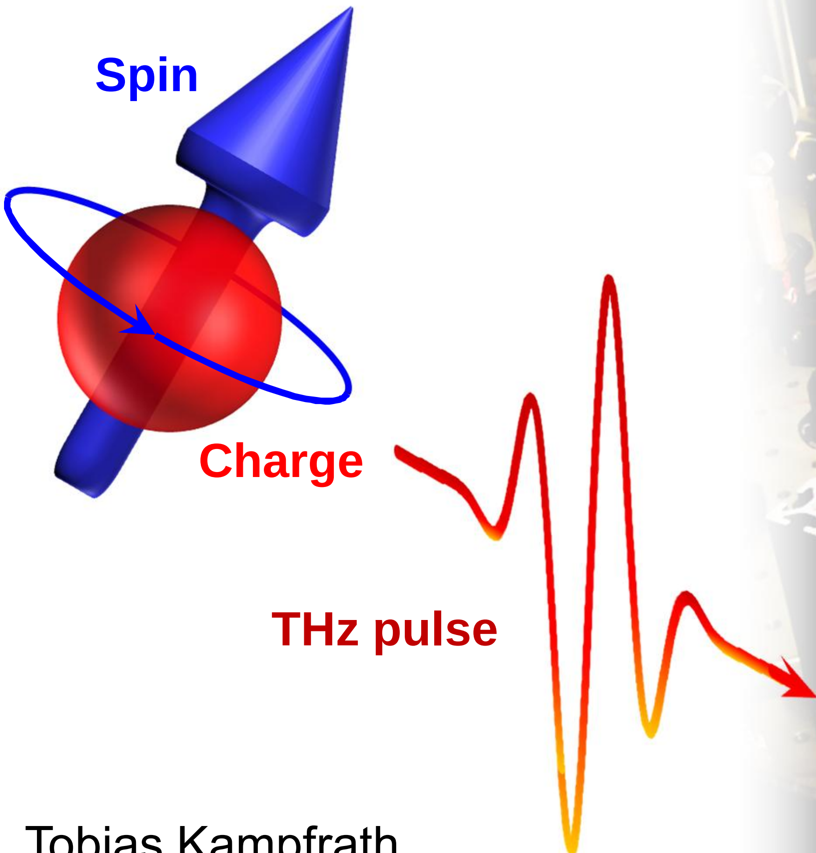
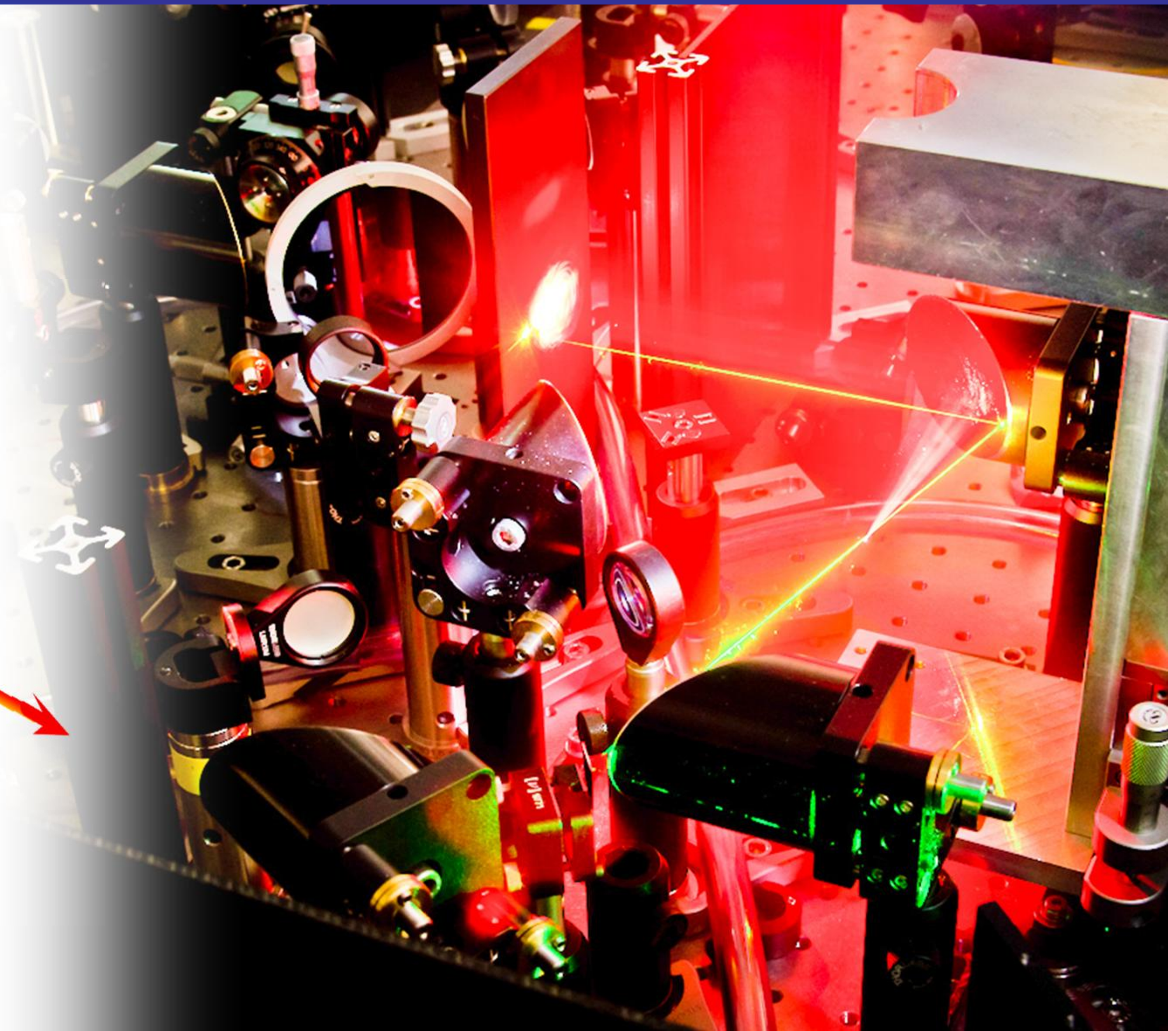
Probing and controlling spin dynamics with THz pulses





Tobias Kampfrath
Freie Universität Berlin
and FHI/Max Planck Society

PhD students: organize your own symposium

… at the 2019 Spring Meeting of the German Physical Society in Regensburg!

- Your chance to implement a symposium you always wanted to attend
- To get in personal contact with leading scientists at an early stage of your career

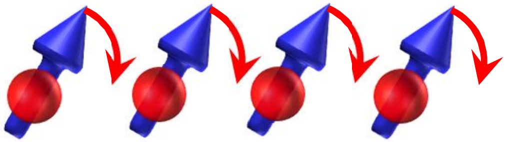
How?

- Pick 1-5 colleagues as organization committee and fill out the online application
  http://www.dpg-physik.de/dpg/gliederung/junge/profil/ateam/wissenschaftlich/tagungen/2019/phd-symposien/announcement.html
- Timely topic related to magnetism
- Invite speakers, compile the program, organize the day
- Deadline: October 15

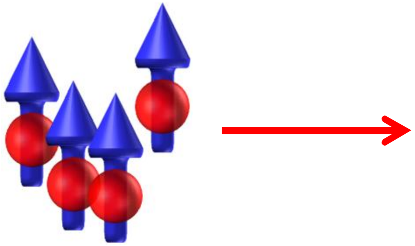
2018	Ultrafast Spin-Lattice interactions
2017	Quantum Magnets: Frustration and Topology in Experiment and Theory
2015	Quantum phase transitions: Emergent phenomena beyond elementary excitations
2014	Magnon plasmonics
2013	Topological defects in magnetic materials: From devices to cosmos
2012	Spintronics on the way to modern storage technology

Three elementary spin operations

1. **Turn spins around**



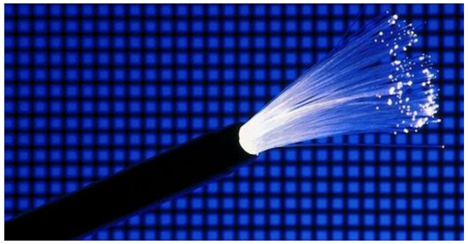
2. **Transport spins**



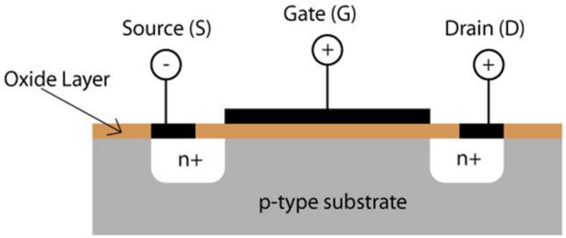
3. **Detect spin dynamics**

Goal: reach speed of other information carriers, i.e. THz bandwidth

Light in fibers: >10 Tbit/s



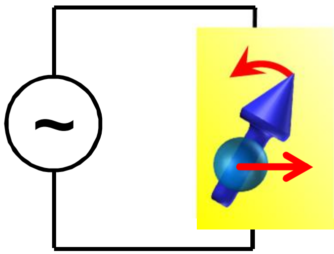
Electrons in a FET: ~1 THz cut-off



How to manipulate magnetic order ultrafast? Two approaches

Spintronics and femtomagnetism

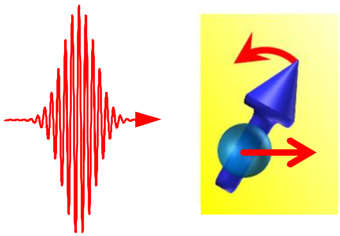
Spintronics: voltages in circuits



- Bandwidth <10 GHz
- Force/torque ∝ applied field

See e.g. Magnetism Roadmap (2017)

Femtomagnetism: fs light fields



- Freq. ~400 THz ⇒ Need rectification
- Force/torque ∝ light intensity

Kirilyuk, Kimel, Rasing, Rev. Mod. Phys. (2010)

DC

0.01 THz

100 THz

1000 THz

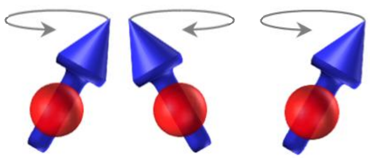
Terahertz gap
1…30 THz
4…120 meV

THz fields + magnetism = useful?

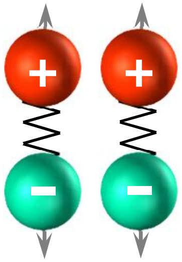
Why THz magnetism?

1) **Reveal speed and initial elementary steps of spintronic effects**
   E.g. spin-Hall, spin-Seebeck and GMR

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



Magnons

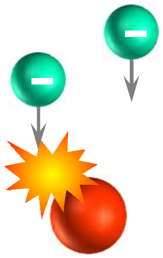


Phonons

Bound electron states:

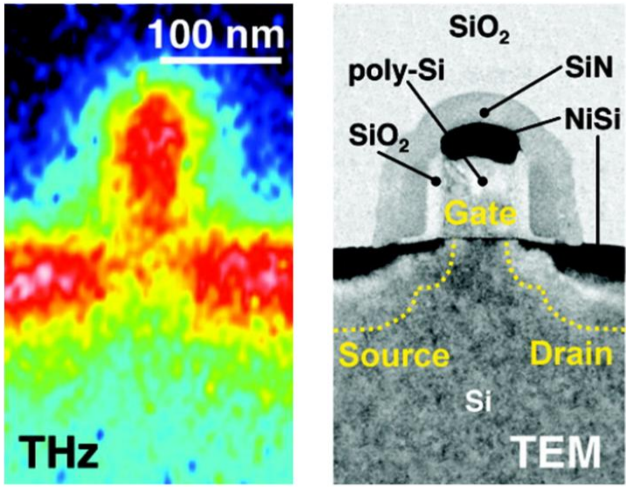
- Cooper pairs
- Excitons

1 THz ≙ 4 meV



Intraband transport

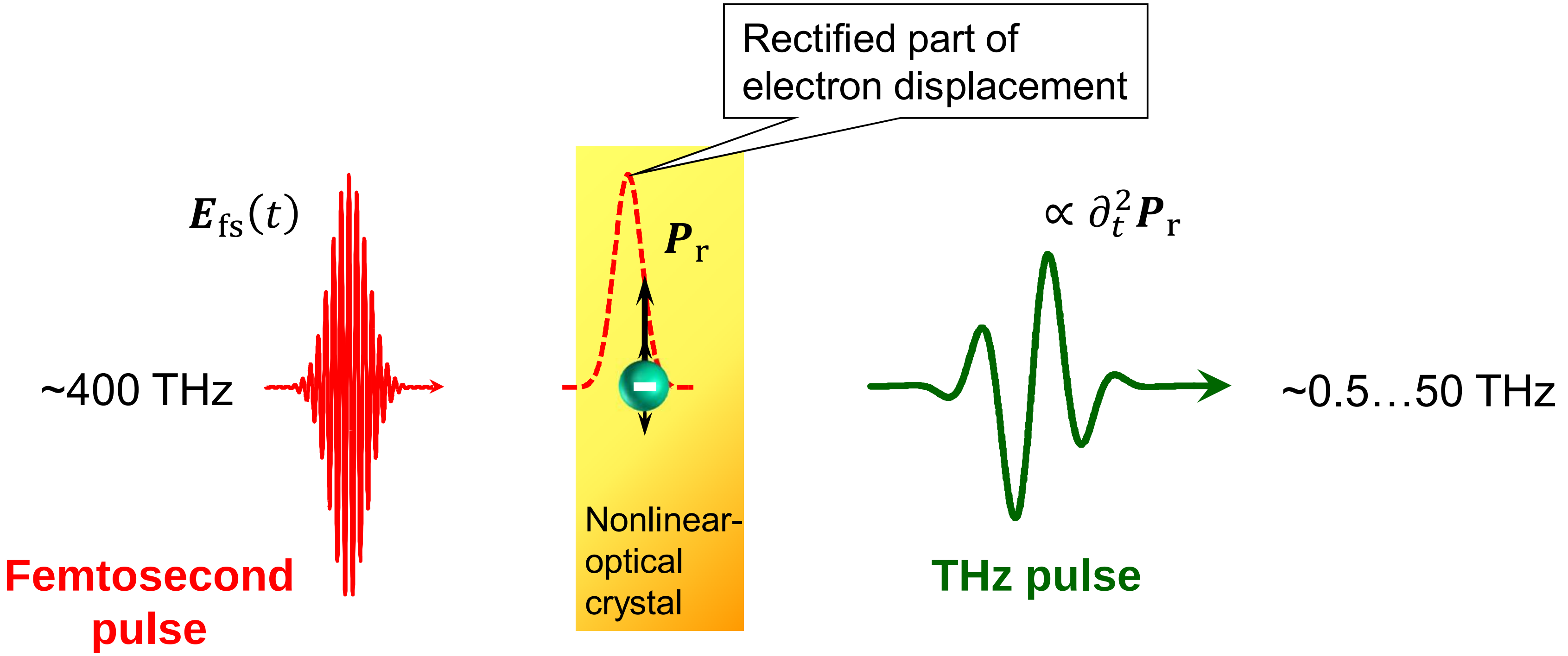
3) **Reward for THz technology**
   e.g. THz sources and modulators for spectroscopy and imaging



Hillenbrand *et al.*, Nano Lett. (2008)

How to get THz pulses?

Intense THz pulses by optical rectification



Nonlinear electron displacement

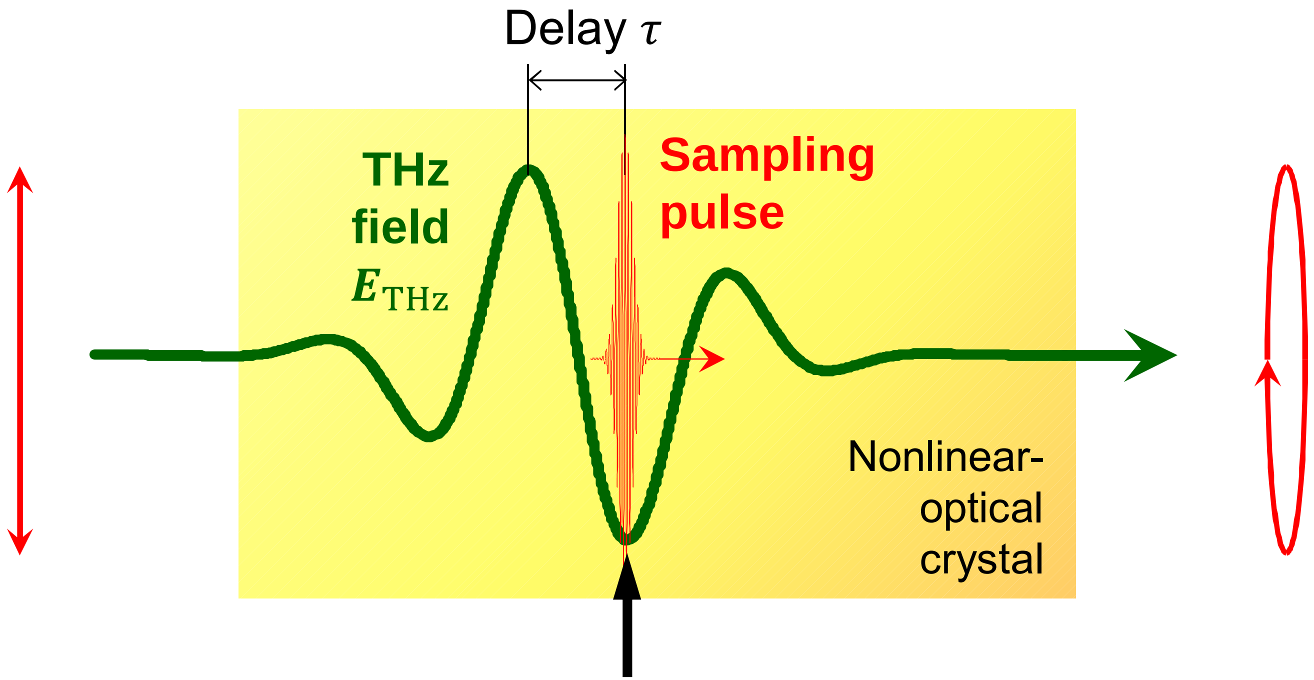
$P_r \propto E_{fs}^2(t)$

$\propto$ 2nd harmonic + |envelope|^2

How to detect the THz pulse?

