



# Attosecond Spintronics

Light wave dynamics driving attosecond coherent spins  
and topological systems

Markus Münzenberg

F. Siegrist *et al.*, Light-wave dynamic control of magnetism,  
Nature **571**, 240–244 (2019)

Florian Siegrist, **Martin Schultze**, TU Graz/ MPQ Garching  
**Sangeeta Sharma**, MBI Berlin/ MPI Halle



# Collaborations



@spintronicsHGW



## Attosecond spin dynamics:

Florian Siegrist, Martin Schultze, TU Graz/ MPQ  
Garching

Sangeeta Sharma, MBI Berlin, MPI Halle

## THz emitter:

J. Nötzold, S. Mährlein, Lukas Braun, Tobias  
Kampfrath, *Fritz Haber Institute*

Marco Battiato, Pablo Maldonado, Peter  
Oppeneer, *Uppsala University*

F. Freimuth, Y. Mokrousov, S. Blügel, *FZ Jülich*

Mathias Kläui, *Mainz University*



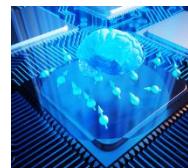
Priority program  
Topologische  
Isolatoren



Priority program  
Skyrmionics



FET Open SpinAge



META ZIK PlasMark



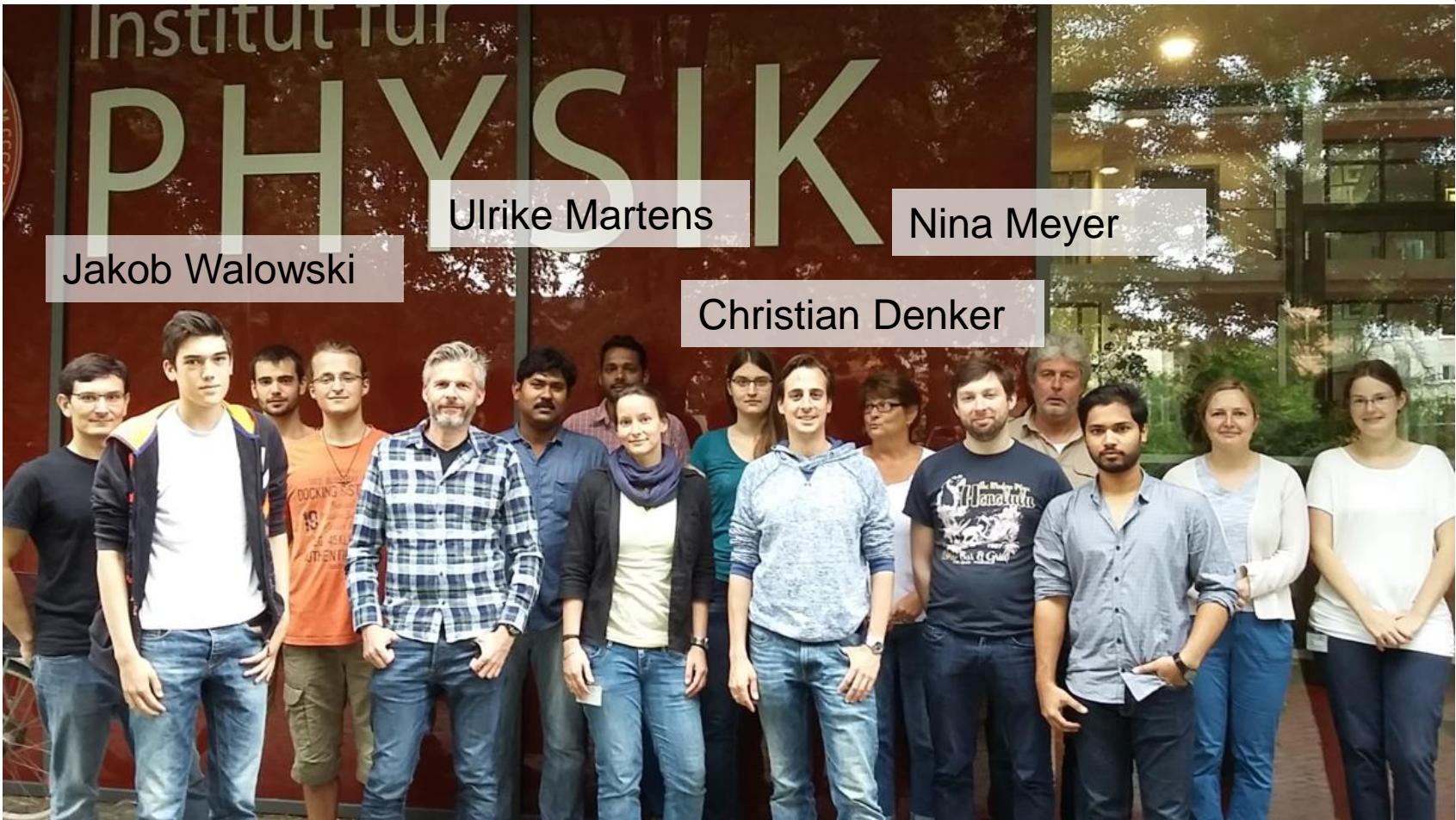
Bundesministerium  
für Bildung  
und Forschung

DAAD

# Contributions



@spintronicsHGW



Priority program  
Topologische  
Isolatoren



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Skyrmionics



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DAAD

# Outline

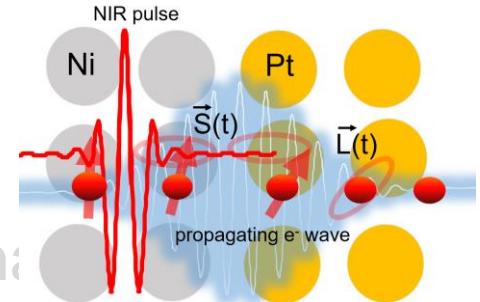


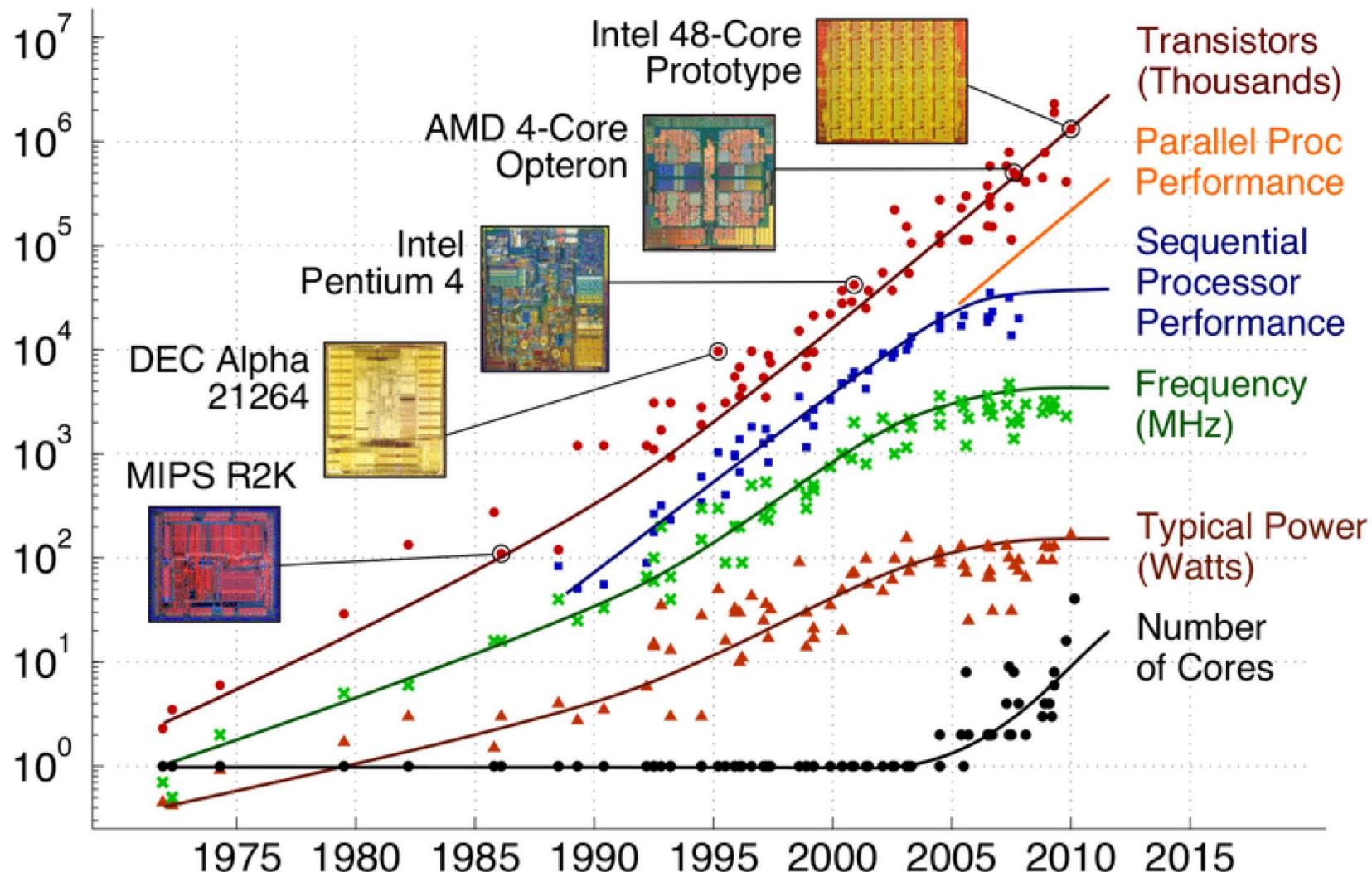
- A coherent attosecond spintronics?
- The nature of femtosecond spin dynamics
- THz spintronic emitter – *noncoherent*
- *Topological Insulators*
- Lightwave electronics - *coherent*
- Summary

