrivial signal change upon sample reversal
- Indicates both even ($\dot{M}$) and odd signals contribute

**Separate even and odd contributions**

# F monolayer: Odd and even emission



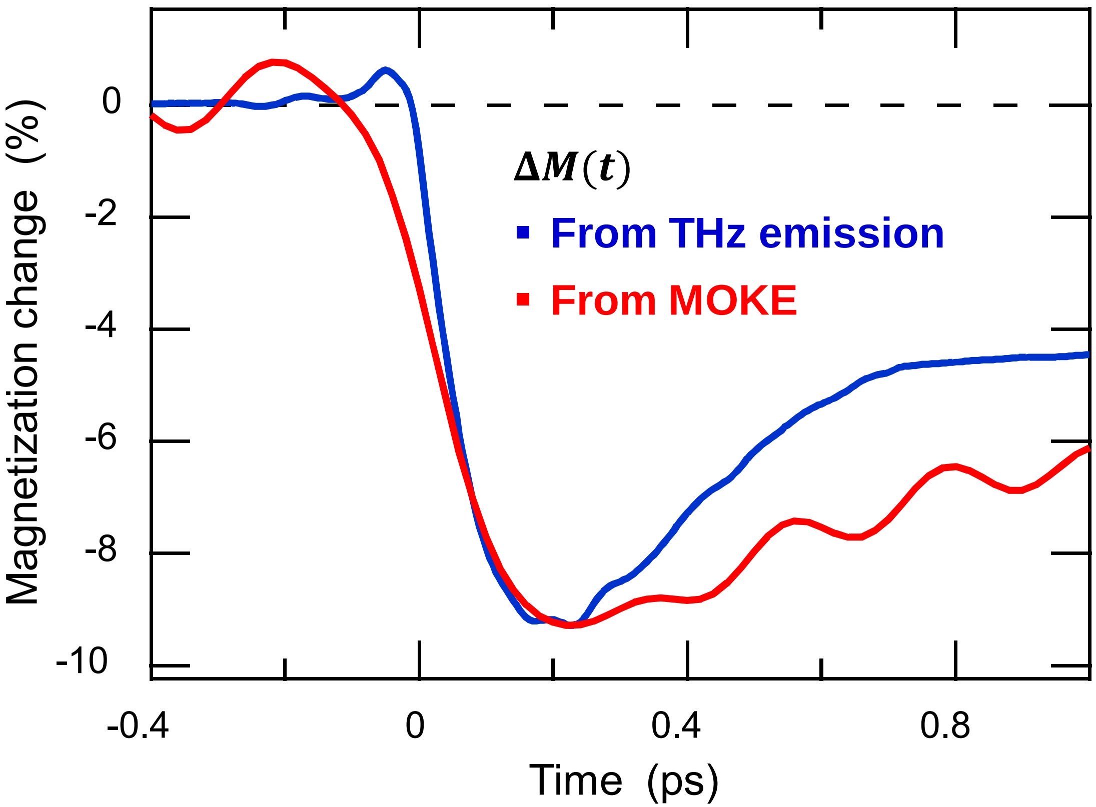

**Odd signal: Considerable**

- Origin not fully clear: Due likely to film inhomogeneities along growth direction

**Even signal: We assign it to ultrafast demagnetization, $E_{\dot{M}} \propto \dot{M}$**

- Signal has right polarity and order of magnitude
- Extract $M(t)$ and check temporal dynamics