Reviews: Hoffmann, Fülöp, J. Phys. D (2011); Reimann, Rep. Prog. Phys. (2007)

THz detection: electro-optic sampling



Wu, Zhang, APL (1995)

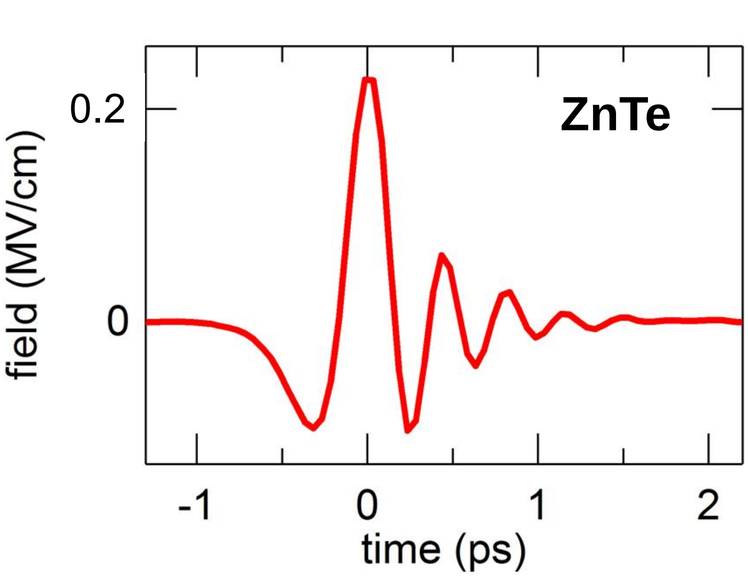
Electrooptic effect:

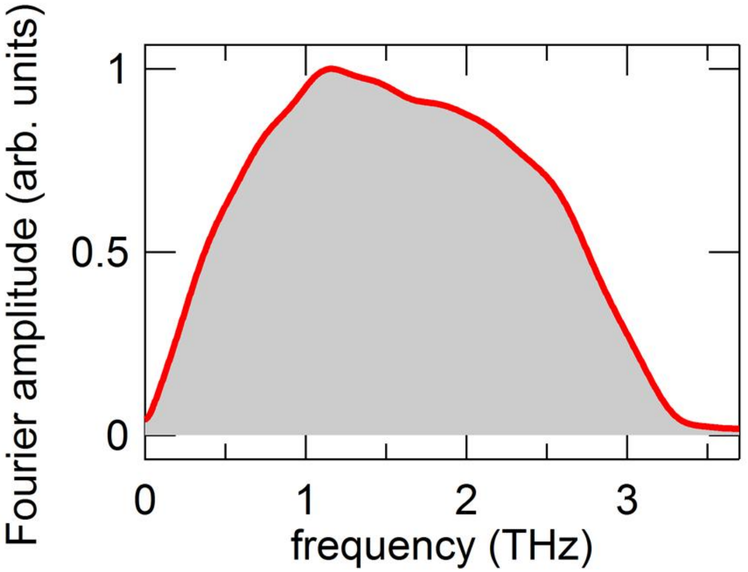
Change in refractive index $\propto \boldsymbol{E}_{\text{THz}} \Rightarrow$ Crystal becomes birefringent

Scan ellipticity of sampling pulse vs $\tau \Rightarrow$ Get THz electric field $\boldsymbol{E}_{\text{THz}}(\tau)$

A typical THz pulse…

Example of an ultrashort THz pulse





- Duration down to 50 fs
- Peak fields up to ~30 MV/cm (~10 T)
- Detection of *full transient field*, threshold down to 1 V/m

- Tunable center frequency 0.5…50 THz, i.e. 2…200 meV
- But: gaps between 5 and 15 THz

How to control magnetic order by THz fields?
Consider equation of motion of spins

How can one control spin dynamics?

Equation of motion of spin i

$$\partial_t \langle \boldsymbol{S}_i \rangle = -g\mu_{\mathrm{B}} \langle \boldsymbol{S}_i \times \boldsymbol{B}_{\mathrm{eff}i} \rangle$$

Total effective field acting on spin $\boldsymbol{S}_i$

In Heisenberg-type magnet

$$\boldsymbol{B}_{\mathrm{eff}i} = (\boldsymbol{B}_{\mathrm{ext}} + \boldsymbol{B}_{\mathrm{SSMD}i}) + \frac{\boldsymbol{v}_i}{c} \times \boldsymbol{E}_i + \sum_{j \neq i} J_{ij} \boldsymbol{S}_j$$

Zeeman coupling to external field + other spins

SO coupling to total electric field

Exchange coupling

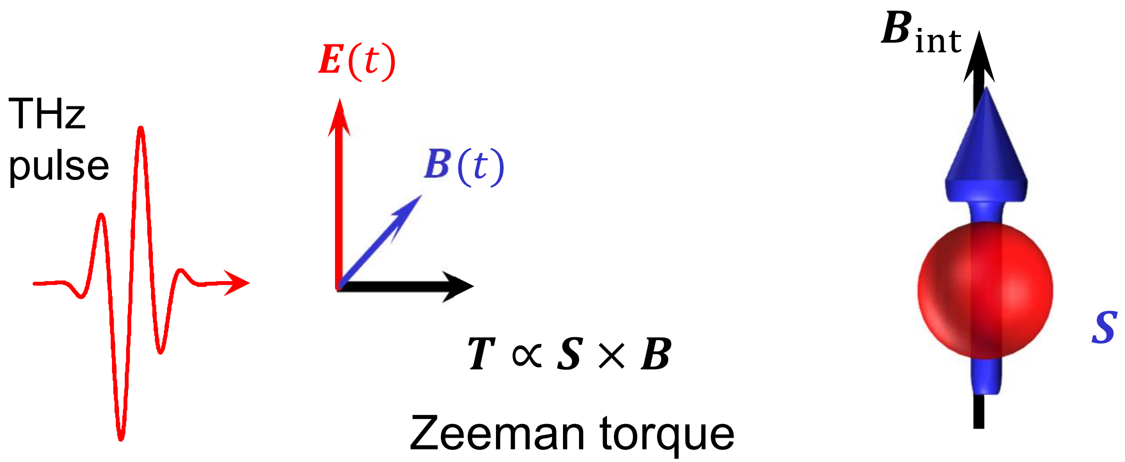
$\boldsymbol{B}_{\mathrm{eff}i}$ is *the* handle to (ultrafast) control over magnetic order

- Directly by external fields $\boldsymbol{B}_{\mathrm{ext}}$, $\boldsymbol{E}_{\mathrm{ext}}$ (→Kim)
- Indirectly by modulation of coupling parameters (e.g. J_{ij}) using light, currents, strain, heat, … (→Kirilyuk, Kalashnikova)

Start simple: Zeeman torque

How to control spins as fast as possible?

Most natural stimulus: magnetic field $\boldsymbol{B}(t)$

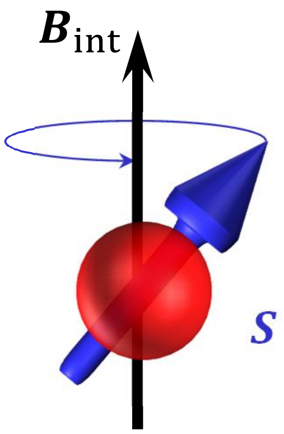


How to control spins as fast as possible?

Most natural stimulus: magnetic field $\boldsymbol{B}(t)$

Most efficient coupling on resonance

Larmor frequency $\hbar\omega_{\mathrm{L}} = g\mu_{\mathrm{B}}|\boldsymbol{B}_{\mathrm{int}}|$



Ferromagnets

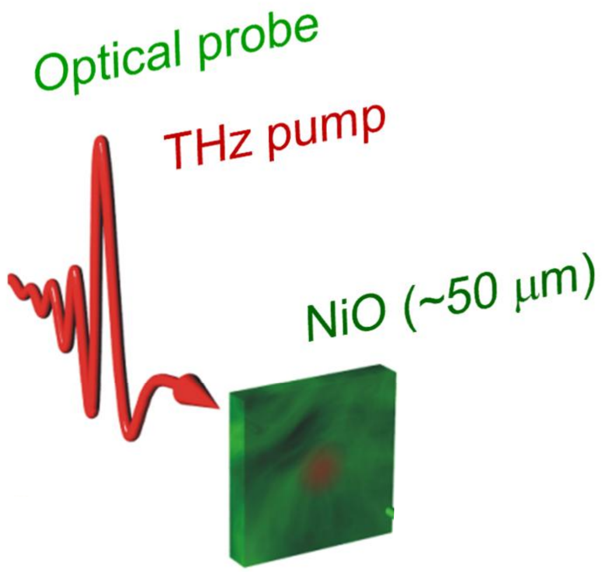
- ω_{L} determined by anisotropy field
- $\omega_{\mathrm{L}}/2\pi \ll 1$ THz

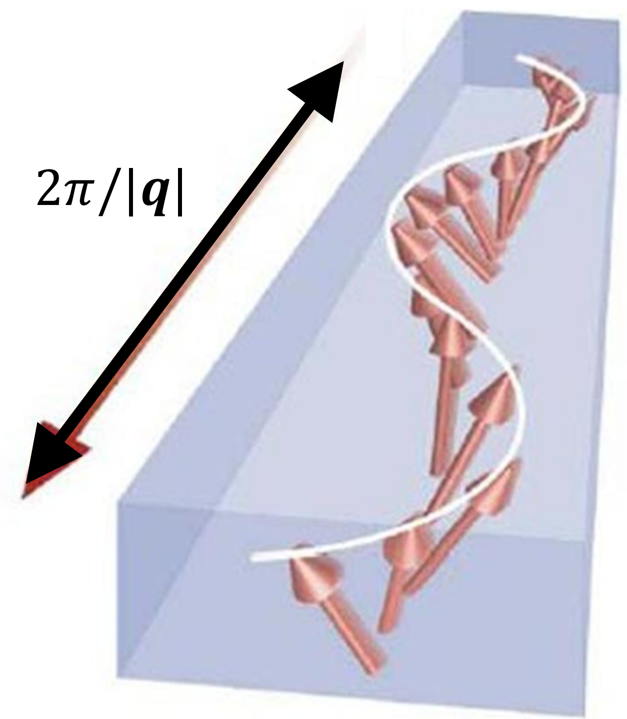
Antiferromagnets

- Exchange causes additional repulsion
- $\omega_{\mathrm{L}}/2\pi \sim 1$ THz

$\Rightarrow$ **Conduct a THz-pump magnetooptic-probe experiment**

THz magnetic pump – infrared probe





Detect Faraday rotation

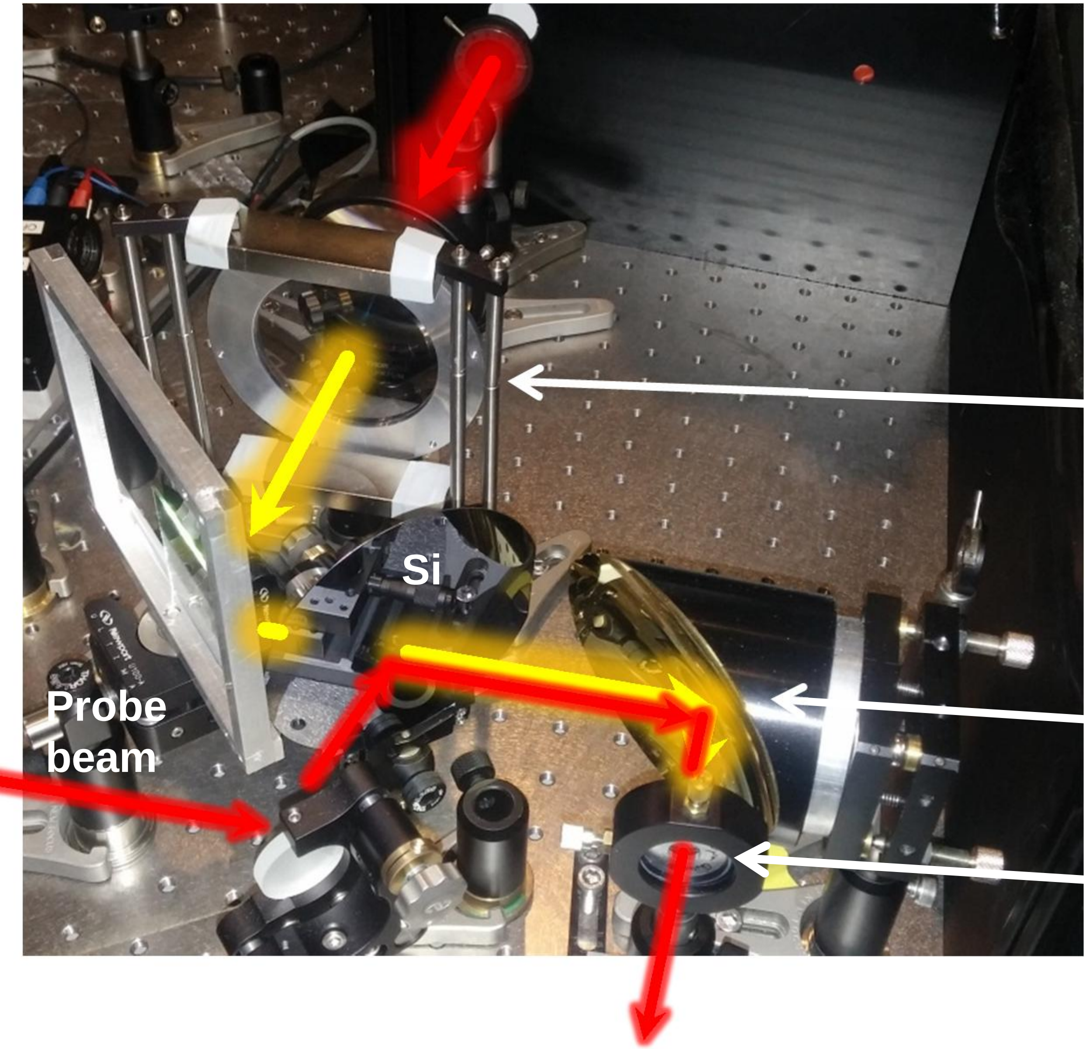
$\propto \boldsymbol{k}_{\mathrm{probe}} \cdot \boldsymbol{M}(t)$

Sample: antiferromagnetic NiO

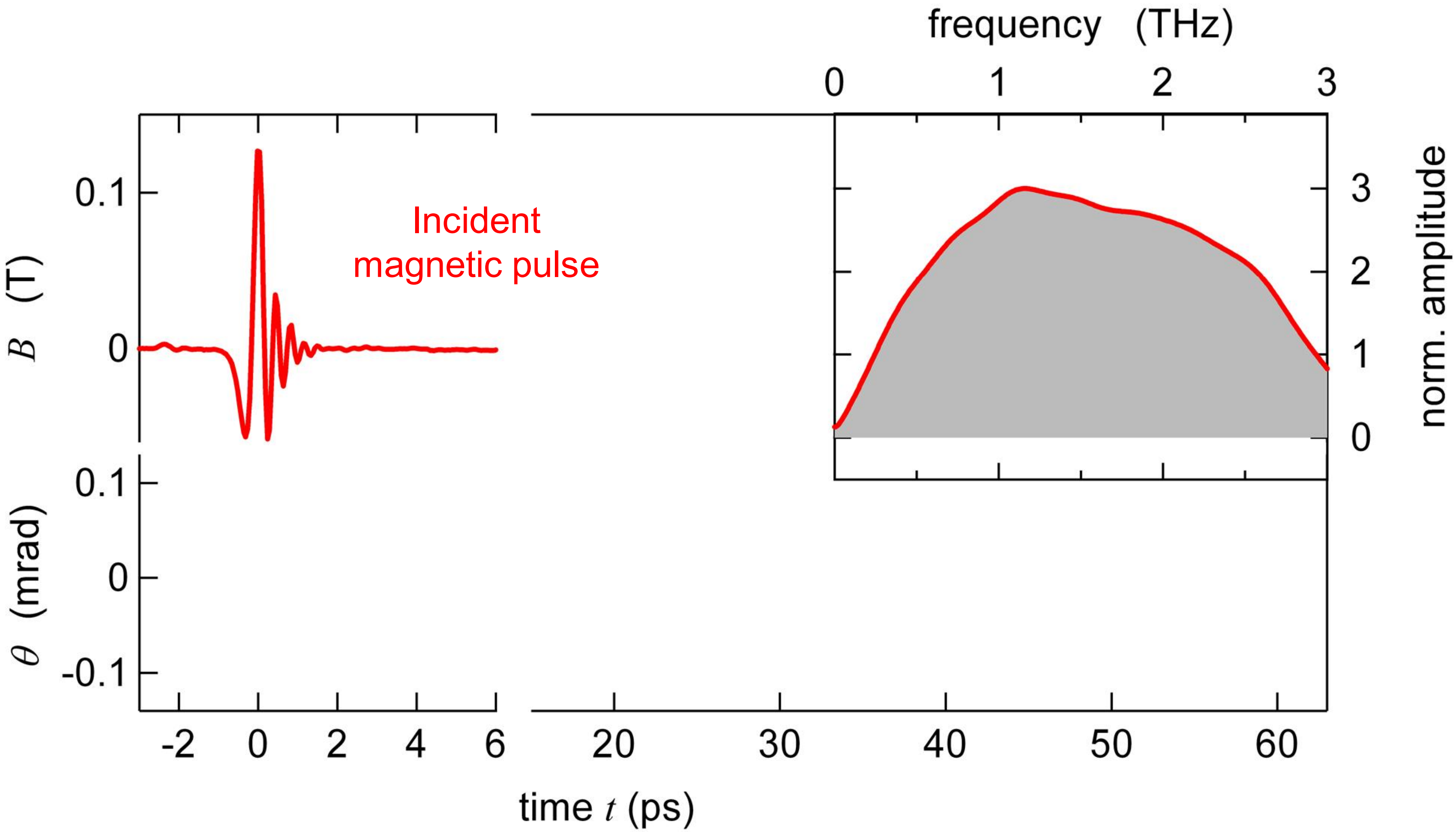
- Neel temperature 523 K
- Magnon ($\boldsymbol{q} = 0$) at 1 THz