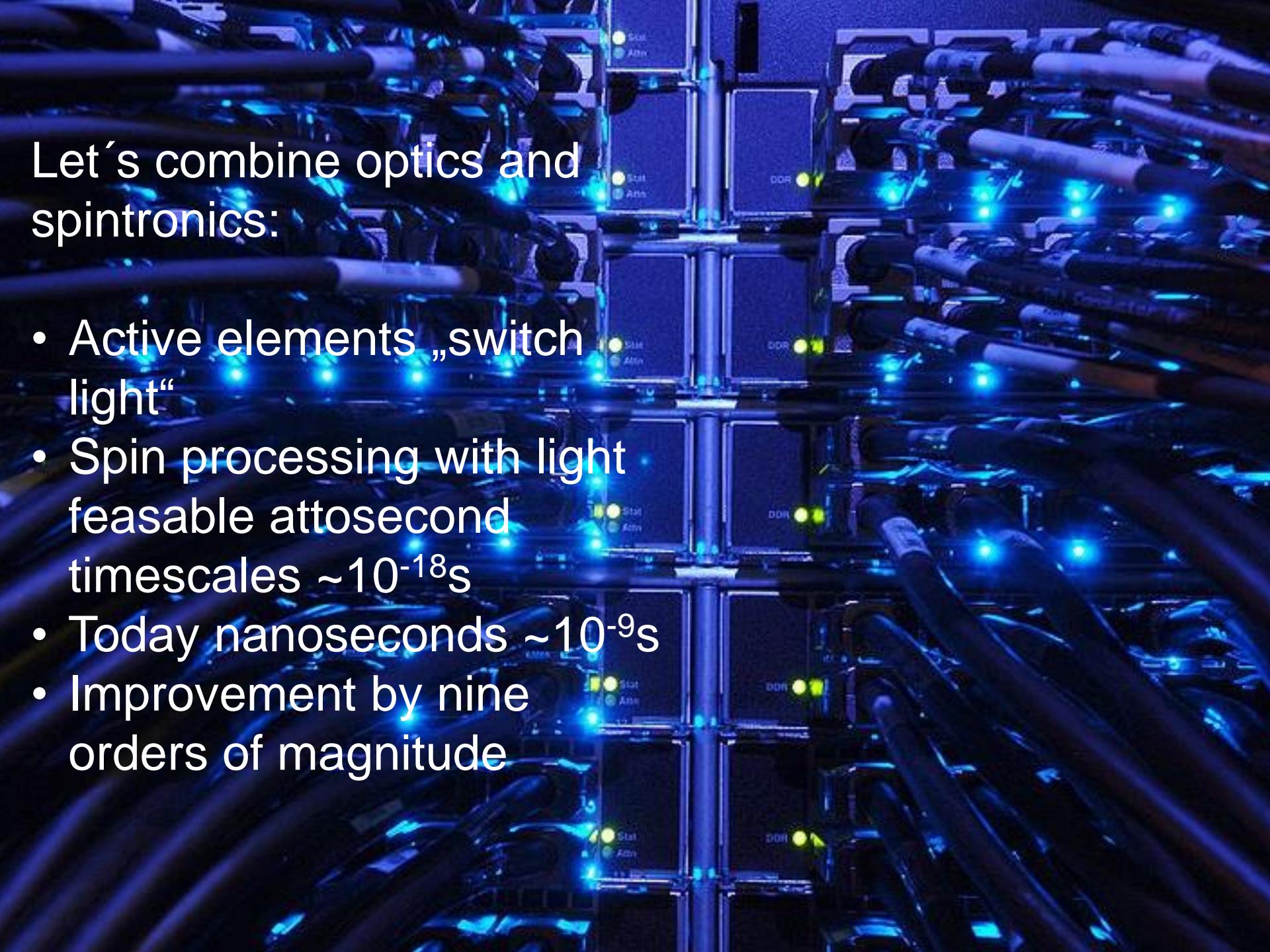
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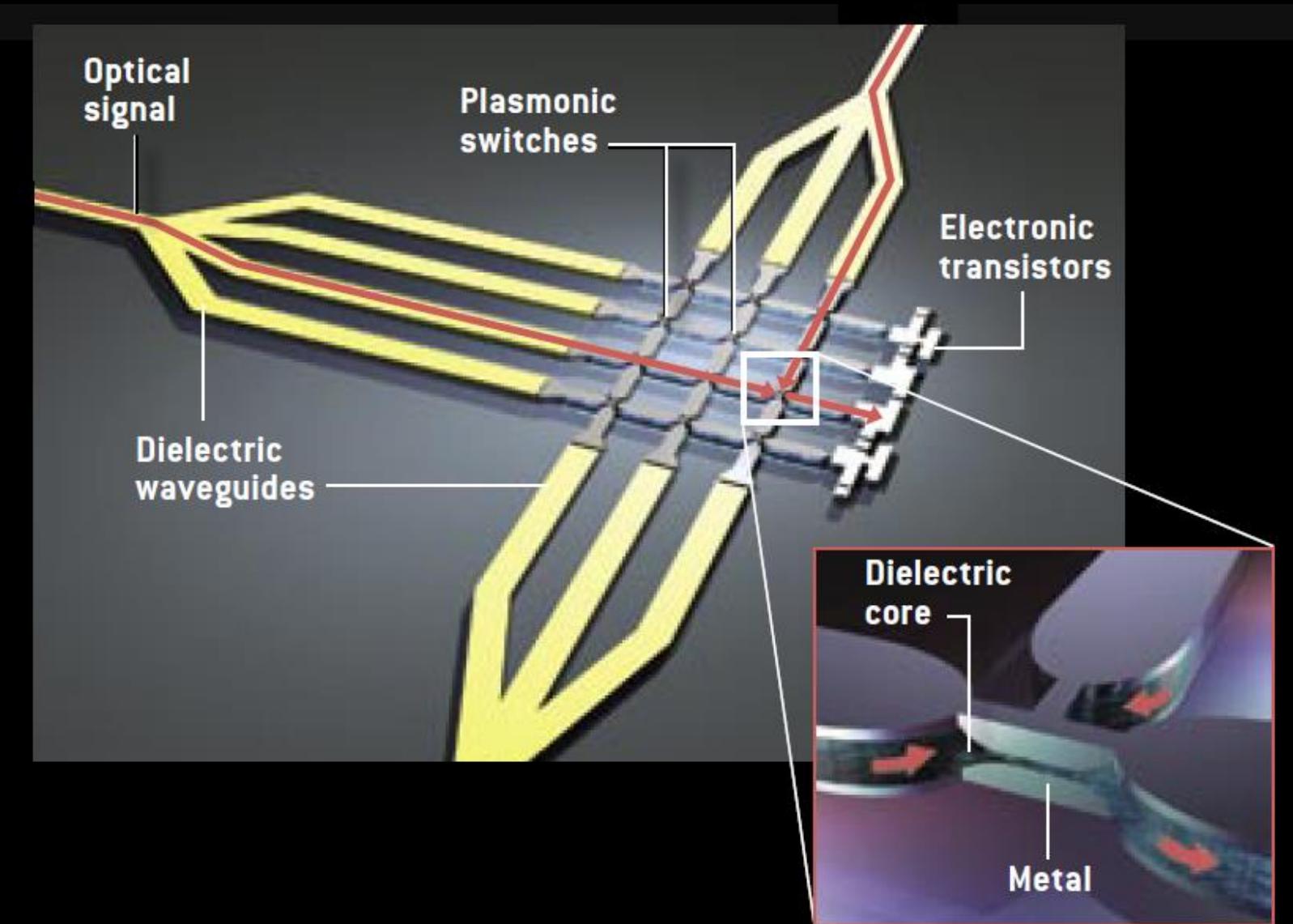




Let's combine optics and spintronics:

- Active elements „switch light“
- Spin processing with light feasible attosecond timescales  $\sim 10^{-18}\text{s}$
- Today nanoseconds  $\sim 10^{-9}\text{s}$
- Improvement by nine orders of magnitude