# F monolayer: $M(t)$ dynamics




**Approximate agreement of**

- THz-extracted $M(t)$
- $M(t)$ from MOKE

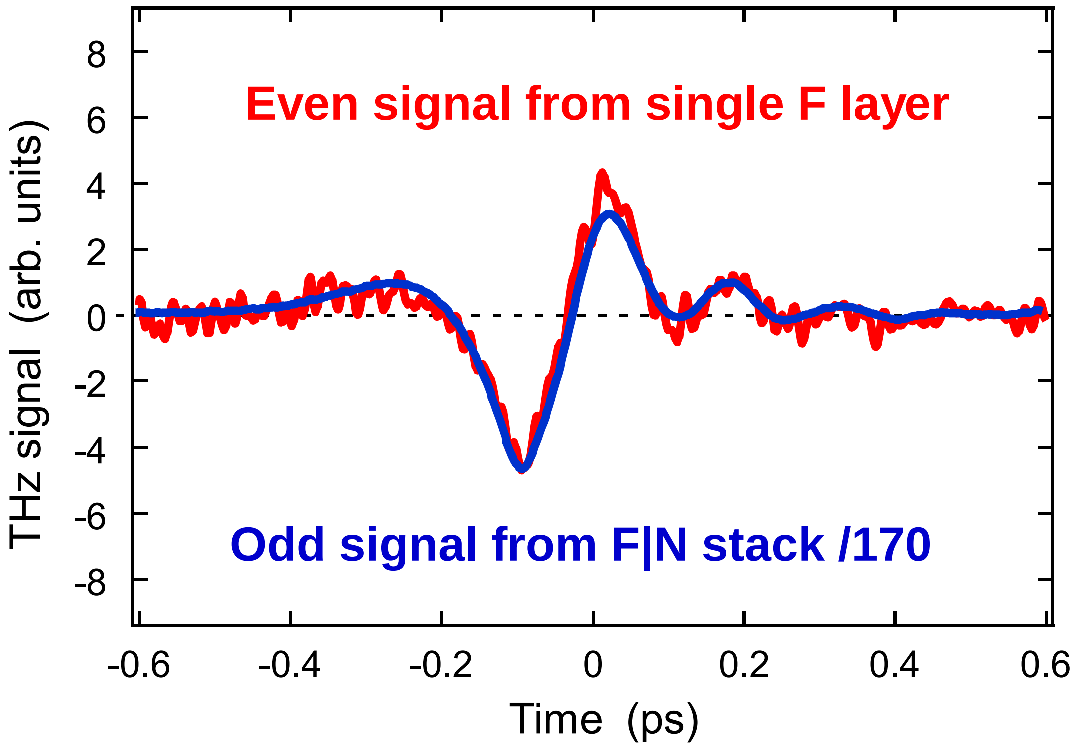
Huisman, Kimel *et al.*, PRB (2015)

Razdolski, Bovensiepen, Melnikov *et al.*, J. Phys. D (2017)

The even signal from the F single layer arises from ultrafast demagnetization

**Now compare: Even signal from F ↔ Odd signal from F|N**

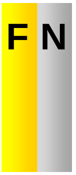
# Even from F vs odd from F|N




Rouzegar, Brandt, Reiß, Brouwer, Woltersdorf, TK *et al.* (2020)

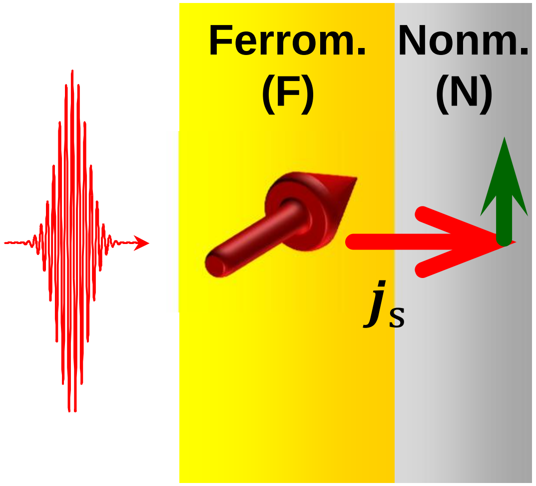
**Even emission from single F layer** $\propto$ **Odd emission from F|N stack**