In the lab…

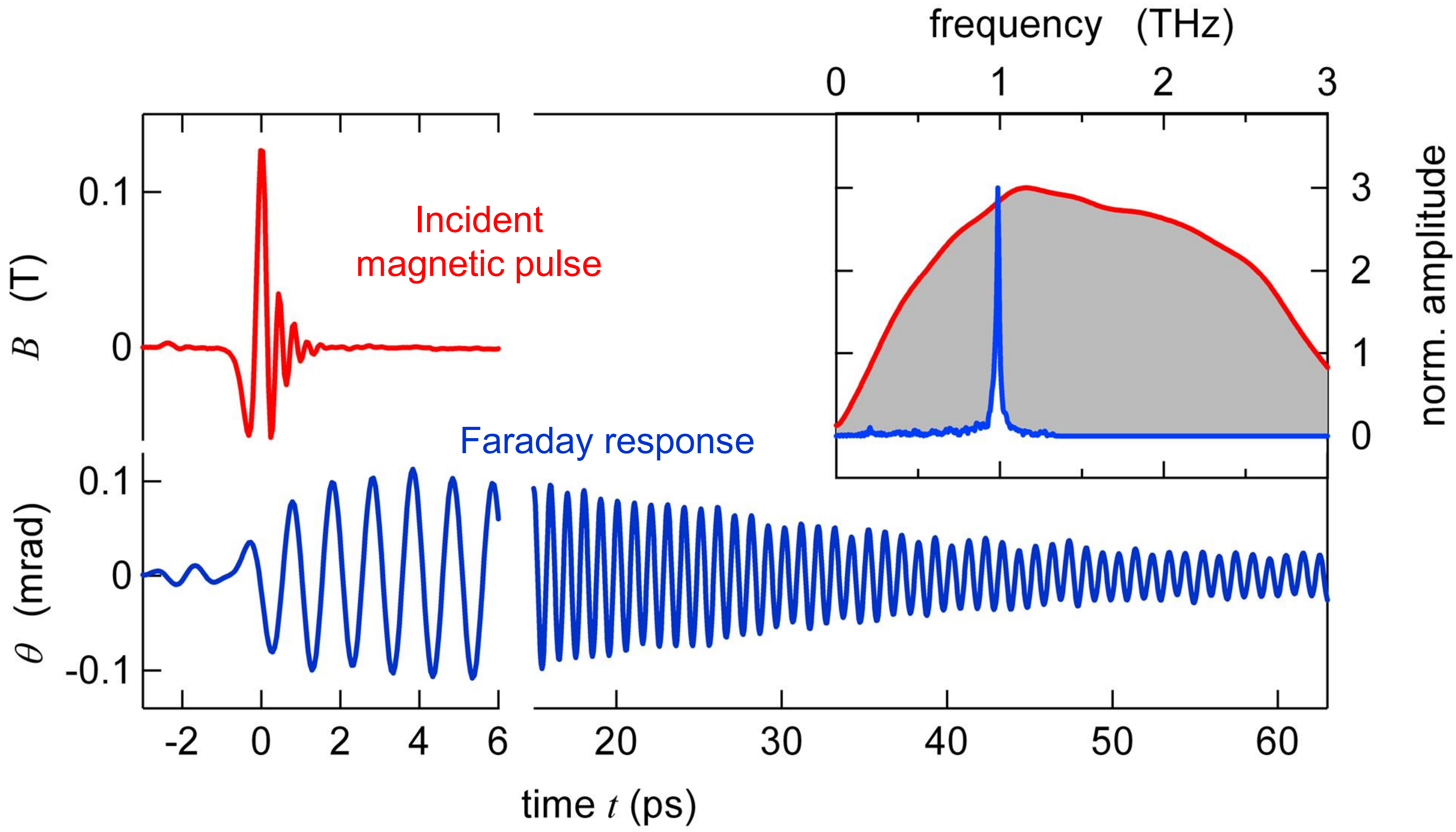
Simplistic THz setup in the lab



THz-induced magnon oscillation



THz-induced magnon oscillation



Oscillation at 1 THz, decay time ~40 ps $\Rightarrow$ Signature of $\boldsymbol{q} = 0$ magnon at 1 THz

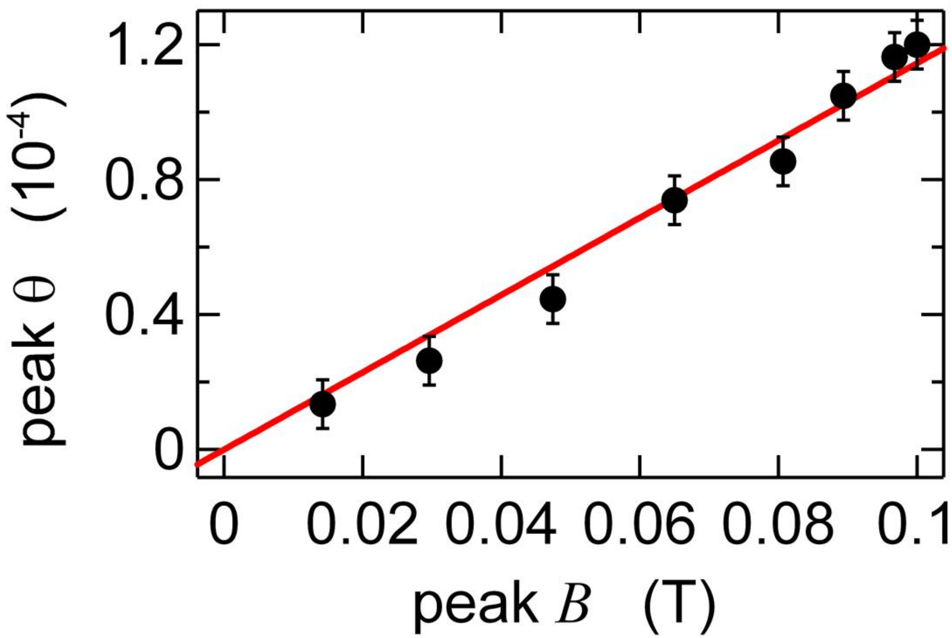
Driven by electric or magnetic field component?

The magnon is driven by the magnetic field

Observation:

Induced magnetization
$\propto$ driving field

Is $\boldsymbol{M} = C\boldsymbol{E}$ possible?

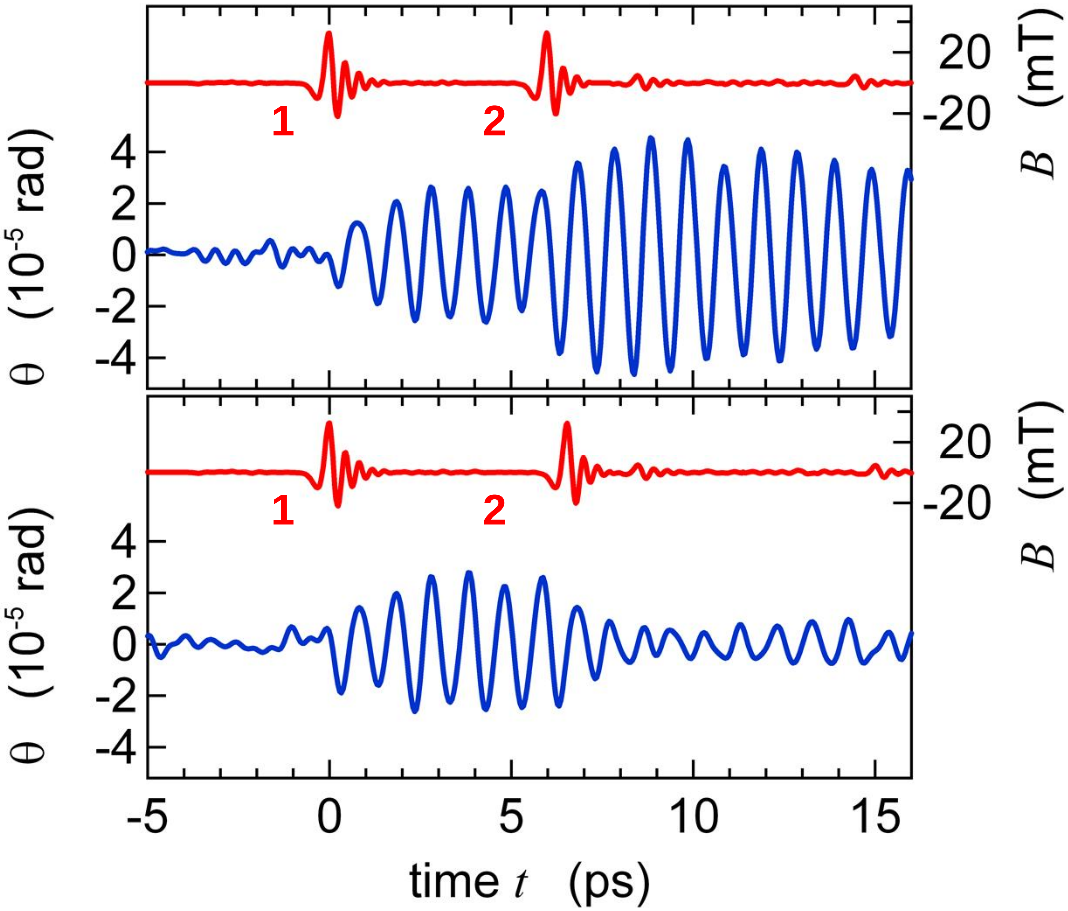


- NiO is centrosymmetric $\Rightarrow C = 0$
- No linear magnetoelectric effect in centrosymmetric NiO

Driving force is magnetic (not electric) field

Idea: use double pulses to control magnon amplitude

Coherent spin control with THz pulse pairs



Second pulse after 6 cycles: amplifies magnon

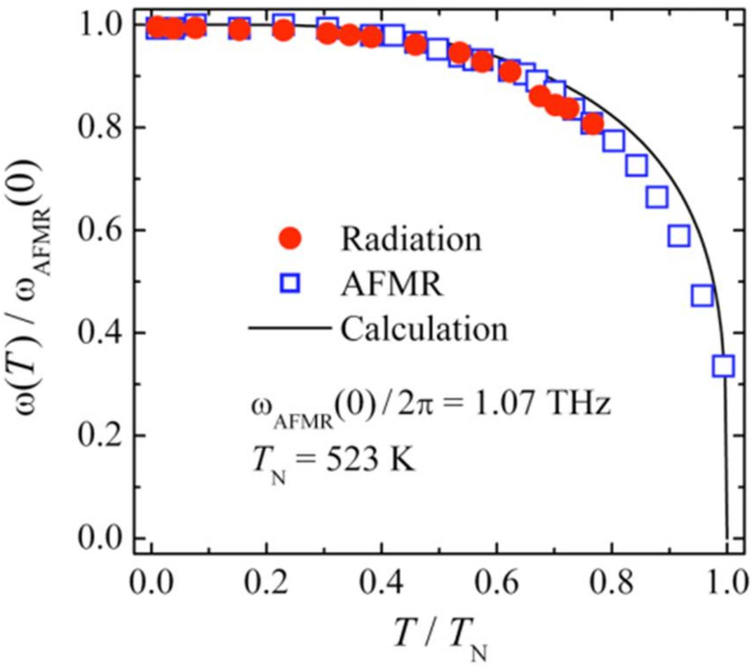
Second pulse after 6.5 cycles: switches magnon off

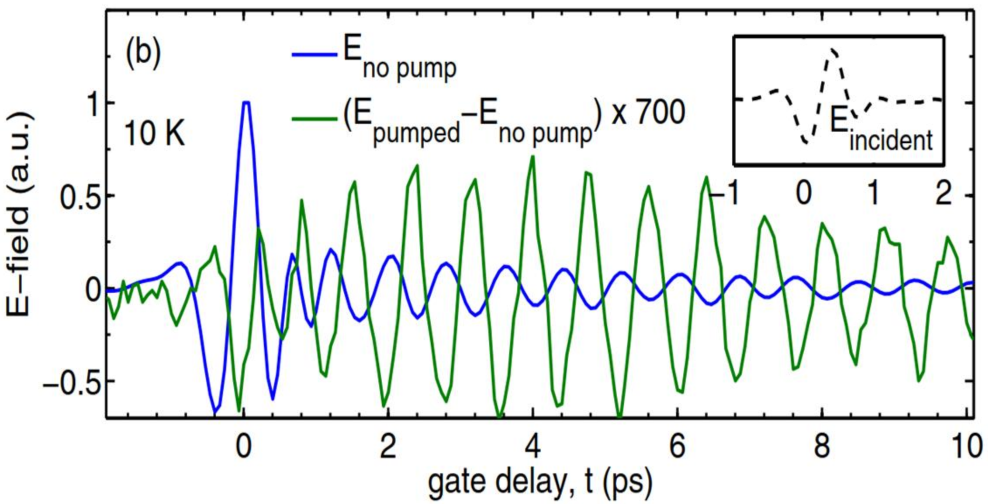
THz spin control is feasible by the simple Zeeman torque of THz magnetic pulses

Kampfrath, Sell, Fiebig, Wolf, Huber *et al.*, Nature Phot. (2011)
Baierl, Kampfrath, Huber *et al.*, PRL (2016)

Interesting application: THz magnon spectroscopy

THz magnon spectroscopy





- Characterization of antiferromagnets
- Magnons probe $B_{\text{exch}}B_{\text{ani}}$

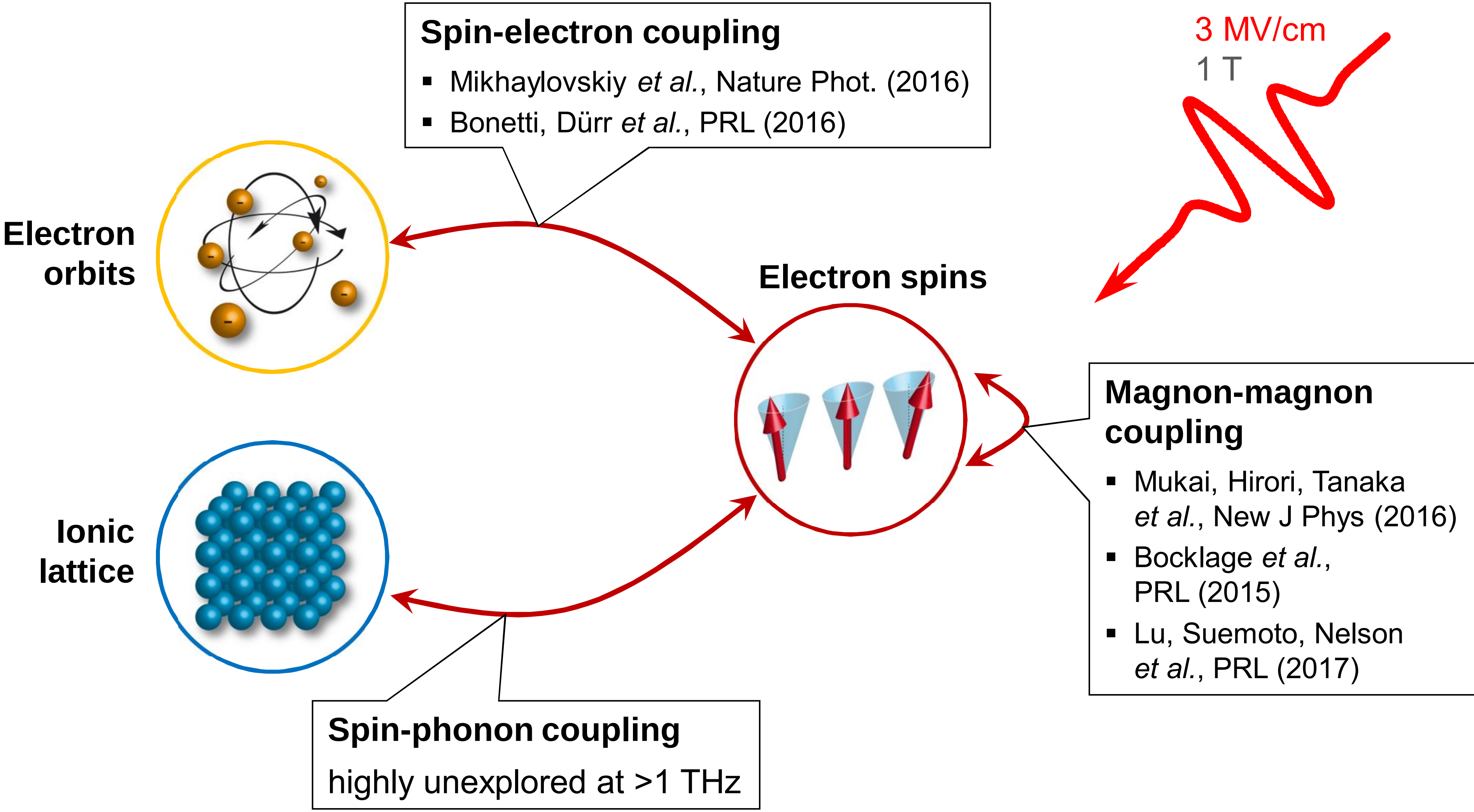
Nishitani, Hangyo *et al.*, APL (2010), PRB (2012)
Kanda, Kuwata-Gonokami *et al.*, Nature Comm. (2012)

- Dynamics of $B_{\text{exch}}B_{\text{ani}}$ following optical excitation

Bowlan, Prasankumar *et al.*, PRB (2016)
Mikhaylovskiy, Kimel *et al.*, Nature Comm. (2015)

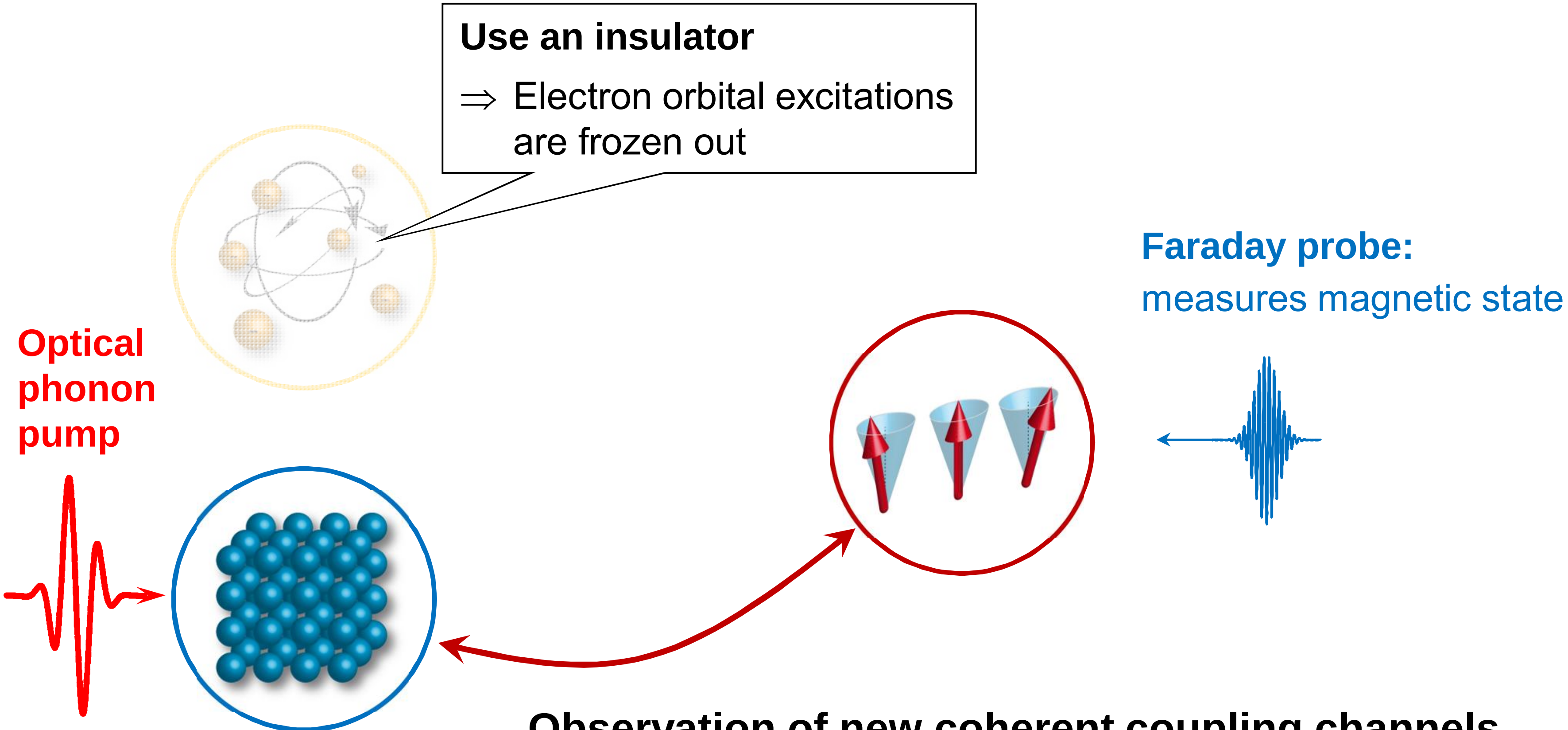
- **Not easy with non-optical methods**
- **Many more opportunities with stronger THz fields: probe spin couplings**

Reveal elementary spin couplings



How to probe coupling of spins and phonons?