# Squeeze light: plasmonics

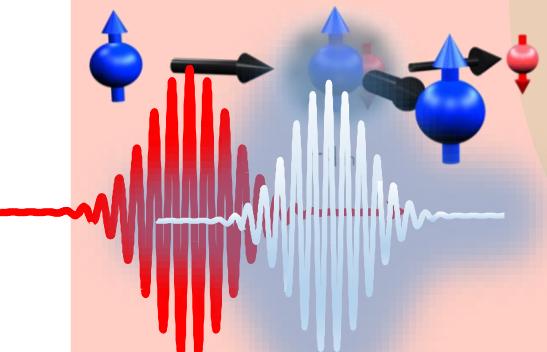


# Novel spintronic Photonic THz applications

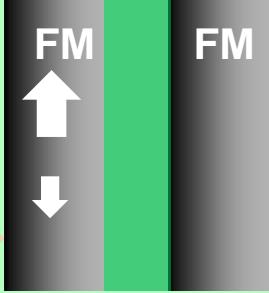


## Photonics

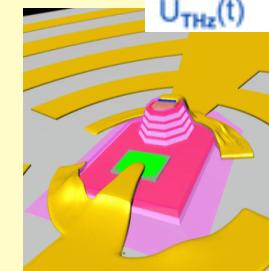
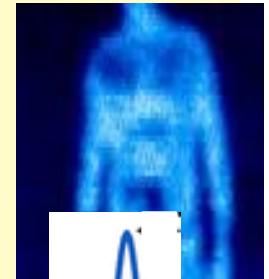
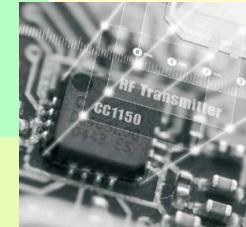
Attosecond  
coherent  
clocking



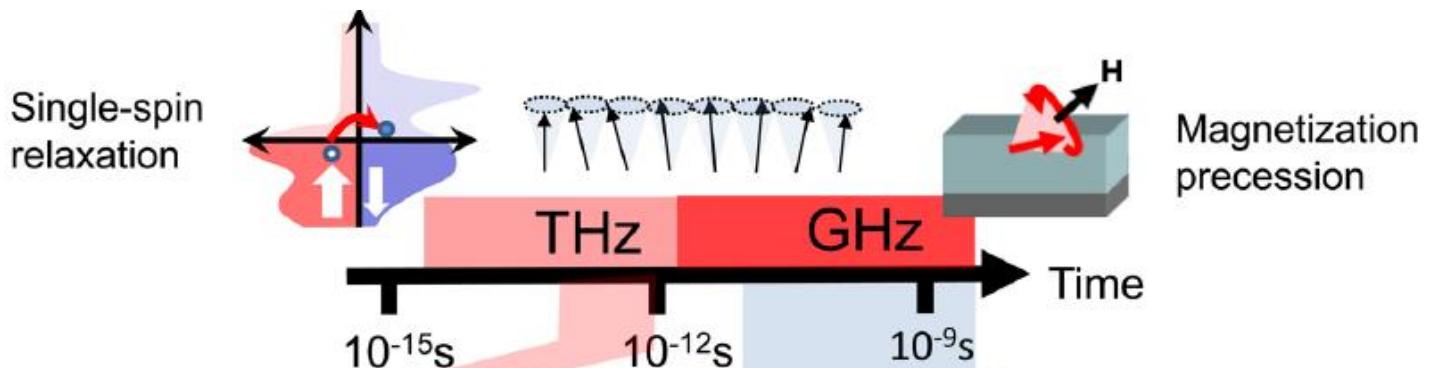
## Spintronics



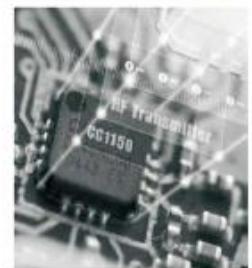
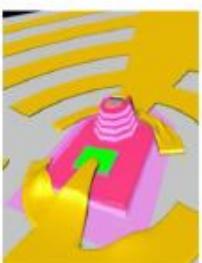
## Electronics THz applications



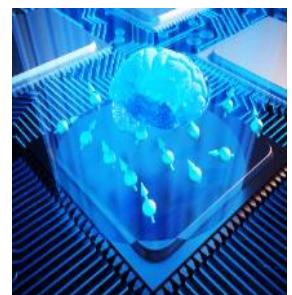
# Novel spintronic THz applications



Applications: security, bio scanner, pharmaceutics and food control, mobile phone, inter-chip wireless, data bit addressing and transfer



**Brain inspired computing - SOT oscillators addressed in plasmonic chips**



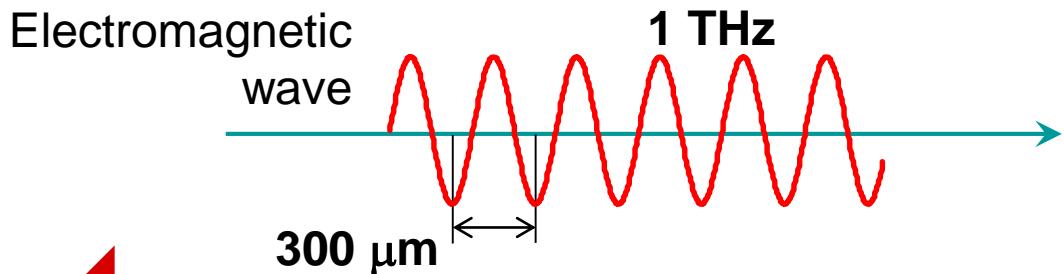
**Communication and data frequencies:**

3G and LTE: 0.7 to 2.6 GHz 5G: 24.25 – 27.5 GHz

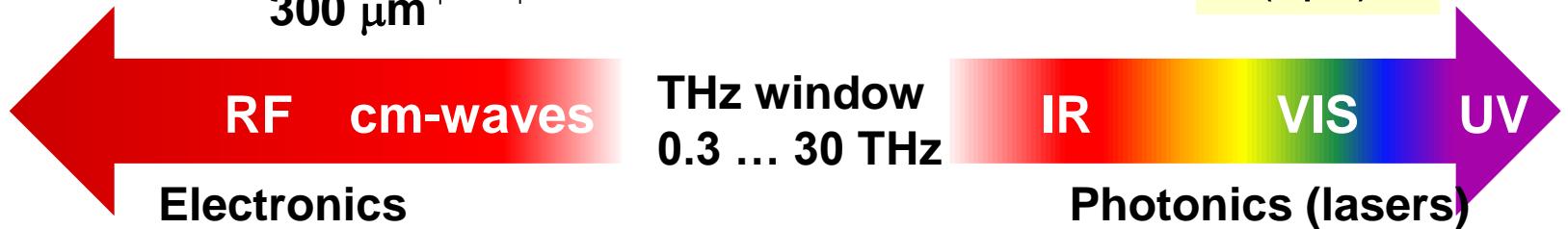
*6G reaches THz frequency: > 95 GHz to 3 THz range*

FET Open SpinAge

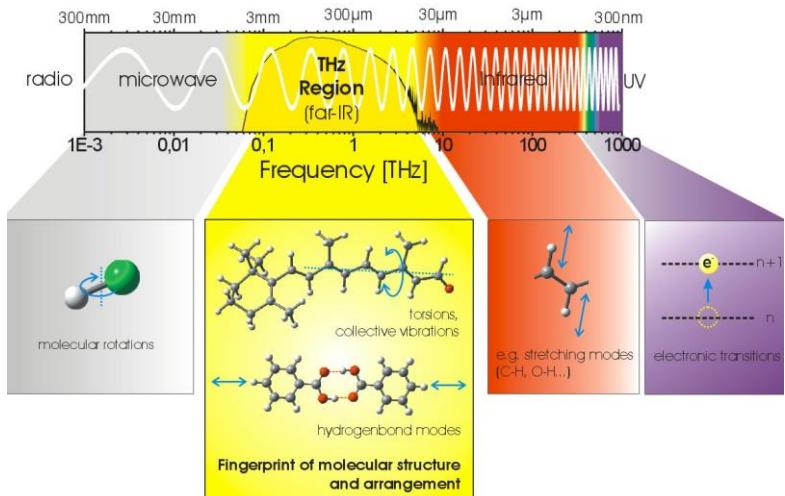
# Novel spintronic THz applications



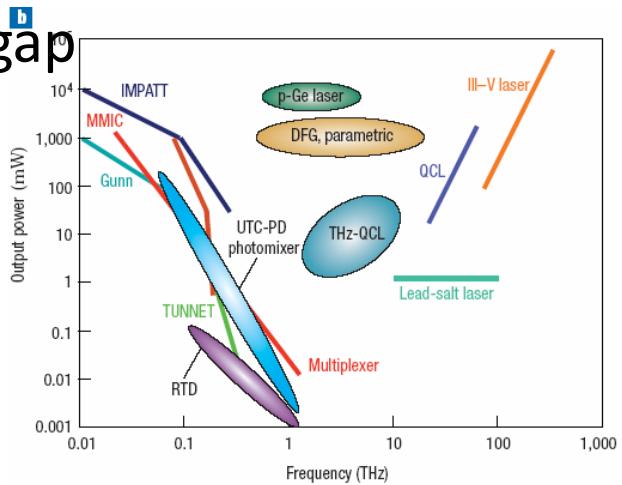
$$\begin{aligned}1 \text{ THz} &= 10^{12} \text{ Hz} \\ &= (1\text{ps})^{-1}\end{aligned}$$