$$E_{\mathrm{even}}^{\mathrm{F}}(t) \propto E_{\mathrm{odd}}^{\mathrm{F|N}}(t)$$

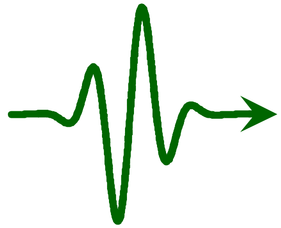


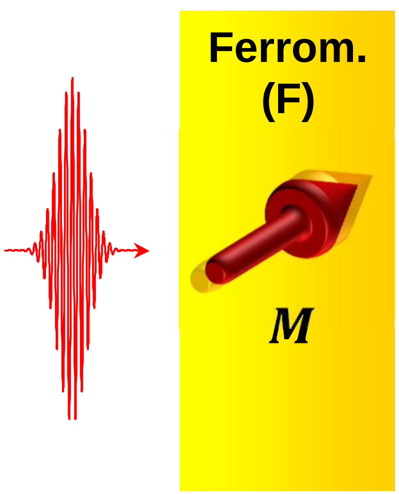

**What does this imply?**

# Transport vs demagnetization

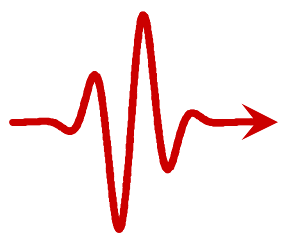



$E_{\mathrm{odd}}^{\mathrm{F|N}}(t) \propto j_s^{\mathrm{F|N}}$






$E_{\mathrm{even}}^{\mathrm{F}} \propto \dot{M}^{\mathrm{F}}$



**Our observations indicate:**

Demagnetization rate of F

∝ Spin current F→N in F|N

$$j_s^{\mathrm{F|N}} \propto \dot{M}^{\mathrm{F}}$$

**Confirmed for**

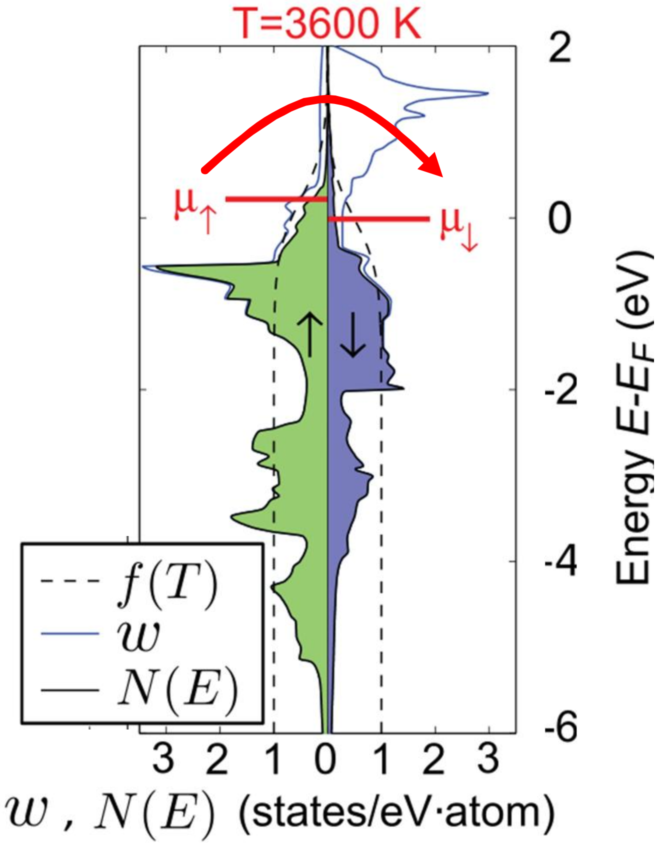
- N = Pt and W
- F = $Co_{70}Fe_{30}$ and $Ni_{80}Fe_{20}$

1) Ultrafast spin transport and
2) Ultrafast demagnetization

**are driven by same force**

**How to explain?**

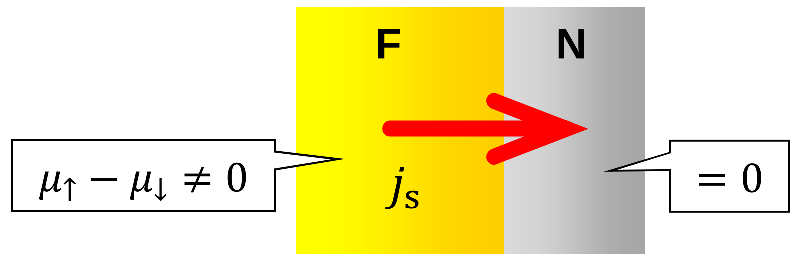
# Interpretation: Driven by $\mu_\uparrow - \mu_\downarrow$




**Spin-dependent Seebeck effect**

Bauer, Saitoh, Wees, Nature Mat. (2013)