Probing spin-phonon coupling



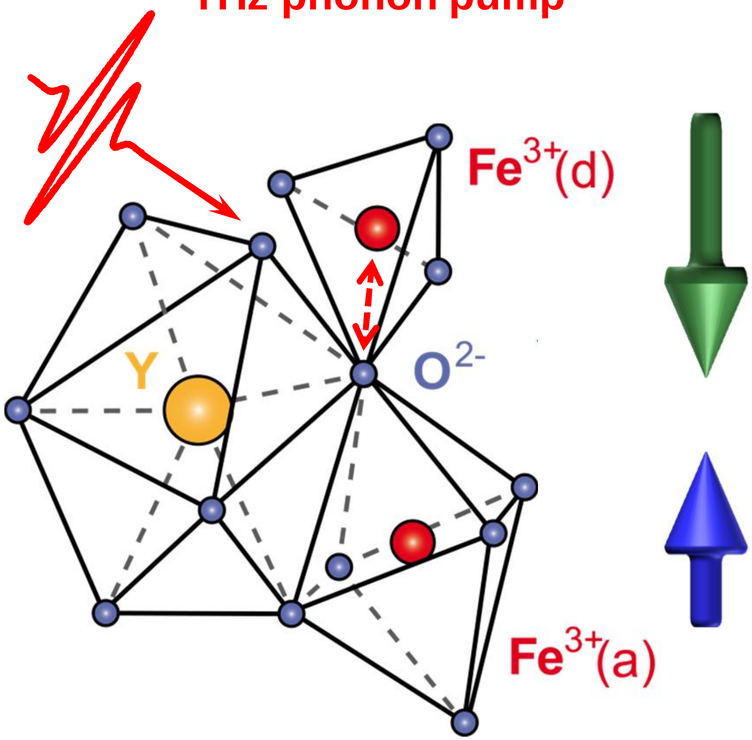
Observation of new coherent coupling channels

- Kubacka, Johnson, Staub *et al.*, Science (2014)
- Nova, Cavalleri *et al.*, Nature Phys. (2016)

How fast is spin-lattice equilibration?

⇒ Study model magnet YIG

Spin-lattice equilibration in YIG



Sample: ferrimagnet YIG

- Has two spin sublattices (a and d)
- Band gap of 2.8 eV
- Magnonic model material

Many open questions, e.g.:

Time scale and mechanism of spin-phonon equilibration unknown

~1 ps	~250 ps	~1 μs
Rezende *et al.*, JMMM (2016)	Schreier *et al.*, PRB (2013)	Xiao *et al.*, PRB (2010)

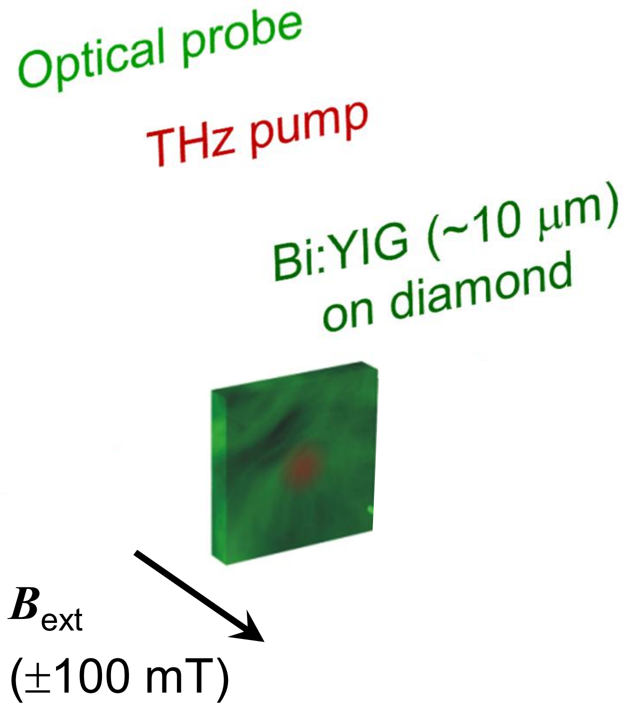
Relevant for

- Magnetization switching
- Spin Seebeck effect

Experiment

- Excite Fe-O lattice vibrations
- Probe spin dynamics from femtoseconds to microseconds

THz lattice pump–magnetooptical probe



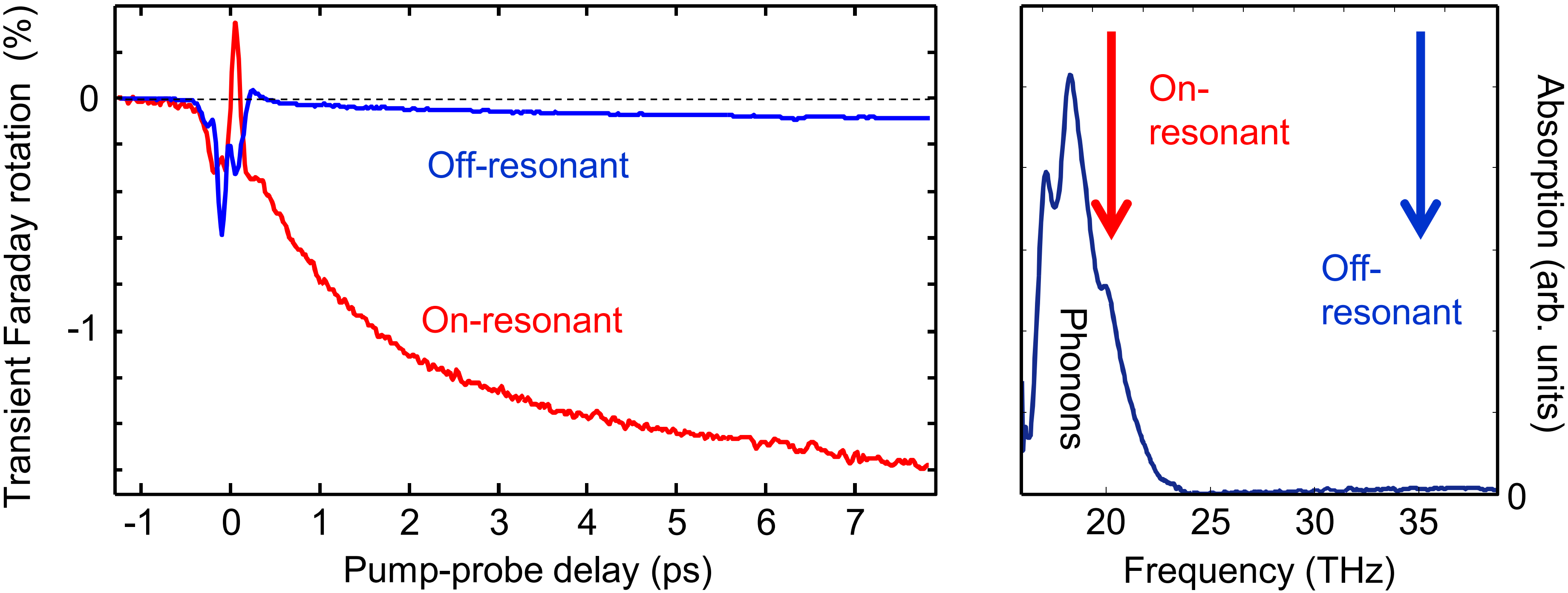
Detect Faraday rotation

$$\theta = A_{\mathrm{a}} M_{\mathrm{a}} + A_{\mathrm{d}} M_{\mathrm{d}}$$

Krumme *et al.*,
Thin Solid Films (1984)

Pump on and off the phonon resonances

Phonon-driven magnetization dynamics

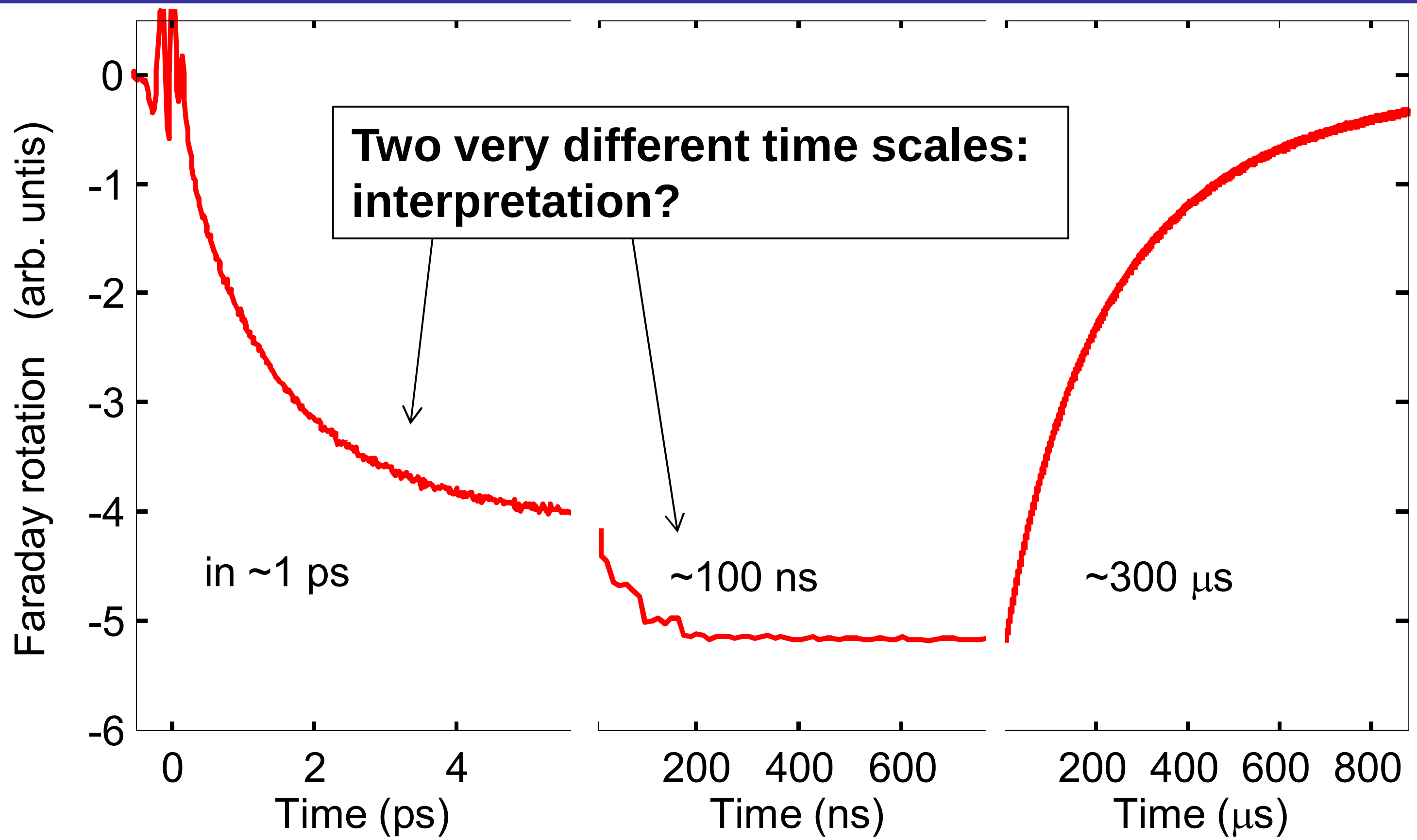


Surprisingly fast loss of magnetic order within ~1 ps:

- ~10^5 faster than lifetime of YIG's zone-center magnons (FMR)
- Response speed is comparable to laser-excited metals

Behavior on longer time scales?

From femtoseconds to milliseconds



Ultrafast magnetic-order quenching

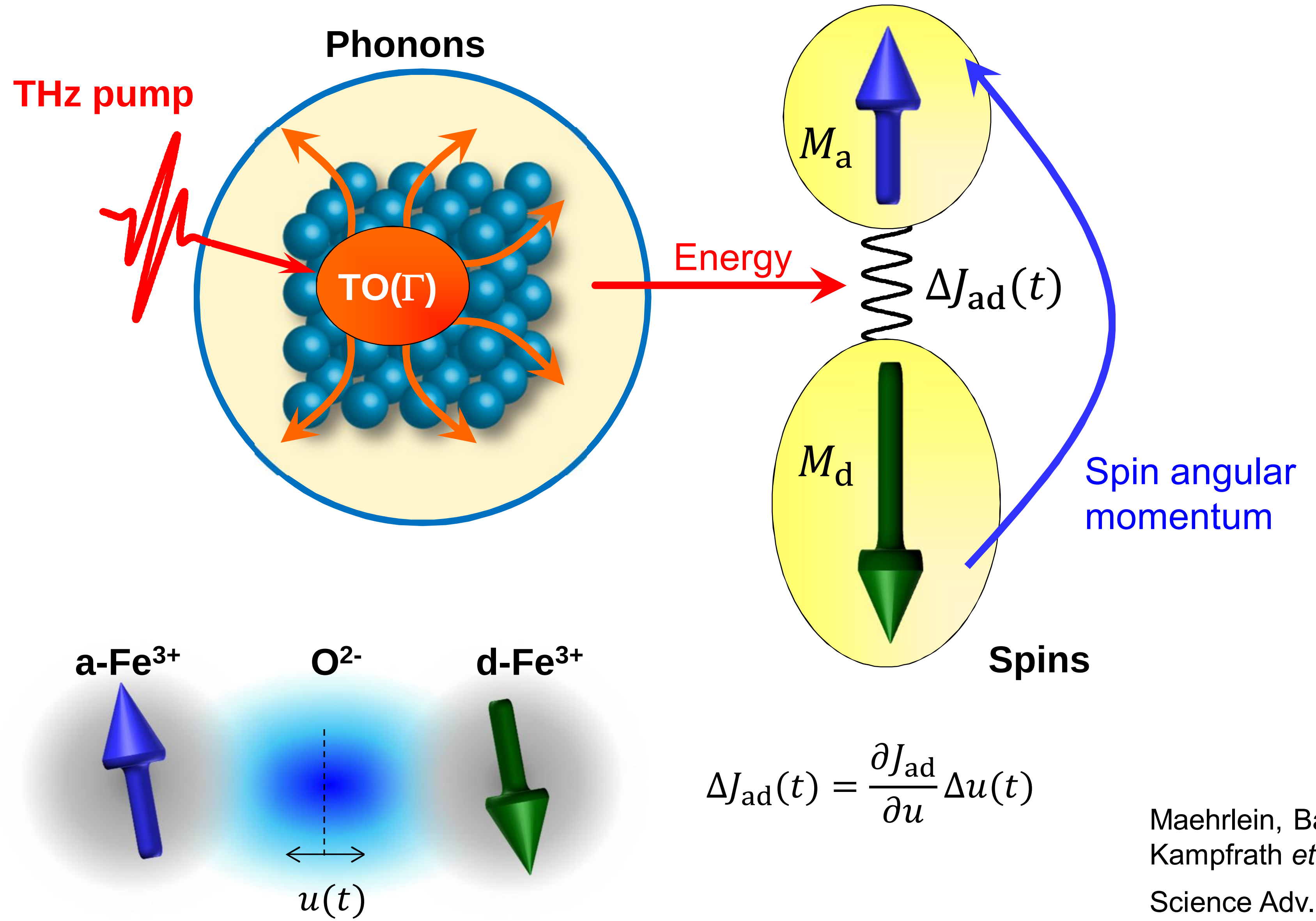
Full equilibration:

deduced from temp.-dependence

Heat flow to substrate:

From simulations, different substrates

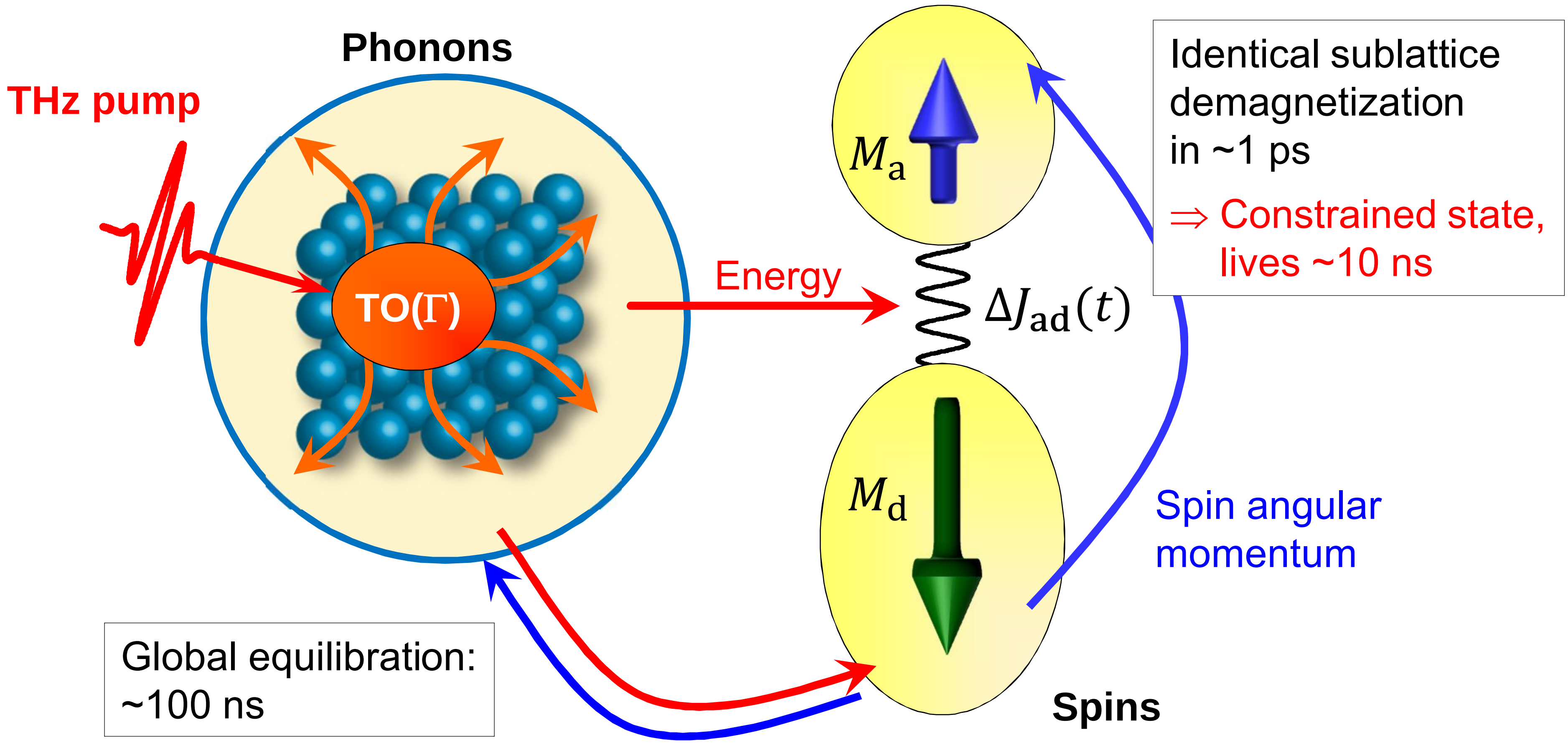
Summary: spin-phonon equilibration in YIG



$$\Delta J_{ad}(t) = \frac{\partial J_{ad}}{\partial u} \Delta u(t)$$

Maehrlein, Barker, Kampfrath *et al.*,
Science Adv. (2018)