Biophysical and medical sensing



Novel THz sources bridge the THz gap



META ZIK PlasMark

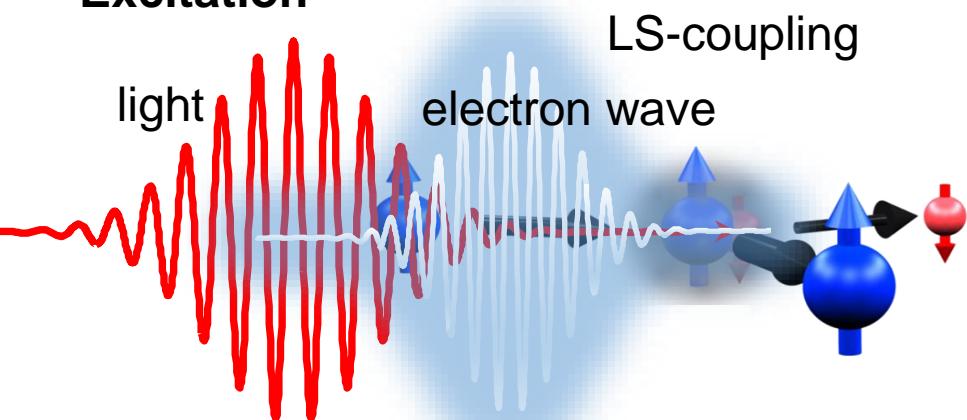
ZENTREN FÜR  
INNOVATIONSKOMPETENZ  
UNTERNEHMEN REGION  
Die BMBF-Innovationsinitiative  
Neue Länder

Bundesministerium  
für Bildung  
und Forschung

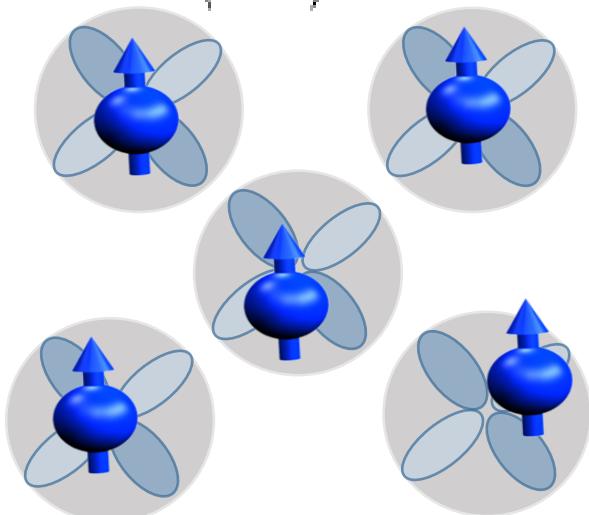
# A coherent attosecond spintronics?



## Excitation



Five atoms, evolution of  $\langle \Delta S^z \rangle$ ?



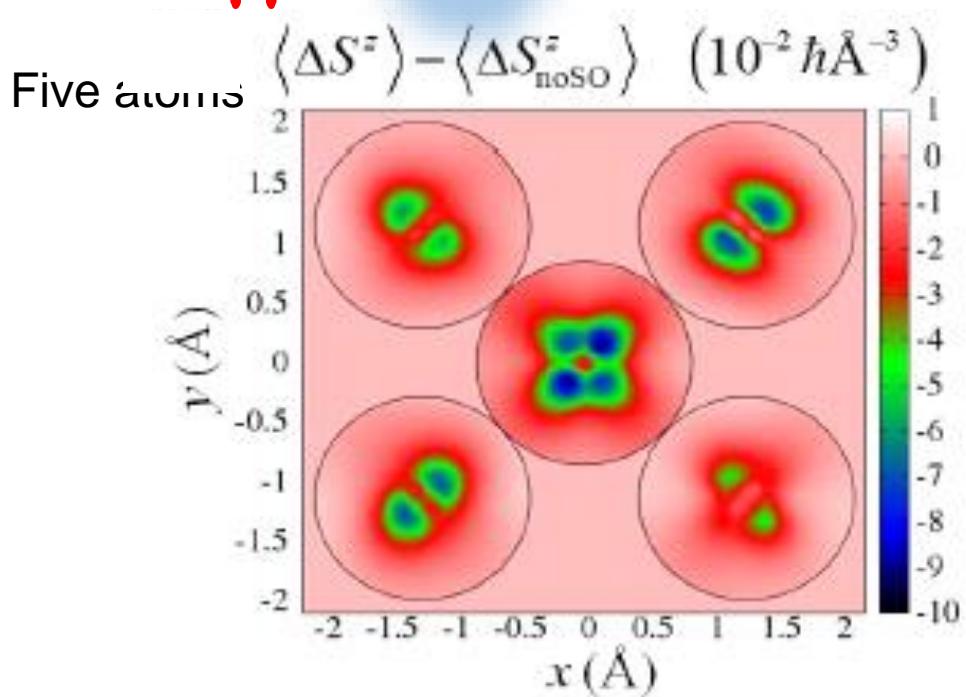
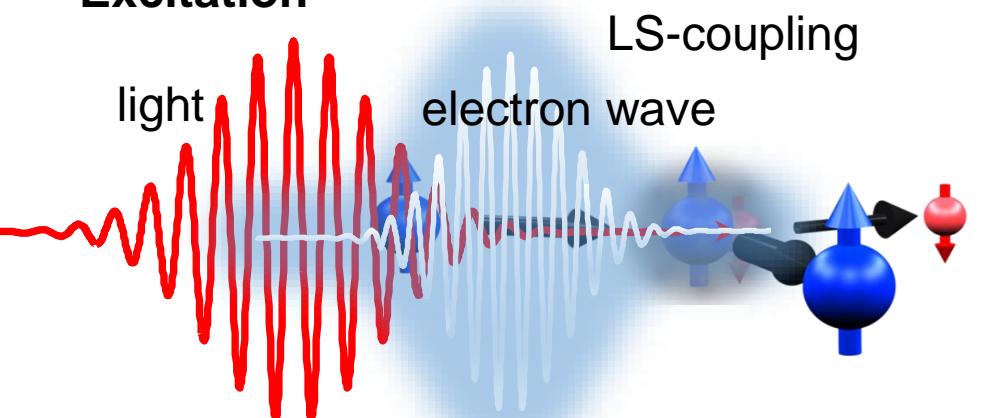
Key to understanding of spin dynamics on atto- to femtosecond timescales (1 as to 10 fs) ab-initio:

## Timeresolved Density Functional Theory (trDFT)

Wolfgang Hübner, Gouping Zhang,  
Hardy Gross, Sangeeta Sharma,  
Stefano Sanvito ...

# A coherent attosecond spintronics?

## Excitation



J. Simoni, et al. Phys. Rev. B 95, 024412 (2017)

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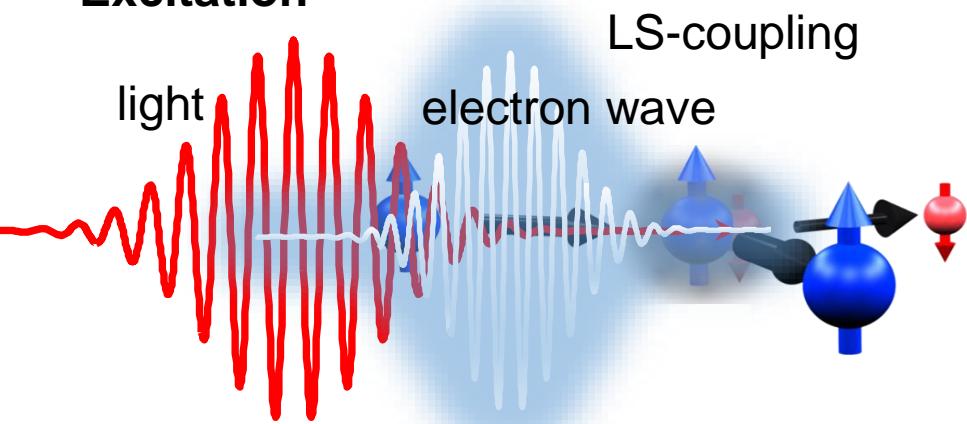
Wolfgang Hübner, Gouping Zhang, Hardy Gross, Sangeeta Sharma, Stefano Sanvito ...