$j_s^{\mathrm{F|N}} \propto \mu_\uparrow - \mu_\downarrow$




**Rethfeld *et al.*:**

$\mu_\uparrow - \mu_\downarrow$ drives spin flow from ↑ to ↓ channel

PRB **90**, 144420 (2014)

$\dot{M}^{\mathrm{F}} \propto \mu_\uparrow - \mu_\downarrow$

**Acremann *et al.*:**

Pump-induced heating of F layer causes $\mu_\uparrow \neq \mu_\downarrow$

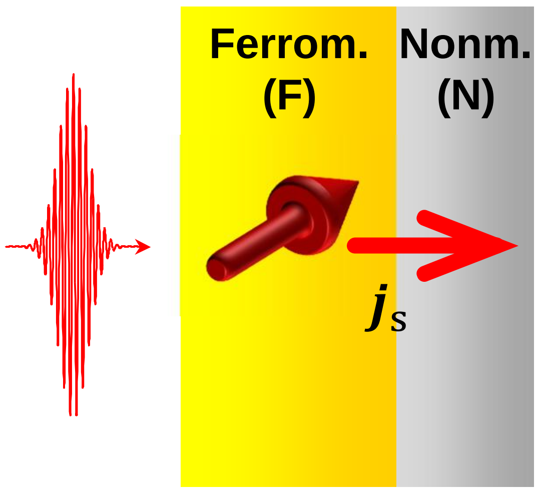
J. Phys. Cond. Mat. (2017)

**Consistent with our observations**

$\dot{M}^{\mathrm{F}} \propto j_s^{\mathrm{F|N}}$

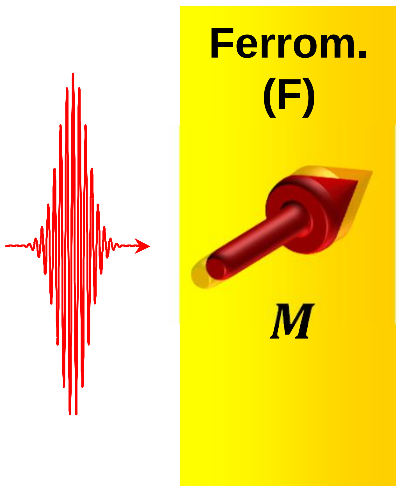
Can be extended to nonthermal states

# Summary




1) Ultrafast spin transport and
2) Ultrafast demagnetization

**are driven by the same force**




**Significance:**

- We can apply all knowledge of 2) to 1)
- Will guide us to maximize spin current from F to N
- Can use N layer as an ultrafast detector of $\mu_\uparrow - \mu_\downarrow$

# Summary

**THz spin transport**

- Enables interesting applications, e.g. THz wave generation, THz spin torque
- First temporal steps of spintronic effects

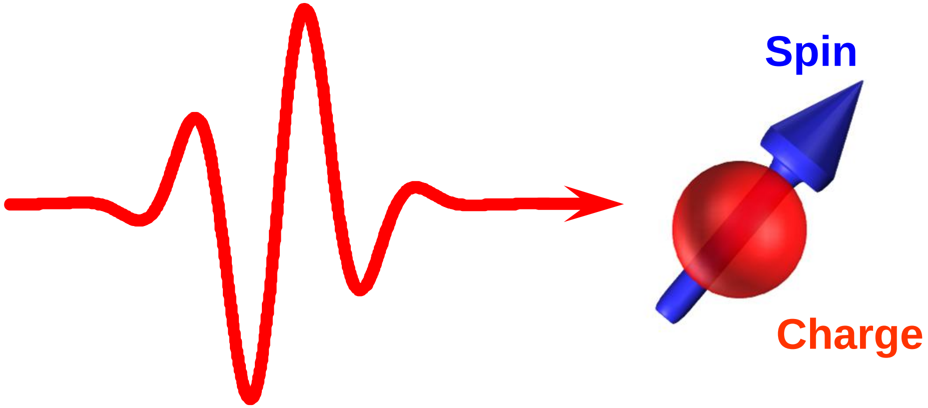
**New insights**

- Ultrafast demagnetization and spin transport are driven by same force

**Future directions**

- Use THz currents to apply spin torque

**THz radiation is a useful tool to probe and control spin dynamics**




**Interested in a project in in THz spintronics?**

→ Contact Tobias Kampfrath