Summary: spin-phonon equilibration in YIG



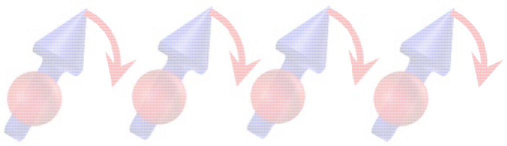
Reveals spin-phonon equilibration in YIG:

- Transfer of energy: in ~1 ps
- …and angular momentum: ~100 ns

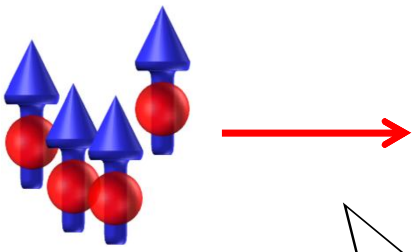
Maehrlein, Barker, Kampfrath *et al.*, Science Adv. (2018)

Three elementary spin operations

1. Turn spins around



2. Transport spins

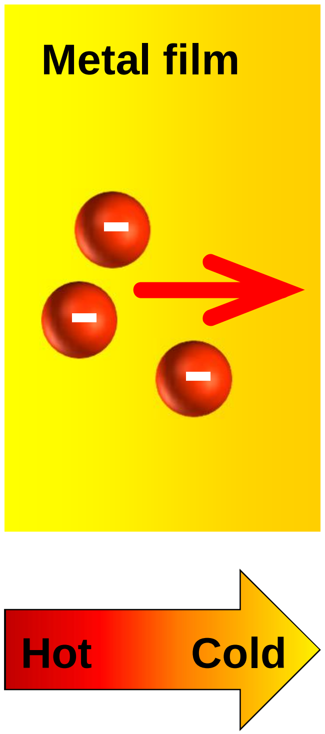


3. Detect spin dynamics

How to launch and detect spin currents ultrafast?

Idea: make use of spin-caloric effects

Heat-driven: the Seebeck effect



Temperature gradient

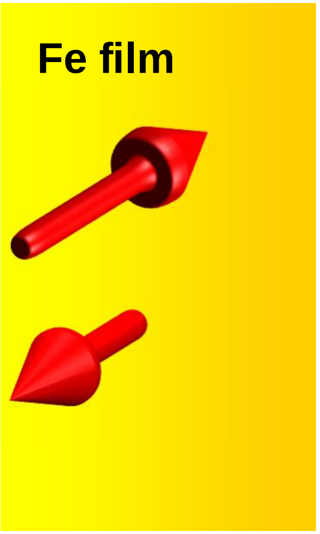
Thomas Seebeck (1821):

A temperature gradient drives an electron current

Ken-ichi Uchida (2008):

In ferromagnets, the Seebeck current is spin-dependent

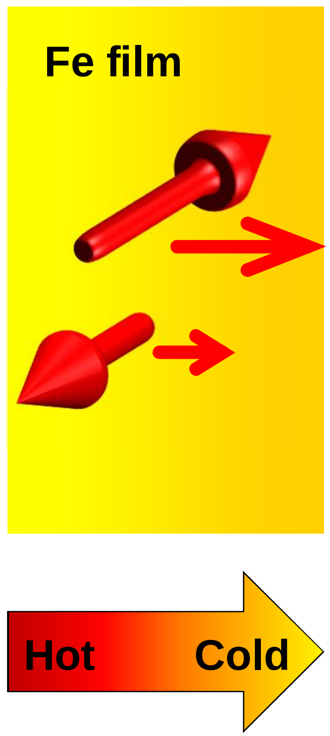
Spin-dependent Seebeck effect (SDSE)



↑ 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

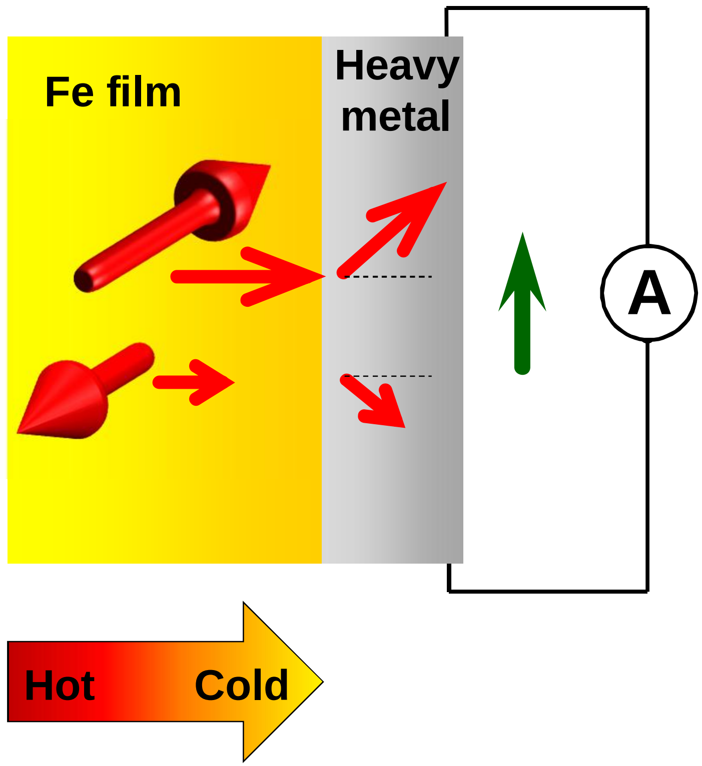
↑ and ↓ electrons have very different transport properties

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

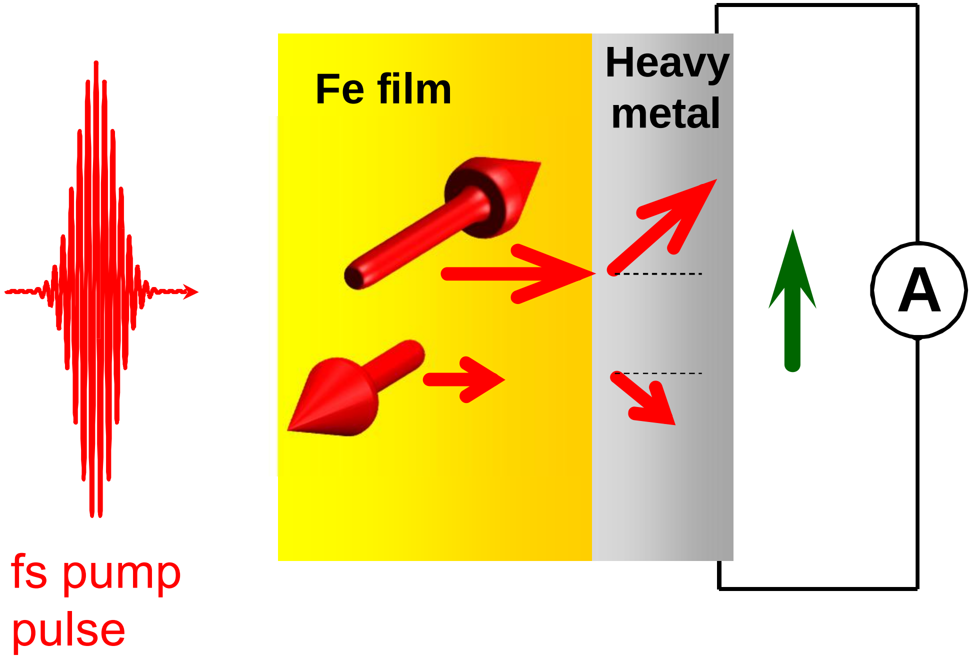
⇒ Transverse charge current

⇒ Spin-to-charge (S2C conversion

Saitoh *et al.*, APL (2006))

How can we induce an imbalance as fast as possible?

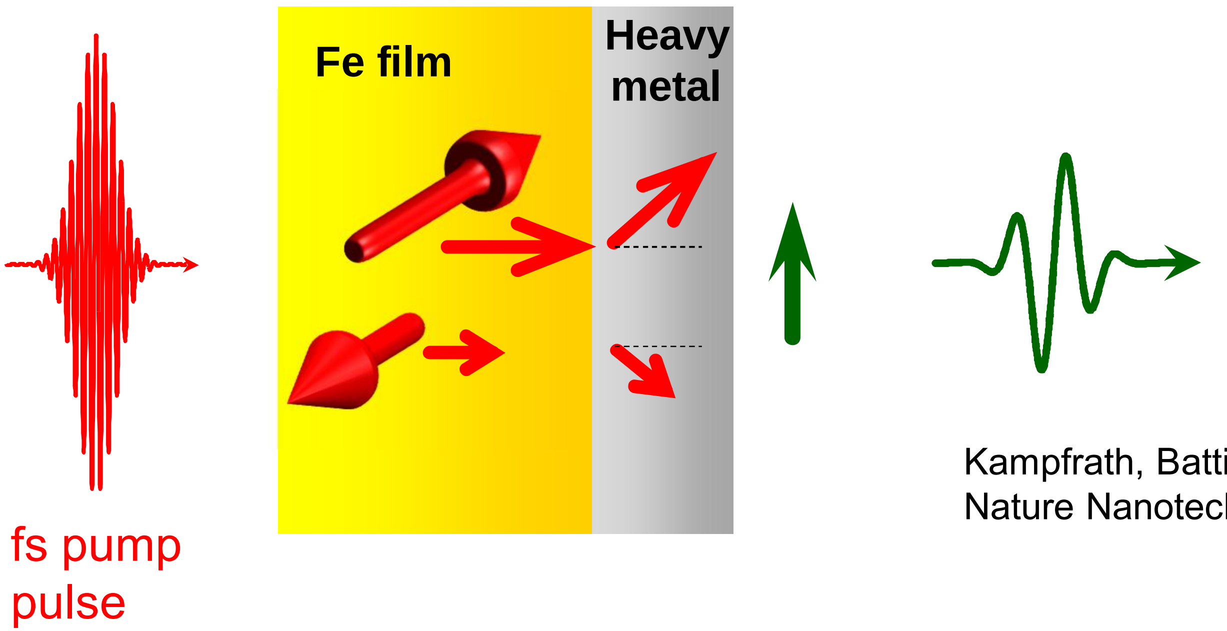
Inverse spin Hall effect (ISHE)



Technical challenge:

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

Inverse spin Hall effect (ISHE)



Emission of electromagnetic pulse (~1 THz)

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

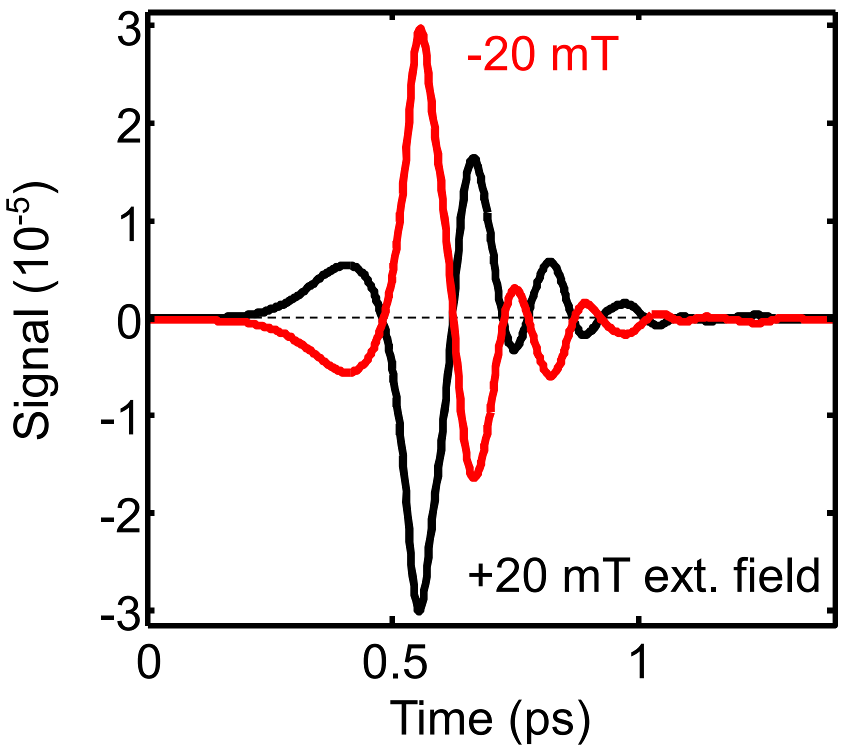
⇒ Measure THz emission from photoexcited FM|NM bilayers

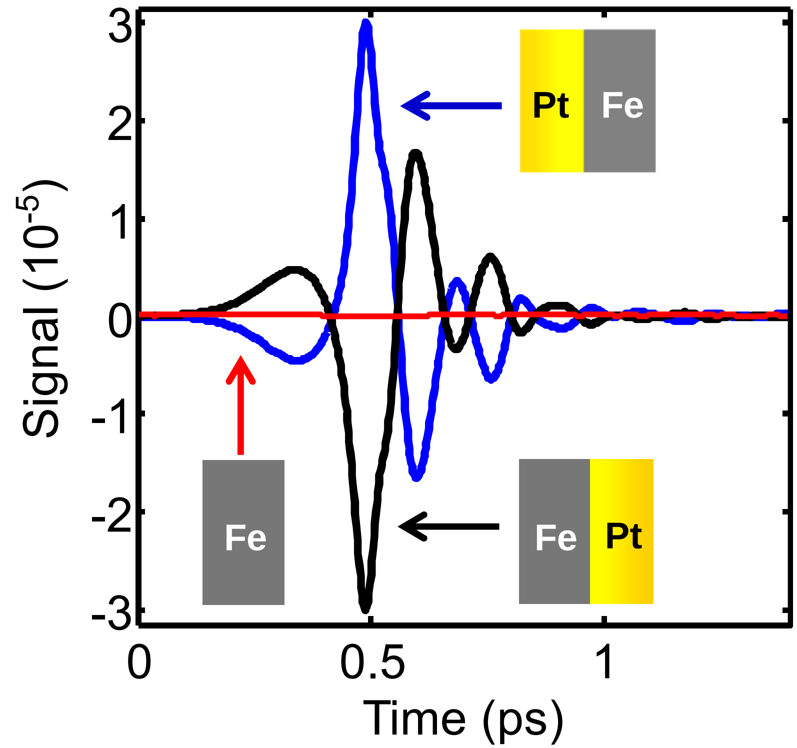
Samples: polycrystalline films (labs of M. Kläui and M. Münzenberg)

Pump pulses: from Ti:sapphire oscillator (10 fs, 800 nm, 2.5 nJ)

A look in the lab…

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

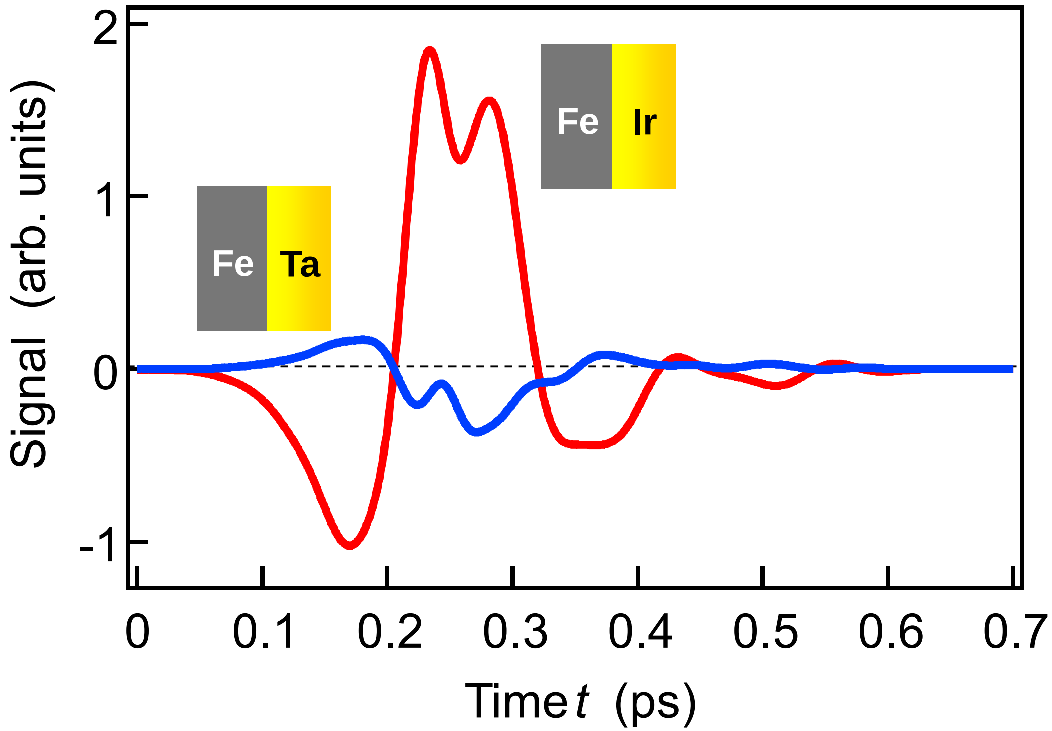
Ultrafast inverse spin Hall effect

Idea:

vary nonmagnetic cap layer

Ta vs Ir:

opposite spin Hall angles, Ir larger



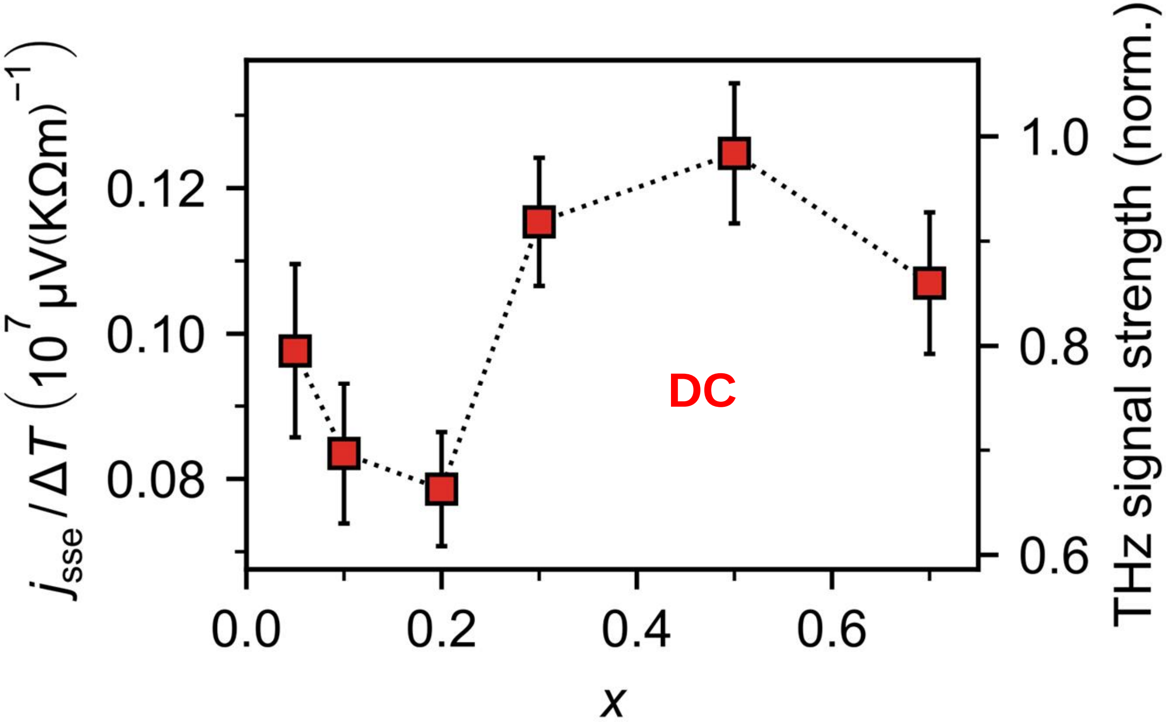
The inverse spin Hall effect is still operative at THz frequencies

