Practical: how to measure ultrafast spin and charge currents



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Heat-driven currents: the Seebeck effect



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)



 \uparrow and \downarrow electrons have very different transport properties

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

Spin-dependent Seebeck effect (SDSE)



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 \Rightarrow Spin-polarized current

Detection with the inverse spin Hall effect

Inverse spin Hall effect (ISHE)



Spin-orbit coupling deflects electrons

- \Rightarrow 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)

 \Rightarrow 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)

How can we detect the THz pulse?

THz detection: electro-optic sampling



Electro-optic effect:

Change in refractive index $\propto E_{THz}(t) \Rightarrow$ Crystal becomes birefringent

Scan ellipticity of sampling pulse vs $\tau \Rightarrow$ Get THz electric field $E_{THz}(\tau)$

A look in the lab...

Simple THz emission setup in the lab



Optical pump beam

Spintronic sample

Parabolic mirror

Electrooptic crystal for sampling of the THz electric field

To detection of probe ellipticity

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

Ta vs lr:

Idea:

vary nonmagnetic cap layer

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)

This has interesting applications... tomorrow

Today: how can we determine the THz-emitting source current?

THz source current



Issue: we do *not* measure E(t)

The transfer function of the THz setup



How can we get the transfer function h?

How to determine the transfer function?

$$S = h * E$$

Goal: determine *h* over large bandwidth (0.3 to 40 THz)

1) Calculate h: requires approximations, e.g. idealized setup

2) Measure h: use a broadband THz reference emitter

Measure
$$S_{ref} = h * E_{ref}$$
 Calculate

Use optically transparent THz emitter:

- $\chi^{(1)}$ and $\chi^{(2)}$ are well known $\Rightarrow E_{ref}$ is quite well predictable
- We choose ZnTe and GaP

Reference emitter: GaP(110), 50 µm thick



 $E_{\rm ref} * h = S_{\rm ref}$

Solve for *h*—directly in the time domain

Experiment vs theory

Spectral amplitude $|\tilde{h}(\omega)|$ Transfer function h(t)h(t) (arb. untis) $| ilde{h}|$ (arb. untis) -0.2 0.2 0.4 20 30 0 10 0 Time t (ps) Frequency $\omega/2\pi$ (THz)

Highly consistent results for *h*



Reference emitter: ZnTe(110), 50 µm thick

Calculated:

- Extended Gaussian beam propagation
- Detector response

Understanding the structure of h



- t = 0: remainder of input δ -peak
- t < 0: faster THz components
- t > 0: slower components, e.g. in Reststrahlen region

Spectral amplitude $|\tilde{h}(\omega)|$



- High pass: DC cannot propagate
- Low pass: e.g. probe duration
- 8...12 THz: Restrahlen band of GaP

Spintronic THz emitter: from *S* **to** *E*



Conclusion

Developed reliable extraction method:

 $\begin{array}{ccc} \text{Measured electro-} & h & \\ \text{optic signal } S(t) & & \\ \end{array} \xrightarrow{h} & \\ \text{directly behind the sample} \end{array}$

Application: quantitative measurement of ultrafast charge transfer in e.g.

- Spintronic multilayers
- Photovoltaic structures
- Molecules: photochemical processes
 - ... so far very rarely implemented

$I(t) \begin{pmatrix} A^+ & E(t) \\ & & & \\ B^- & & \end{pmatrix}$

Future extensions:

Better reference emitters: thinner, stronger, flat spectral output