LS-coupling results in evolution of the magnetization on attosecond timescales in the atom's shell

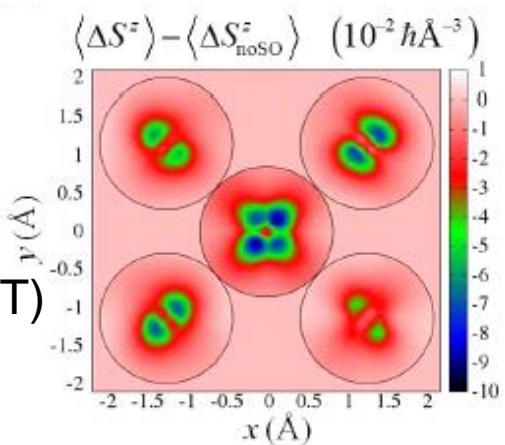
# A coherent attosecond spintronics?



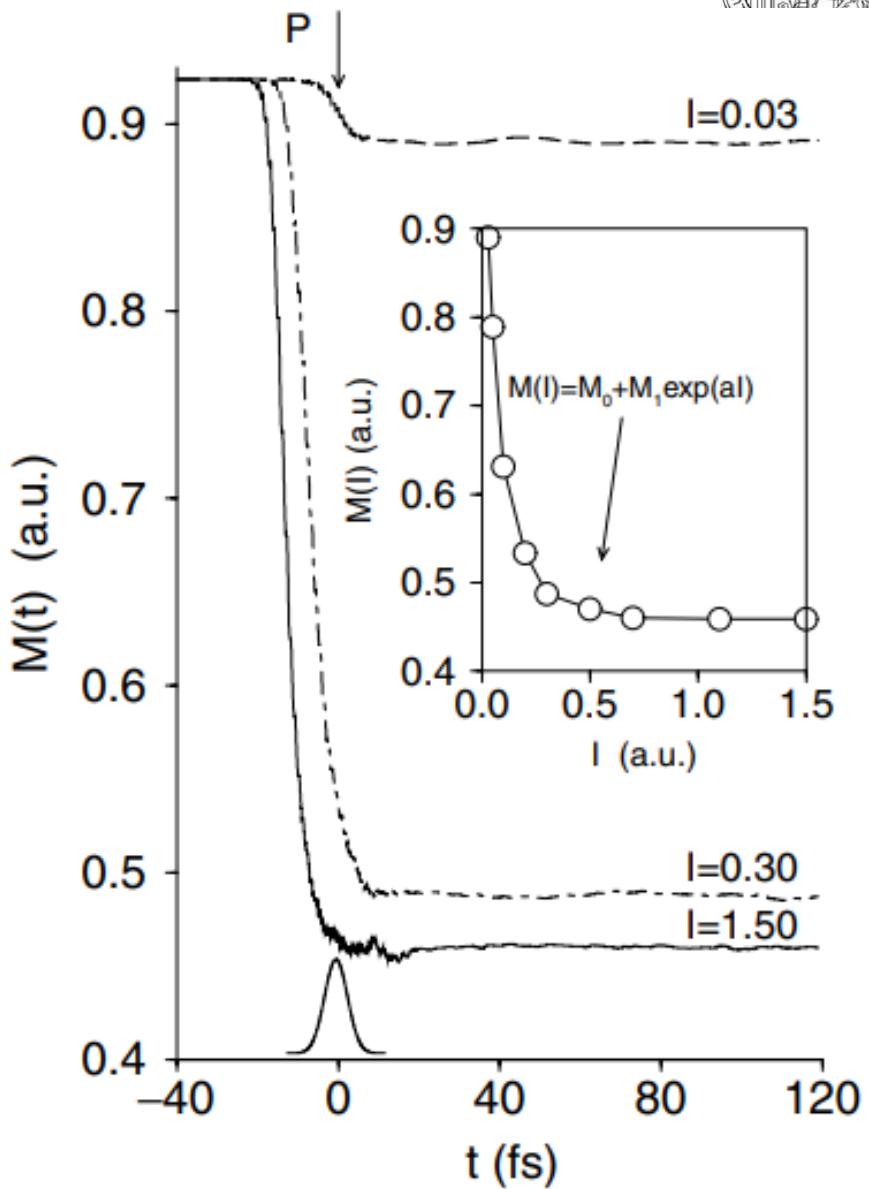
## Excitation



Time  
dependent  
density  
functional  
theory (trDFT)