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

Interesting applications:

1) Rapid material characterization regarding spin-to-charge conversion (S2C)
2) Generation of THz pulses

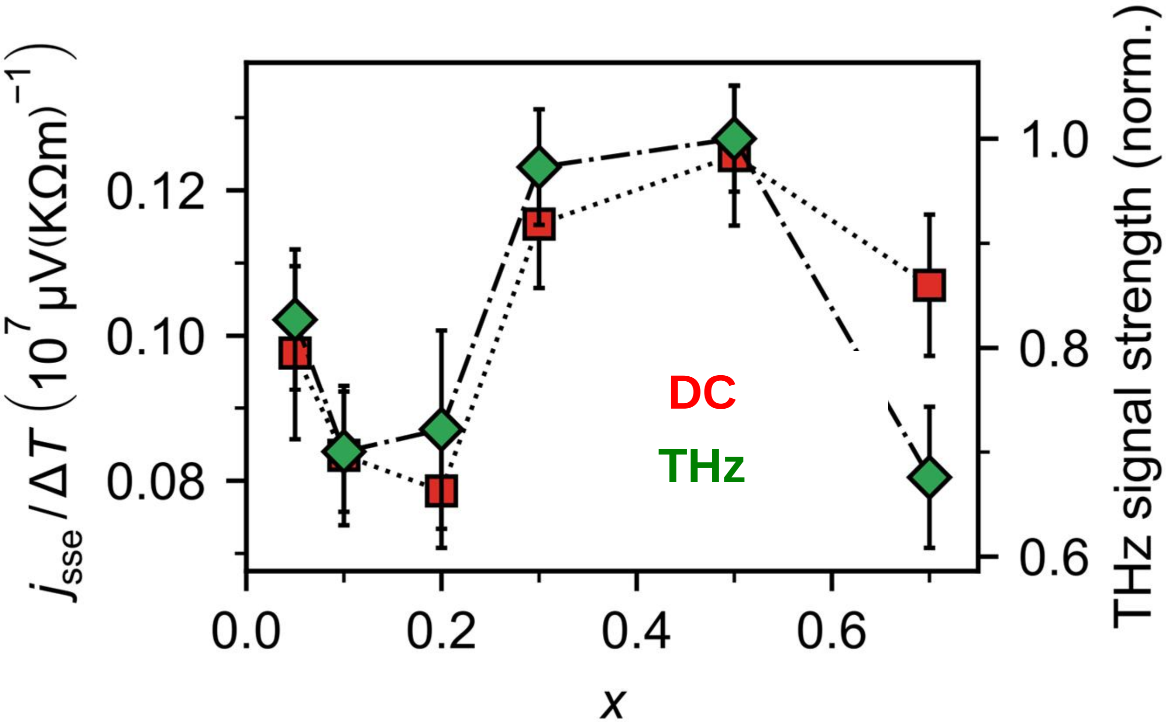
Application 1: characterize S2C strength



N = $Cu_{1-x}Ir_x$

Cramer, Seifert, Kampfrath, Kläui *et al.*, Nano Lett. (2018)

Application 1: characterize S2C strength



N = $Cu_{1-x}Ir_x$

THz emission strength correlates with DC S2C strength

Cramer, Seifert, Kampfrath, Kläui *et al.*, Nano Lett. (2018)

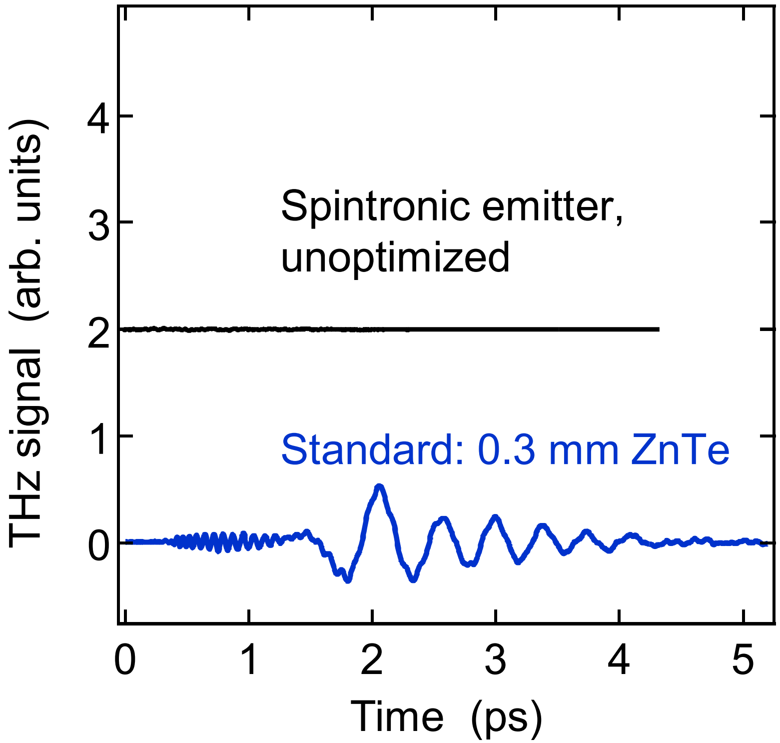
THz emission spectroscopy enables rapid material screening: estimate of the relative spin-Hall conductivity

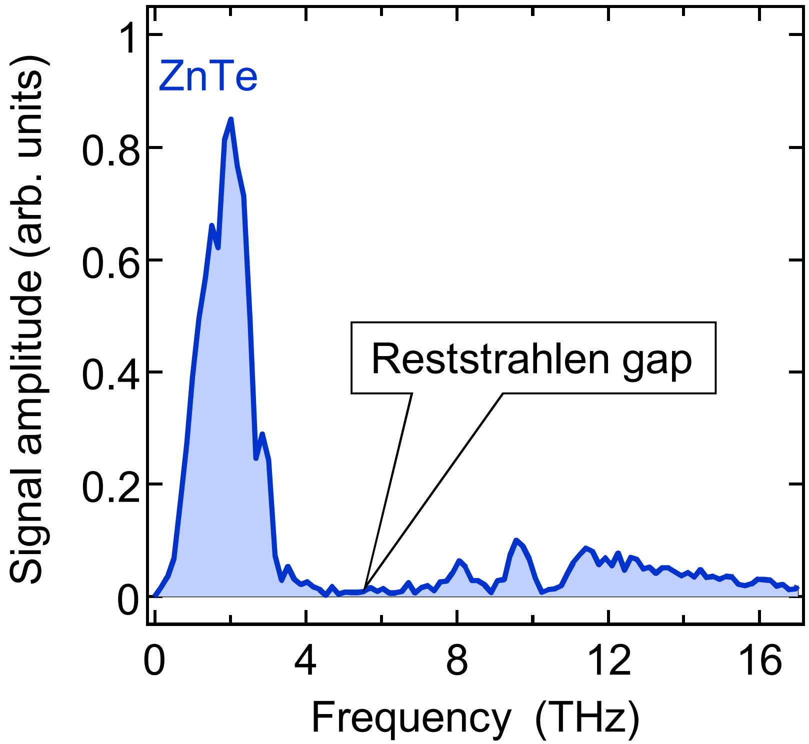
Sasaki, Suzuki, Mizukami, APL (2017)

Seifert *et al.*, SPIN (2017), J. Phys. D (2018)

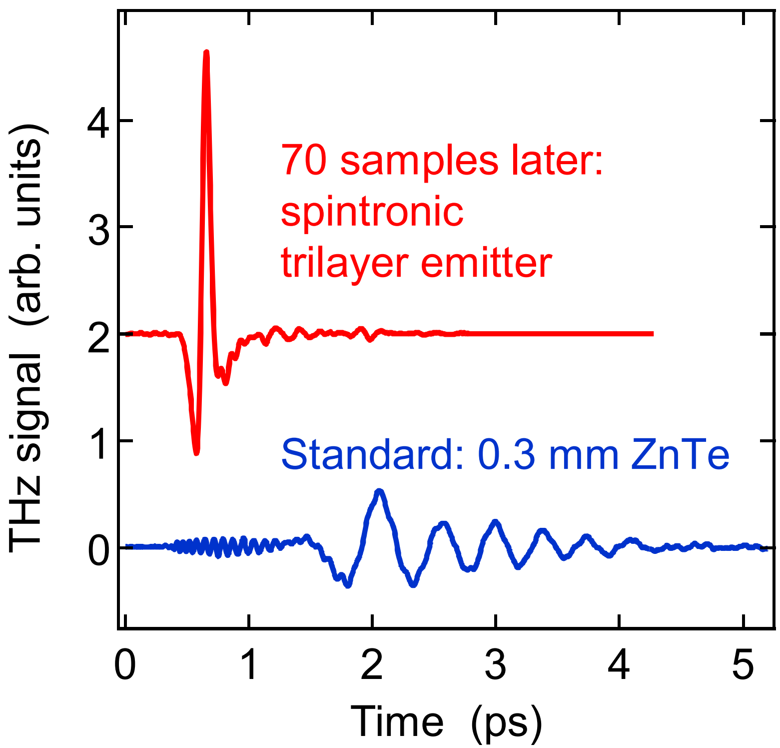
Idea: optimize materials and geometry to maximize the THz amplitude

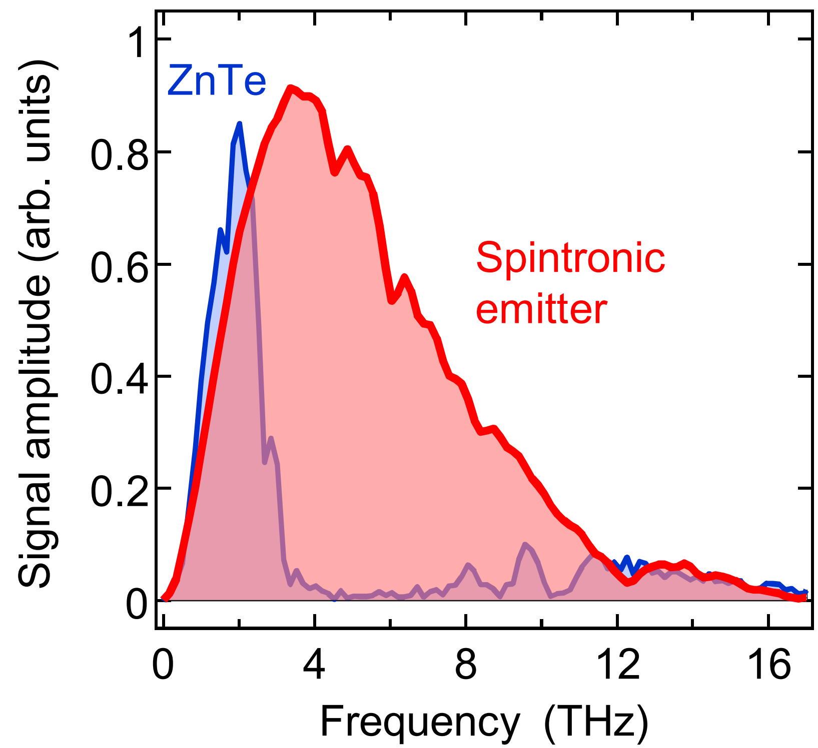
Application 2: spintronic THz emitter





Application 2: spintronic THz emitter





More broadband, efficient and cheaper than standard emitters like ZnTe

Seifert *et al.*, Nature Photon. (2016)

More features…

More features and developments

Upscaling yields 0.3 MV/cm field

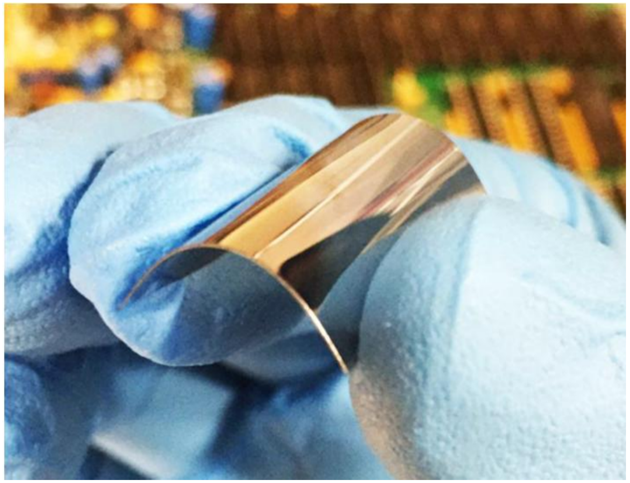
Seifert, Kläui, Kampfrath *et al.*, APL (2017)

Insensitive to pump wavelength

Papaioannou, Beigang *et al.*, arXiV (2018)
Herapath, Hendry *et al.* arXiv (2018)

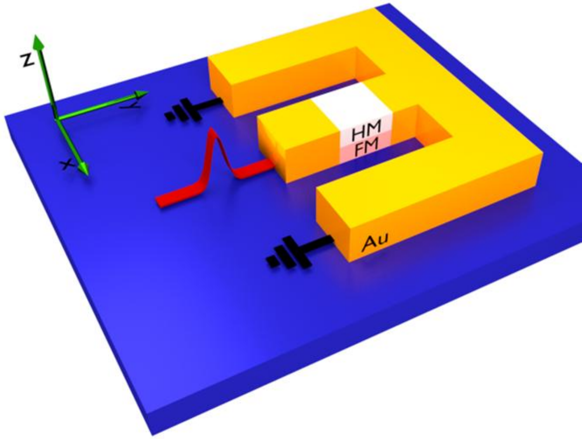
Flexible substrates

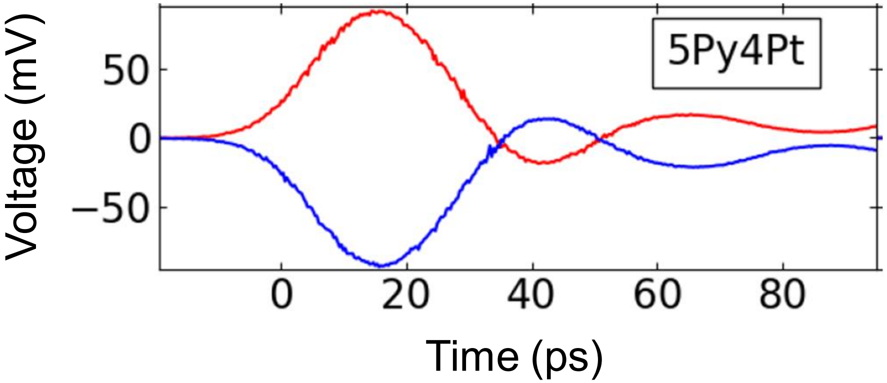
Wu, Yang *et al.*, Adv. Mat. (2016)



On-chip THz source

Weber, Kampfrath, Woltersdorf *et al.* (2018)

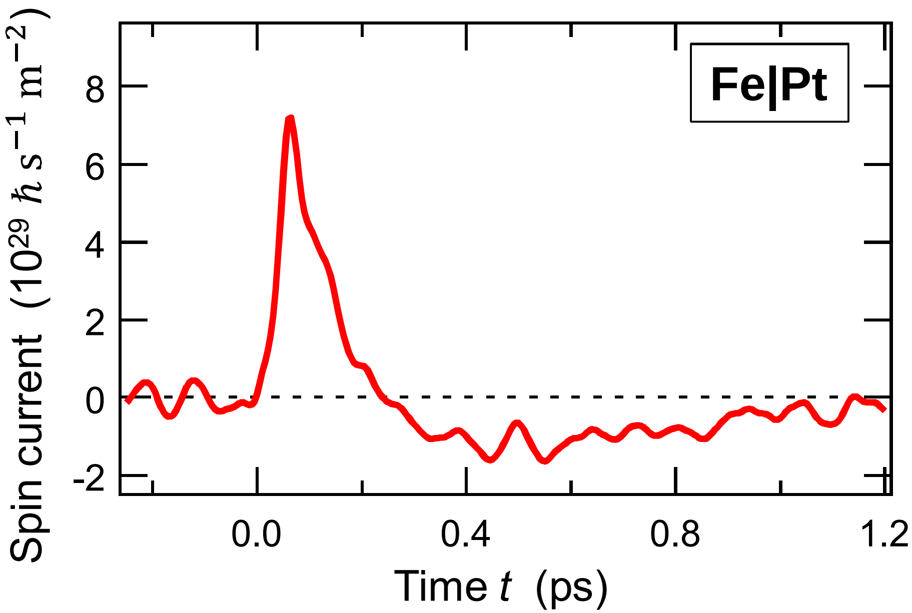


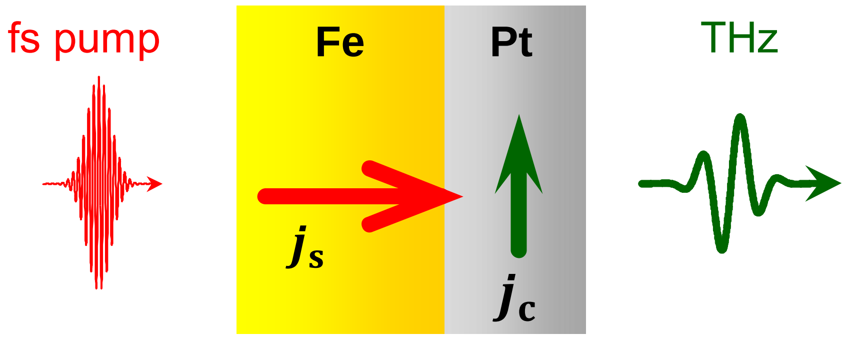


Need better understanding for better performance

What does the driving THz spin current look like?

Dynamics of the spin current





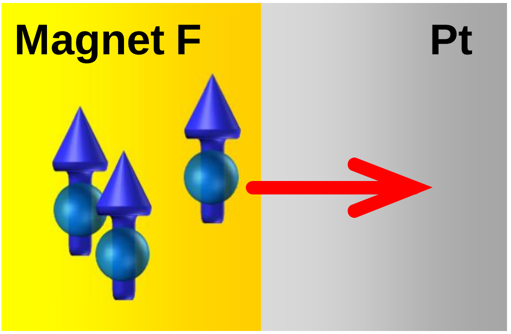
Extremely fast bipolar response

Analysis not straightforward:

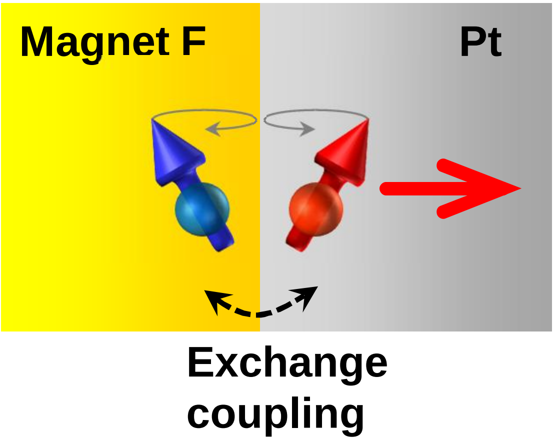
two competing mechanisms of spin transport

Two types of spin transport

By moving electrons



By torque between adjacent spins



Only possible in magnetic metals:

"Spin-dependent Seebeck effect" (SDSE)

Even possible for magnetic insulators:

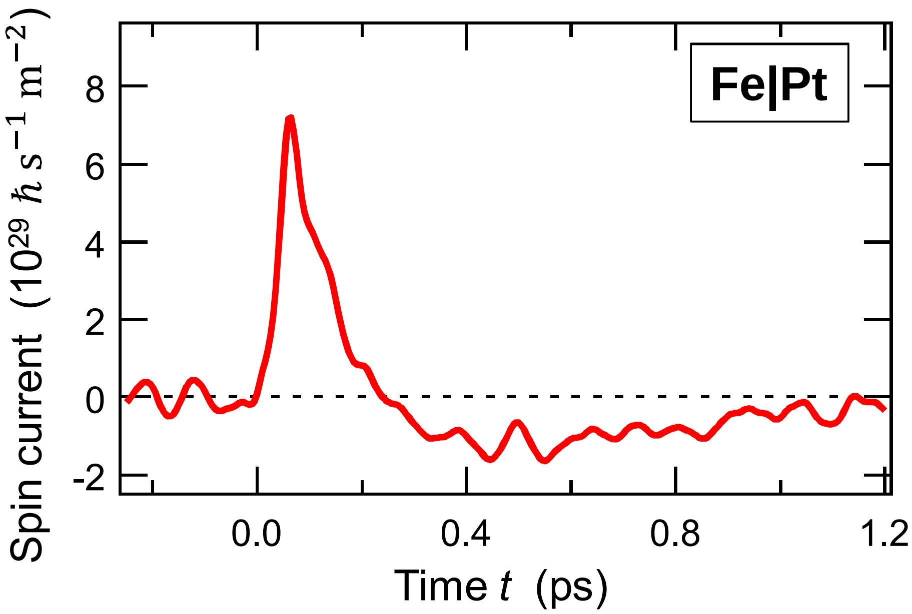
"Magnonic spin Seebeck effect" (SSE), "thermal spin pumping"

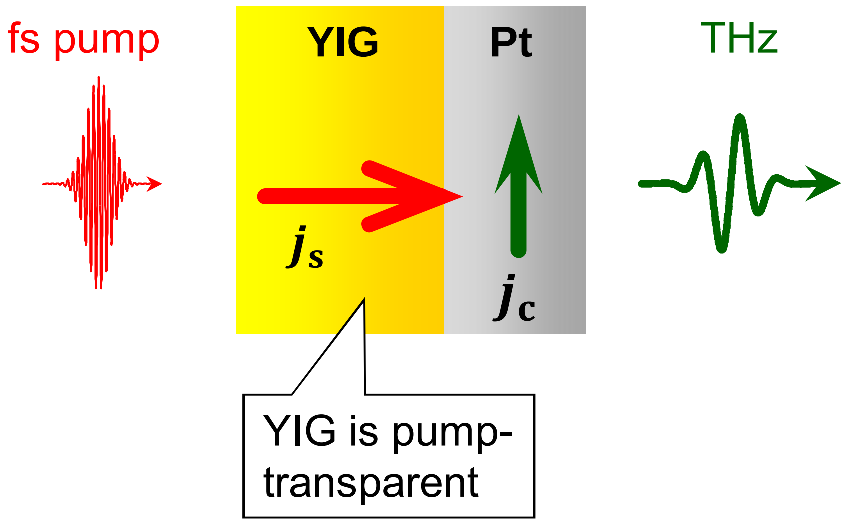
Thus: measure magnetic metals vs insulators

- Reveal relative weight of the two spin-current contributions
- Insulators are potentially simpler to model

Magnet: metal vs insulator

Spin-dependent Seebeck effect



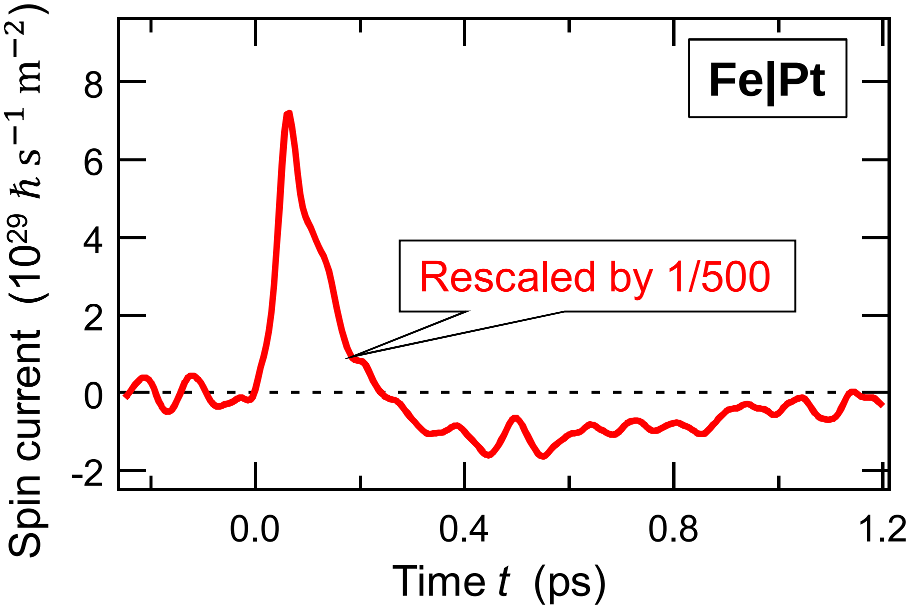


Substitute Fe by insulating YIG

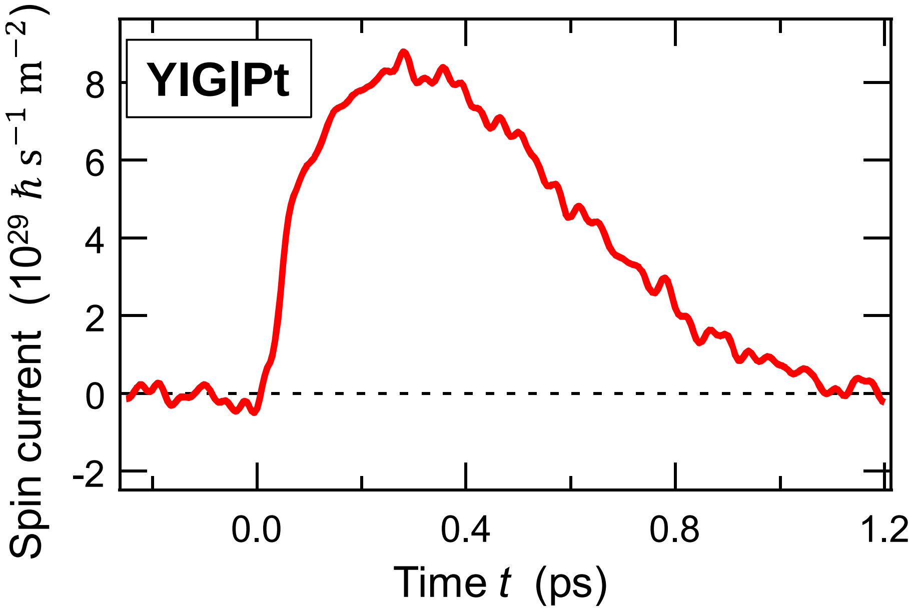
- Switch electron transport off
- Only spin torque possible

Magnet: metal vs insulator

Spin-dependent Seebeck effect



Magnonic spin Seebeck effect



Fe→Pt spin current ~ $10^3\times$ YIG→Pt spin current

⇒ The Fe→Pt spin current has a negligible torque contribution

What determines the dynamics of the magnon current YIG→Pt ?

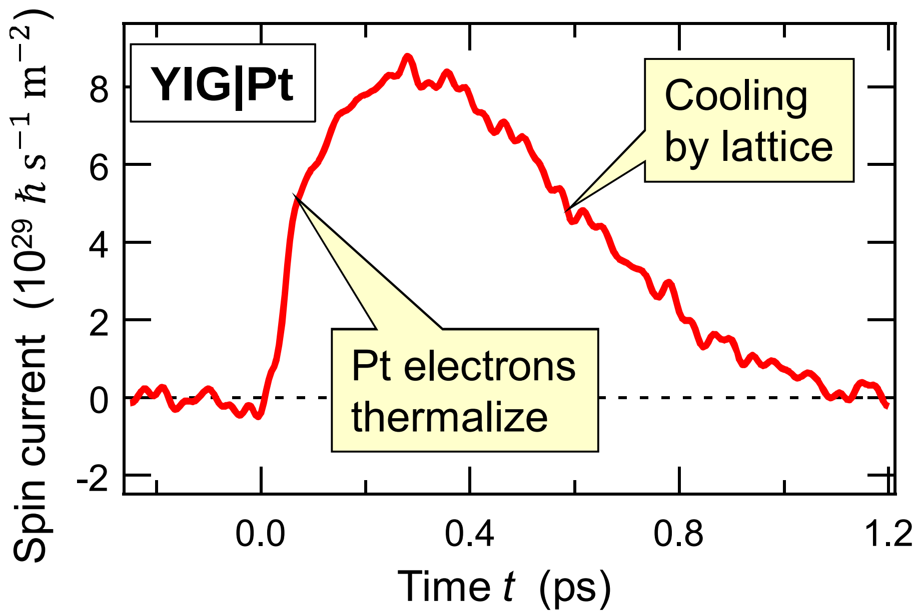
Dynamics of the spin Seebeck current

Lei *et al.*,
PRB **66**, 245420 (2002)

Caffrey *et al.*,
Microsc Thermoph Eng **9**, 365 (2005)

The spin Seebeck current follows the electron temperature in Pt quasi-instantaneously

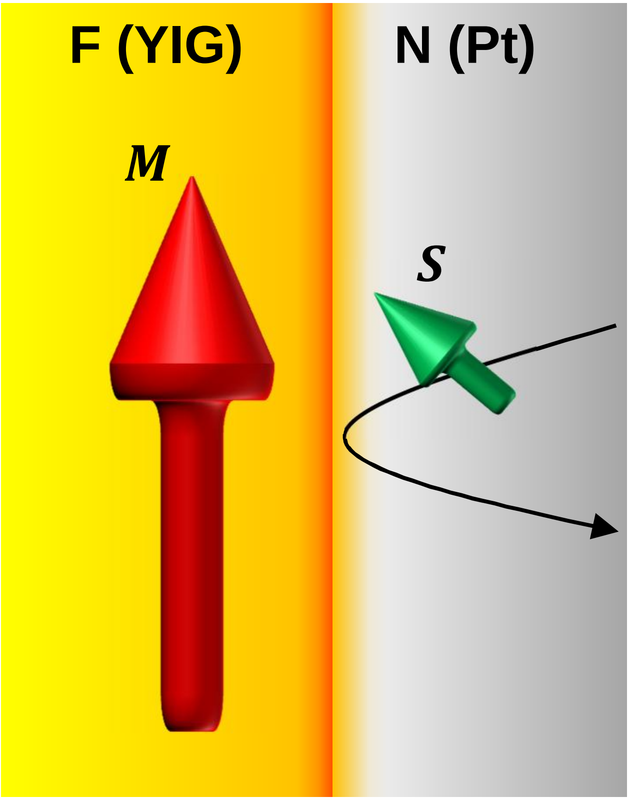
Seifert, Barker, Wolf, Kläui, Kampfrath *et al.*,
Nature Commun. (2018)



- **Why is the spin-current formation so fast?**
- **Why does it rise with electron thermalization?**

Analytical modeling and simulations support the following picture…

The first steps of the spin Seebeck effect



- Pt spin $\boldsymbol{S}$ is incident on interface: $\langle \boldsymbol{S} \rangle = 0$
- Reflected spin $\boldsymbol{S}$ is aligned more parallel to $\boldsymbol{M}$: $\langle \boldsymbol{S} \rangle \upuparrows \boldsymbol{M}$ (similar to STT)

Seifert, Barker, Wolf, Kläui, Kampfrath *et al.*, Nature Commun. (2018)

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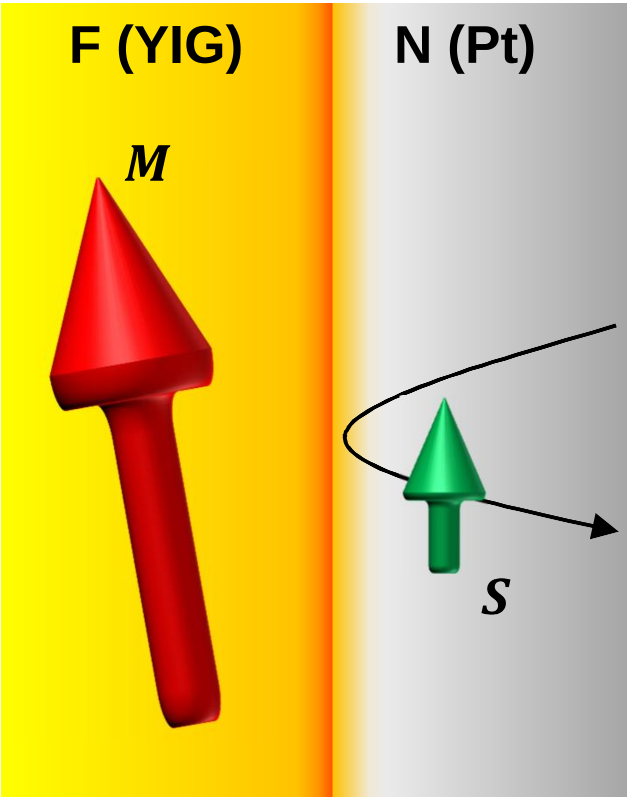

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The spin current j_s is

$\propto$ rate of reflection events

$\propto$ number of electron-hole pairs in Pt

$\Rightarrow j_s$ rises when the photoexcited carriers multiply

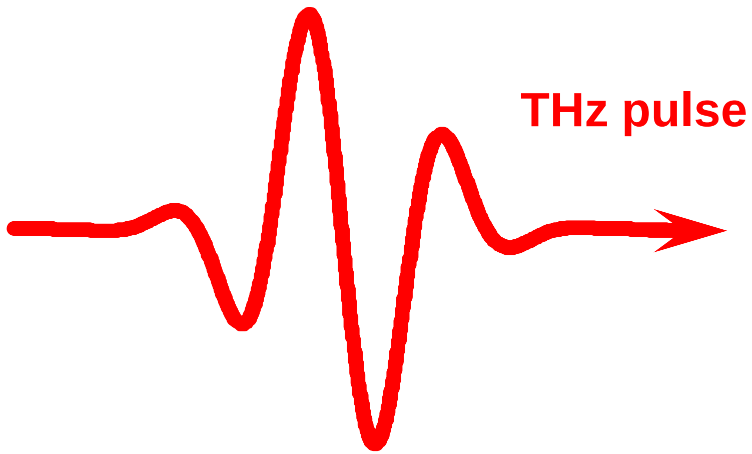
The response is quasi-instantaneous since

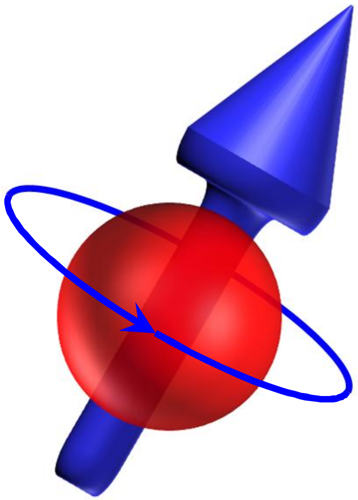
- Pt spins traverse the interface region in <5 fs
- YIG spins react without inertia

Seifert, Barker, Wolf, Kläui, Kampfrath *et al.*, Nature Commun. (2018)