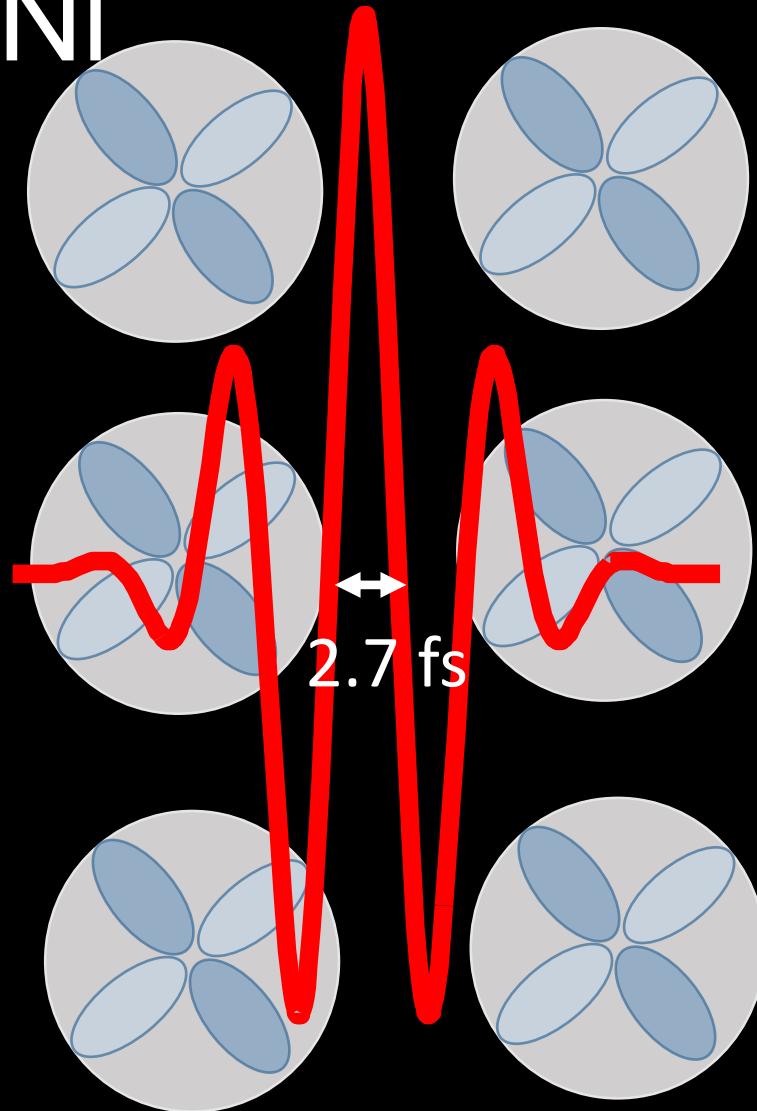


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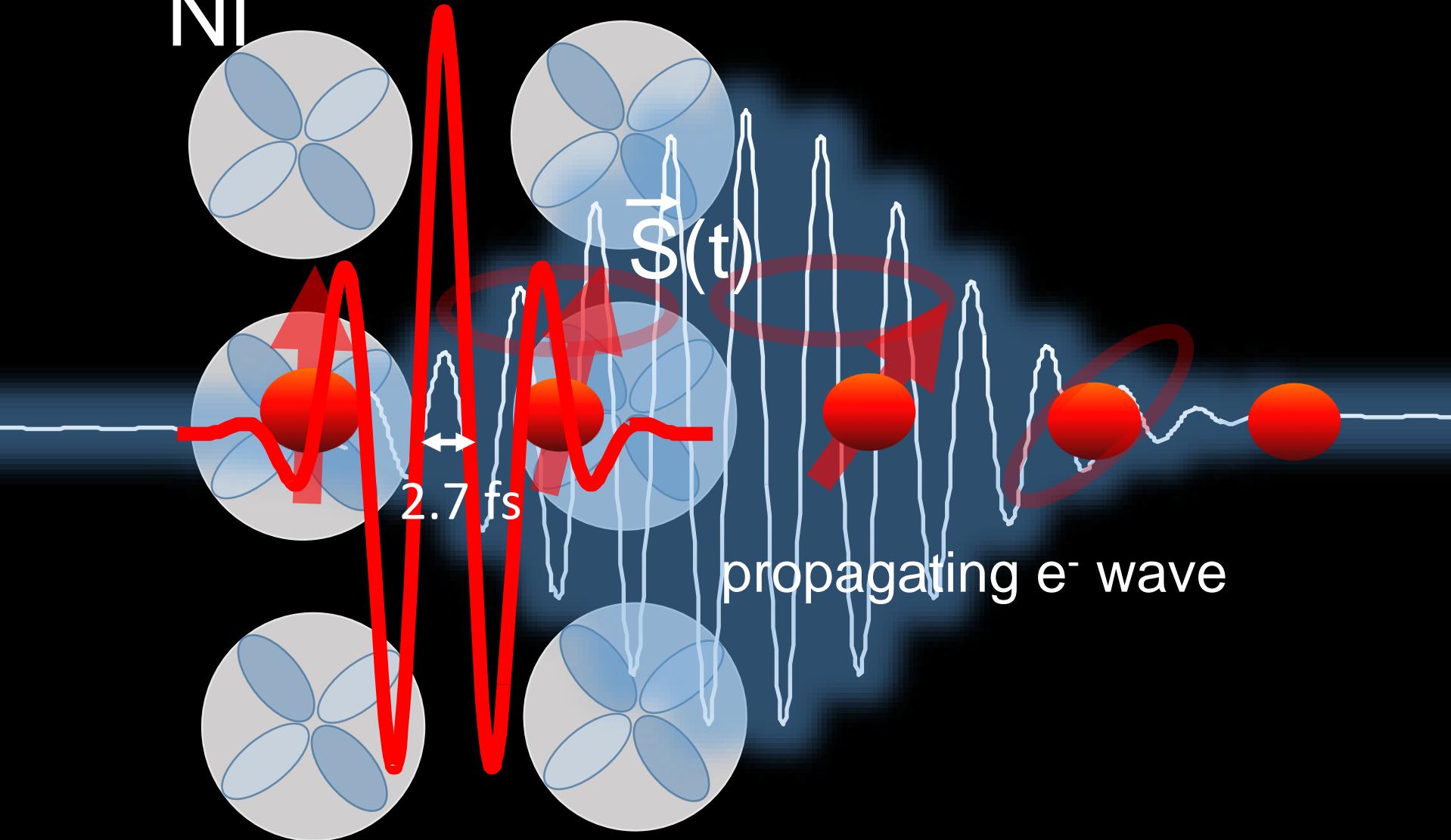
# Few cycle pulse