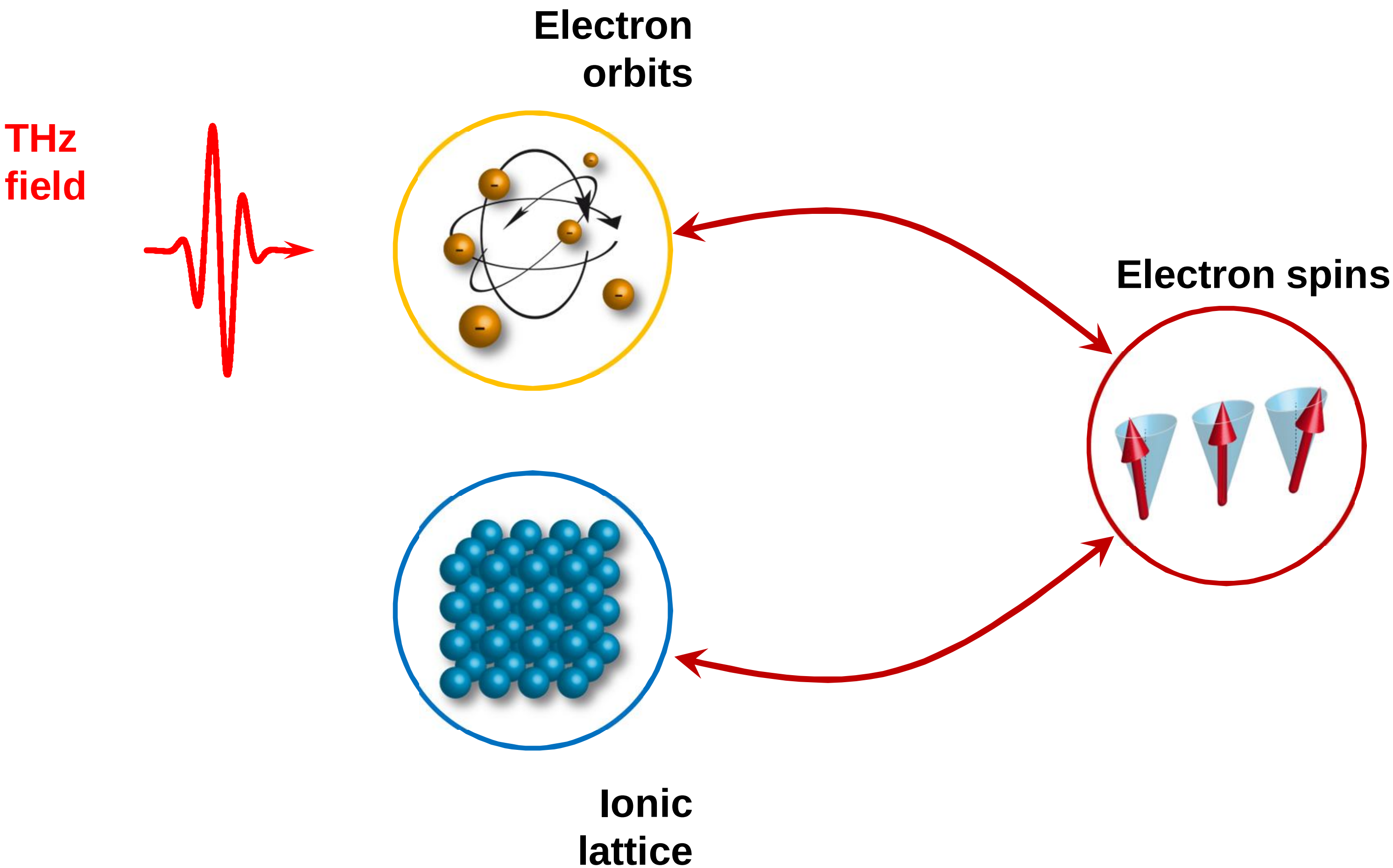
Need thermalized electrons for large spin Seebeck effect

Outlook: toward THz current control





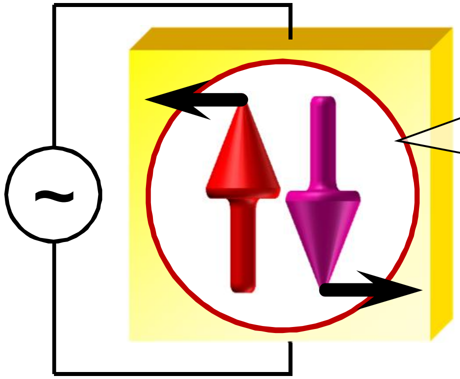
Outlook: THz-field-driven currents



Control over spins with THz-driven currents?

Outlook: switching of antiferromagnets

Electric writing with Ohmic contacts



CuMnAs

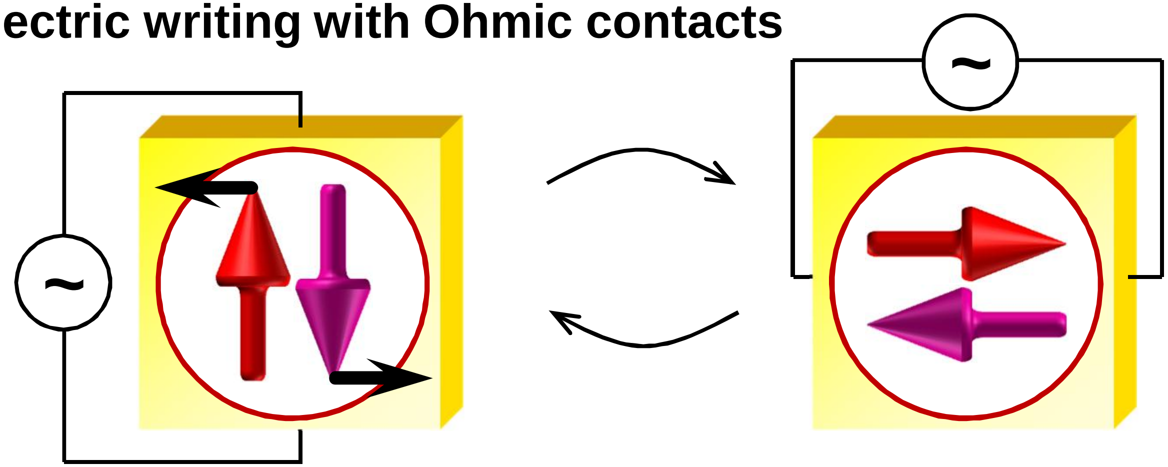
- Antiferromagnetic metal
- Locally broken inversion symmetry

⇒ Current induces staggered magnetic field

Wadley, Jungwirth *et al.*, Science (2016)

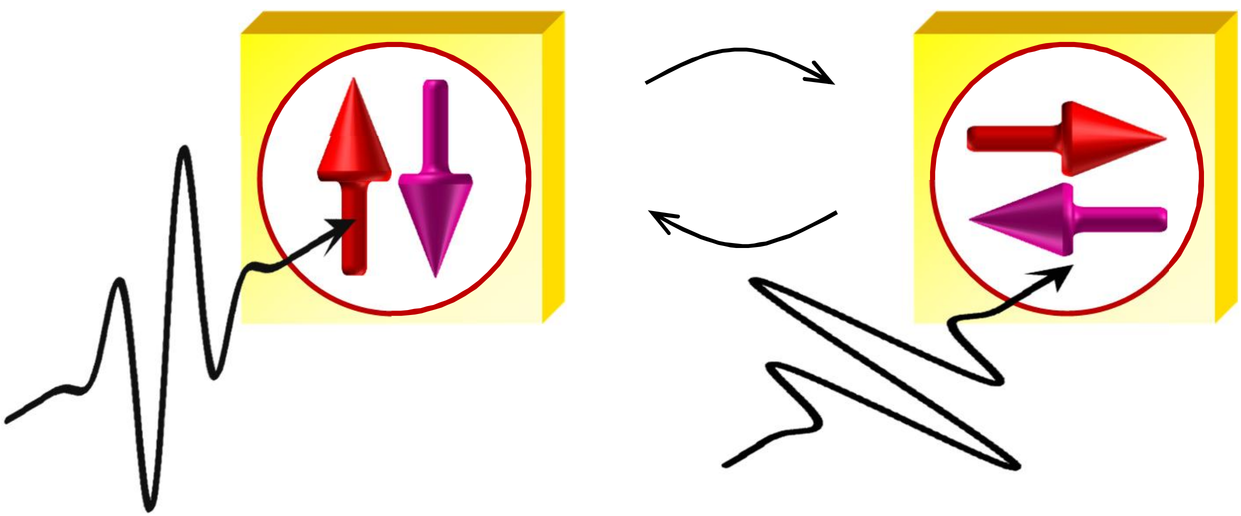
Outlook: switching of antiferromagnets

Electric writing with Ohmic contacts



Wadley, Jungwirth *et al.*, Science (2016)

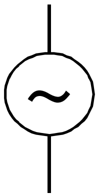
Idea: drive a THz current, contact-free



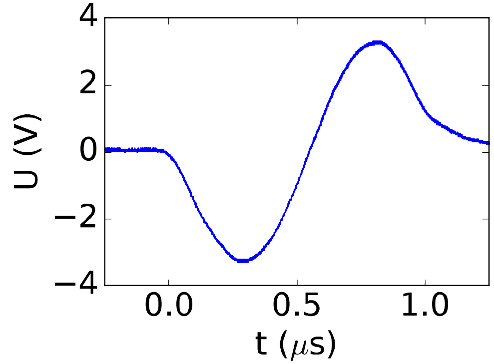
Olejnik, Seifert, Kuzel, Sinova, Kampfrath, Jungwirth *et al.*, Science Advances (2018)

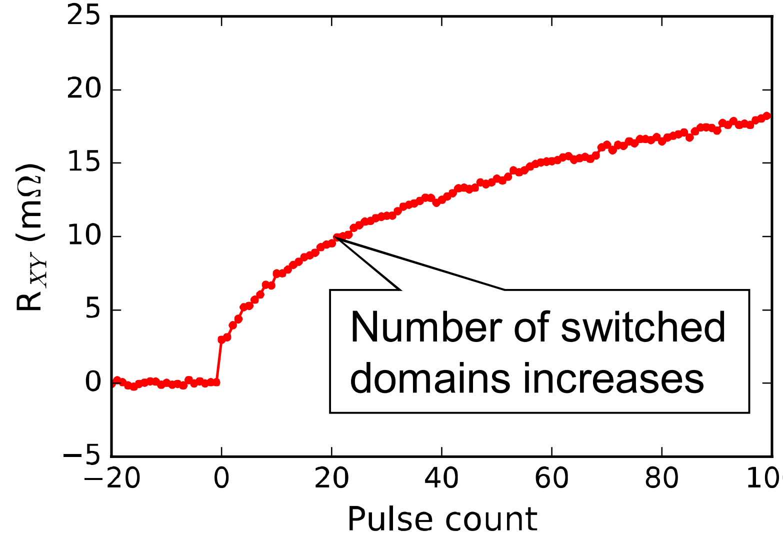
Compare DC vs THz for same sample: probe L with AMR

Writing with MHz and THz fields

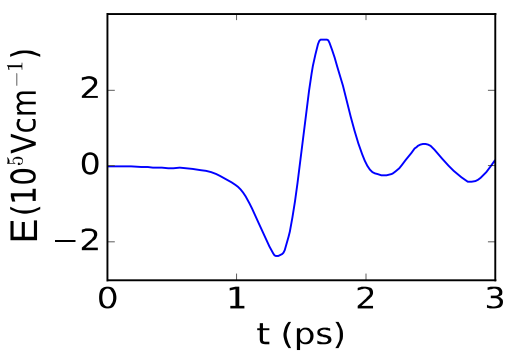


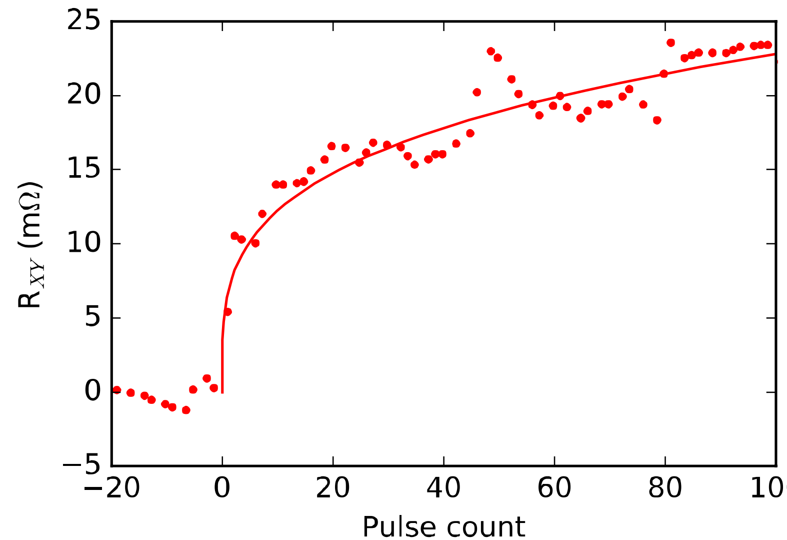
MHz voltage pulse





Free-space THz pulse

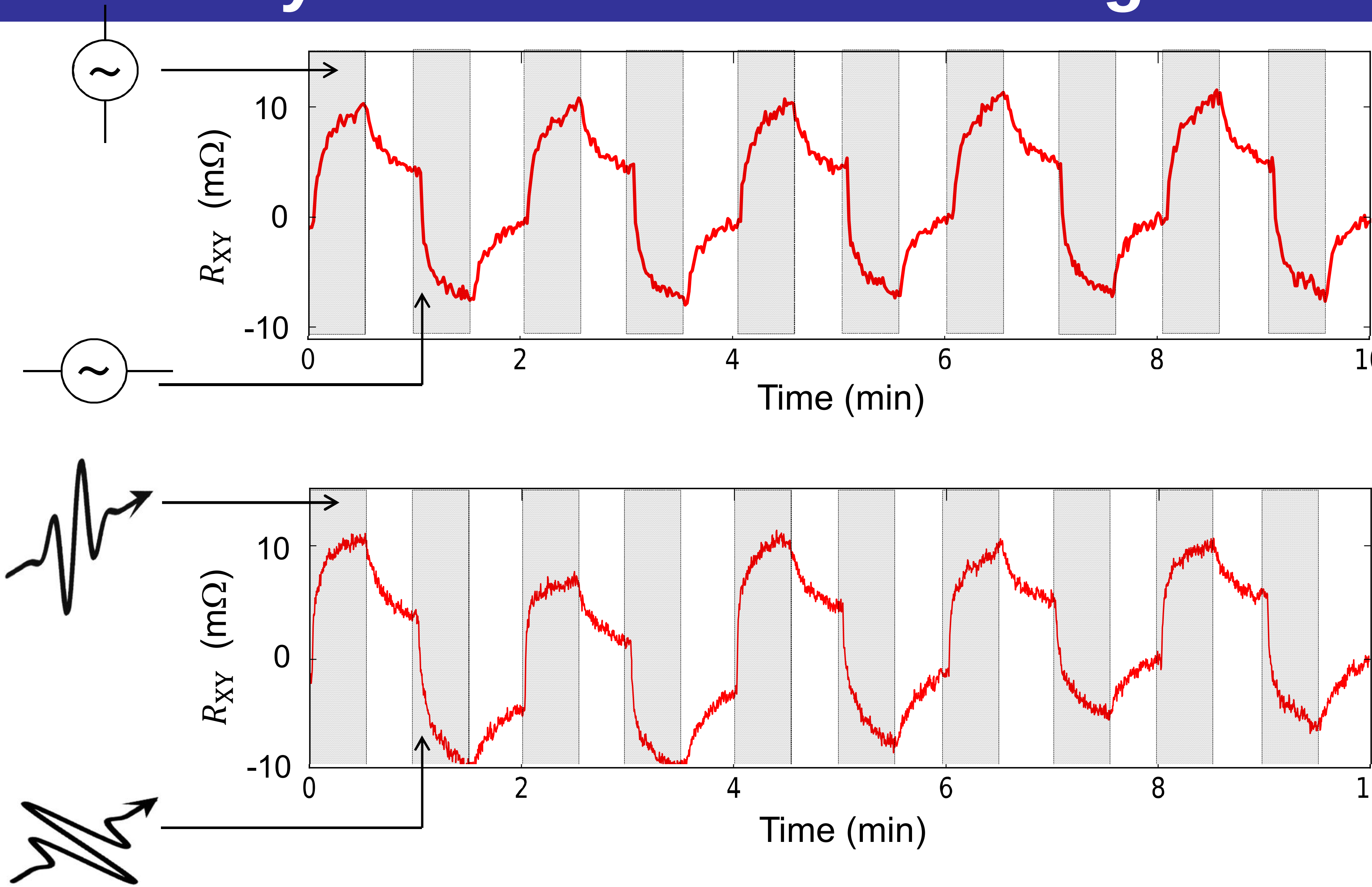




Cyclic operation also possible

Olejnik *et al.*,
Science Advances (2018)

Cyclic MHz and THz writing

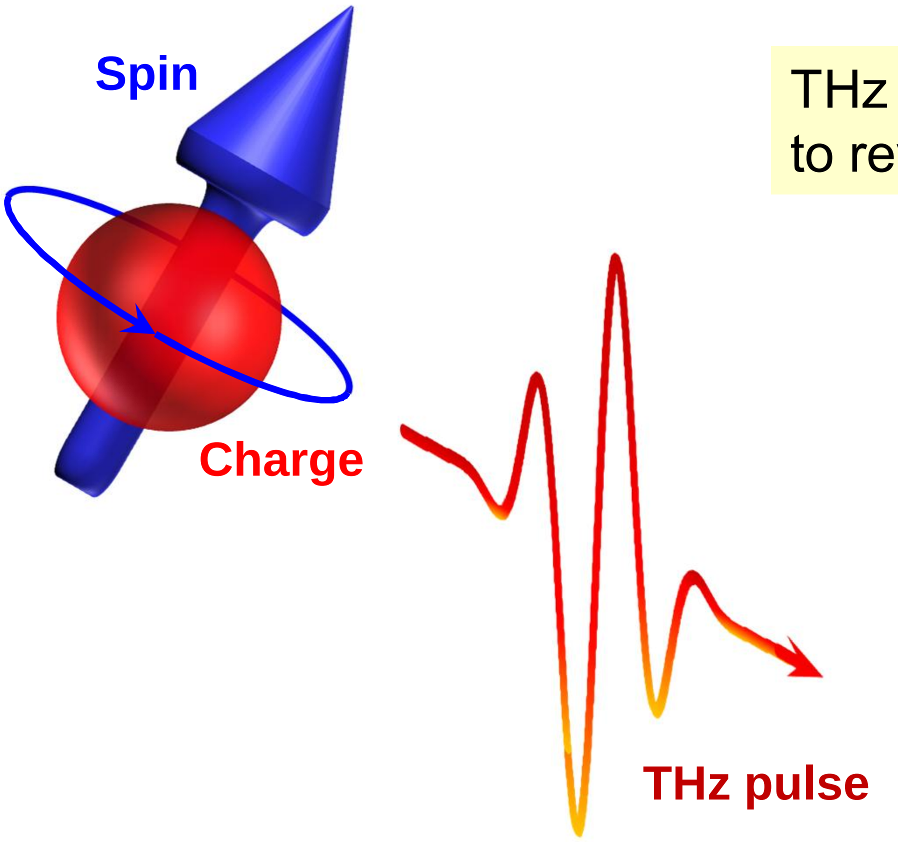


Olejnik *et al.*,
Science Advances (2018)

What about driving phonons?

Summary

- THz fields can access elementary spin couplings (e.g. to phonons)
- Spin Hall and spin Seebeck effects are operative up to 10s of THz
- Studying ultrafast regime permits new insights into spin physics and new applications in THz photonics



THz radiation is a useful tool
to reveal and control spin dynamics