Ni



# Few cycle pulse

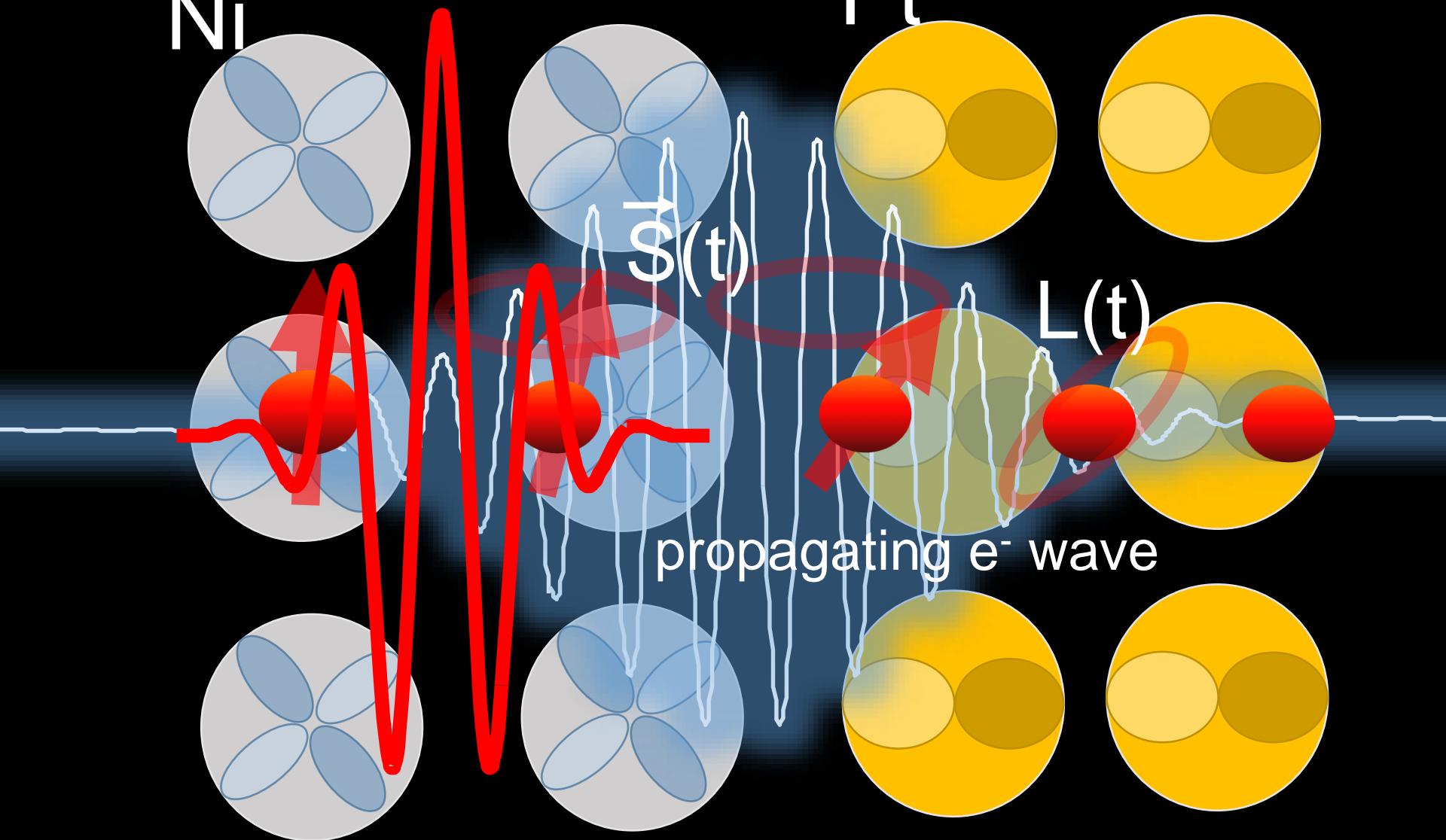
Ni



Few cycle pulse

Ni

Pt

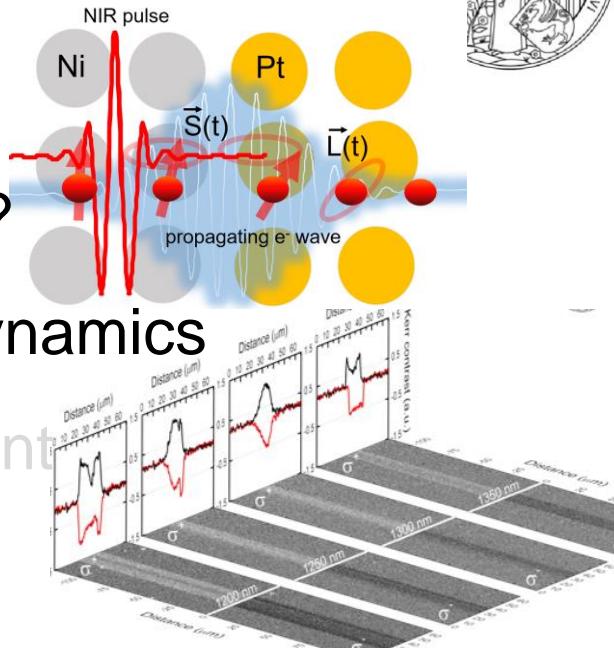


Attosecond Coherent Spintronics

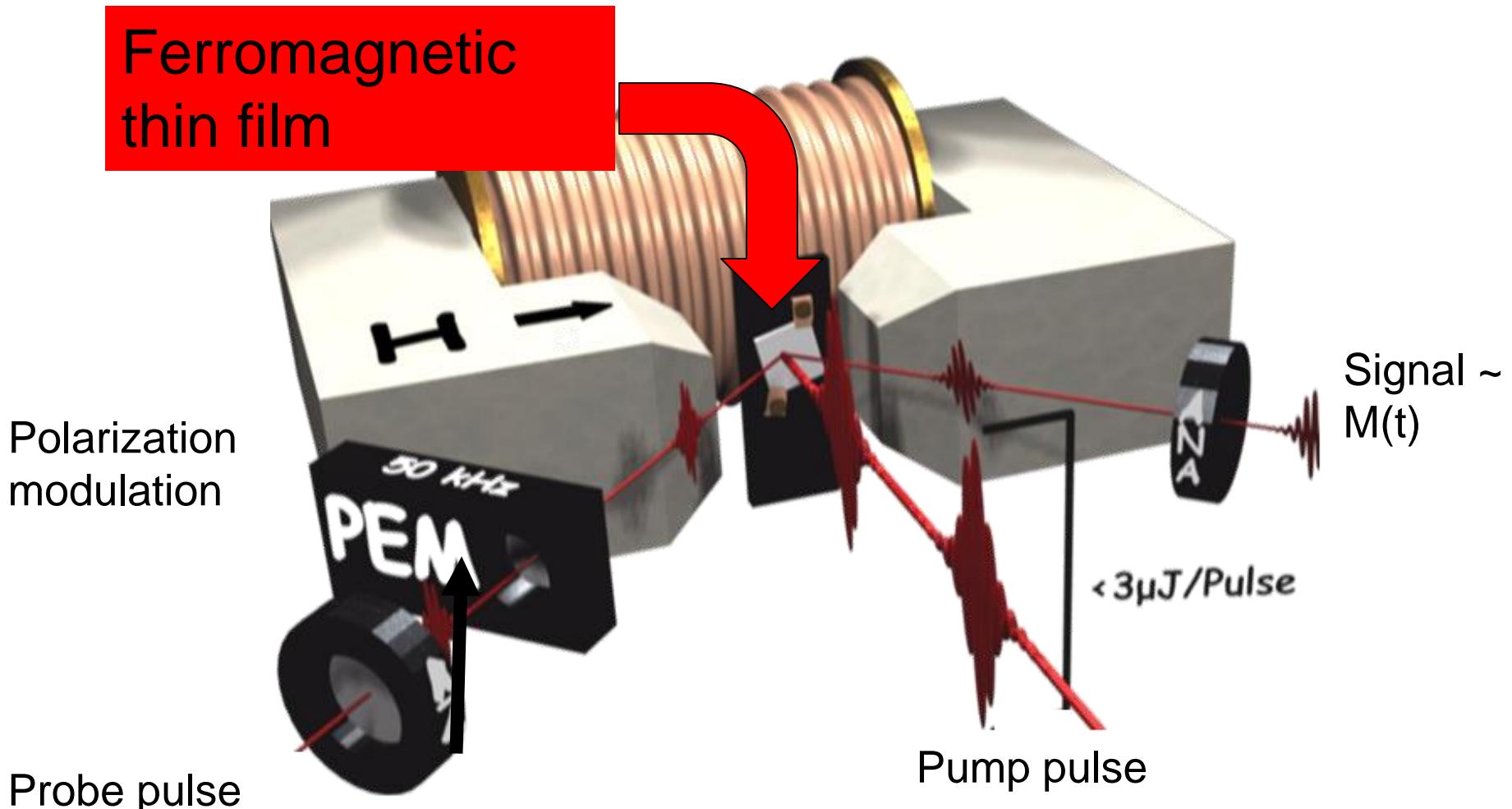
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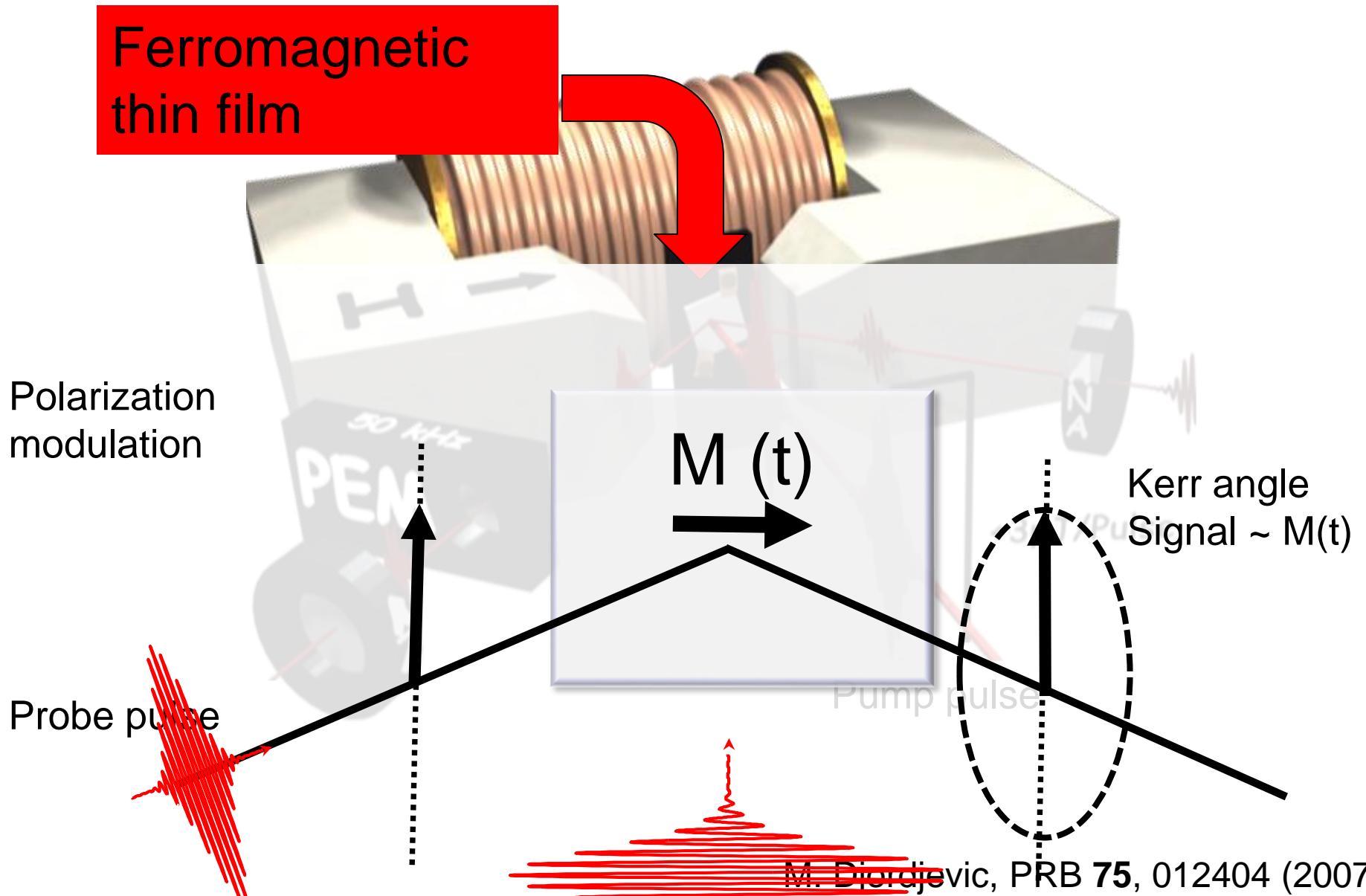


# Femtosecond pump-probe

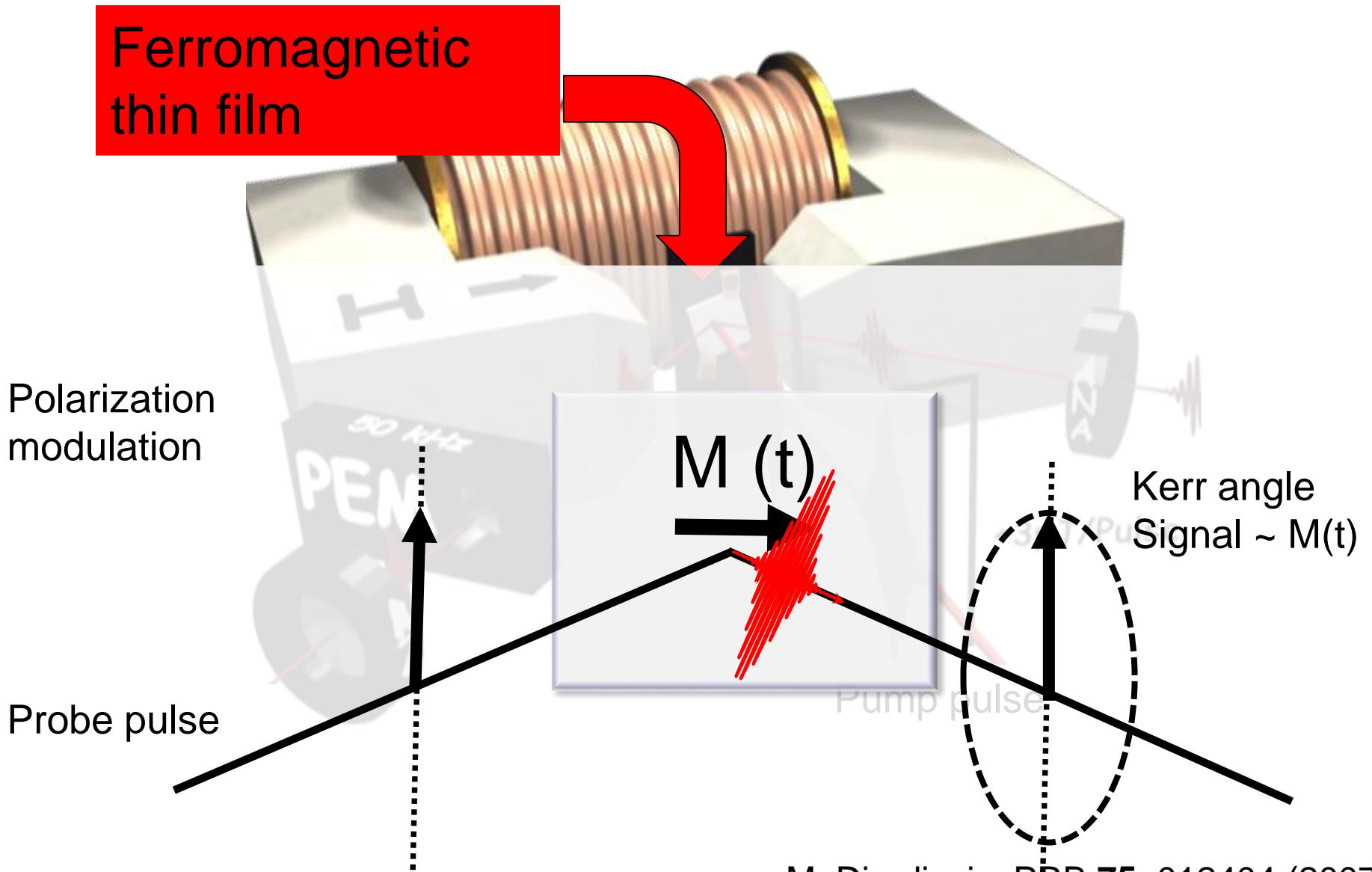


- Access to ultrafast the relaxation (40 fs,  $\lambda=800\text{nm}$ )

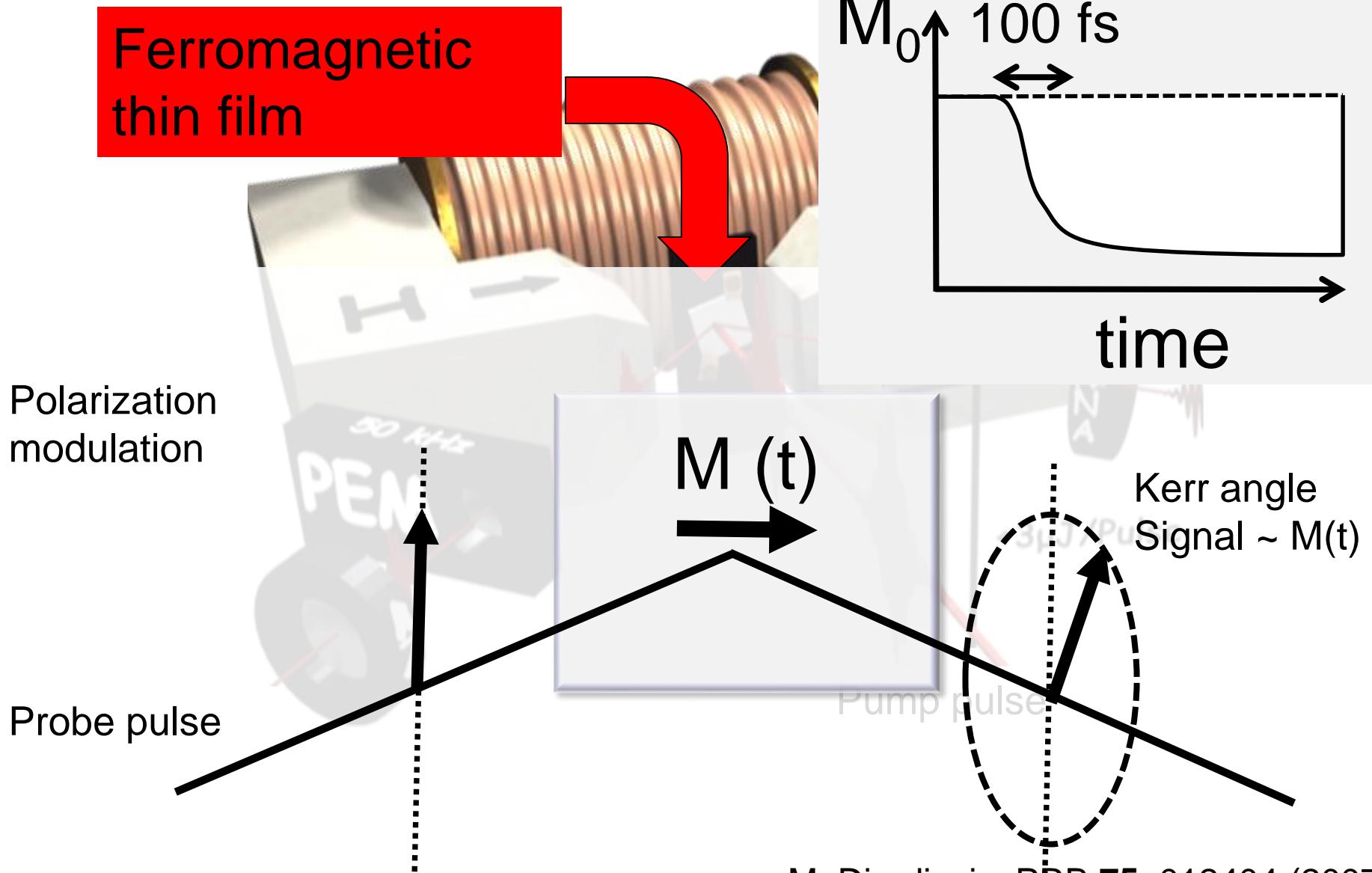
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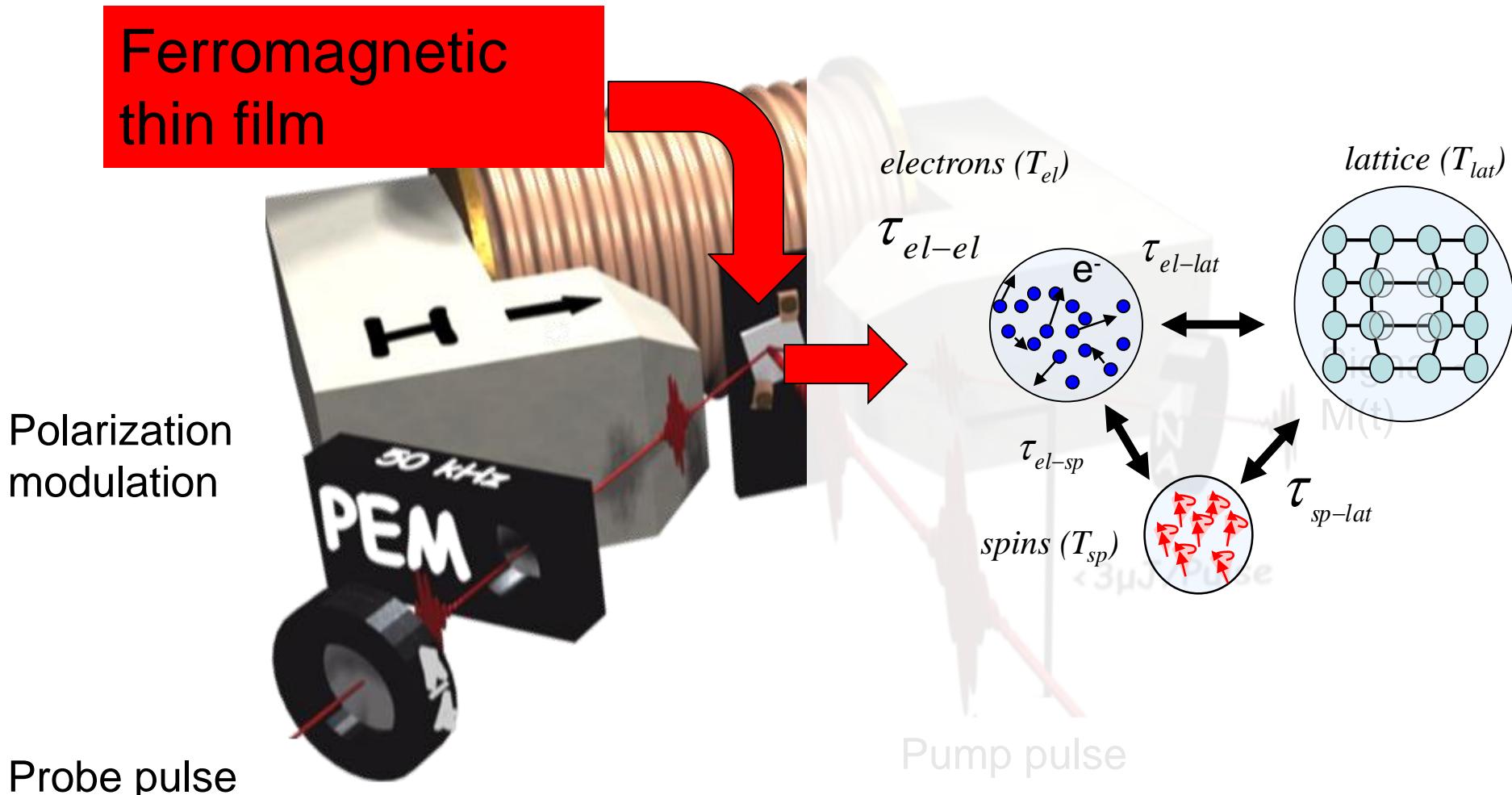
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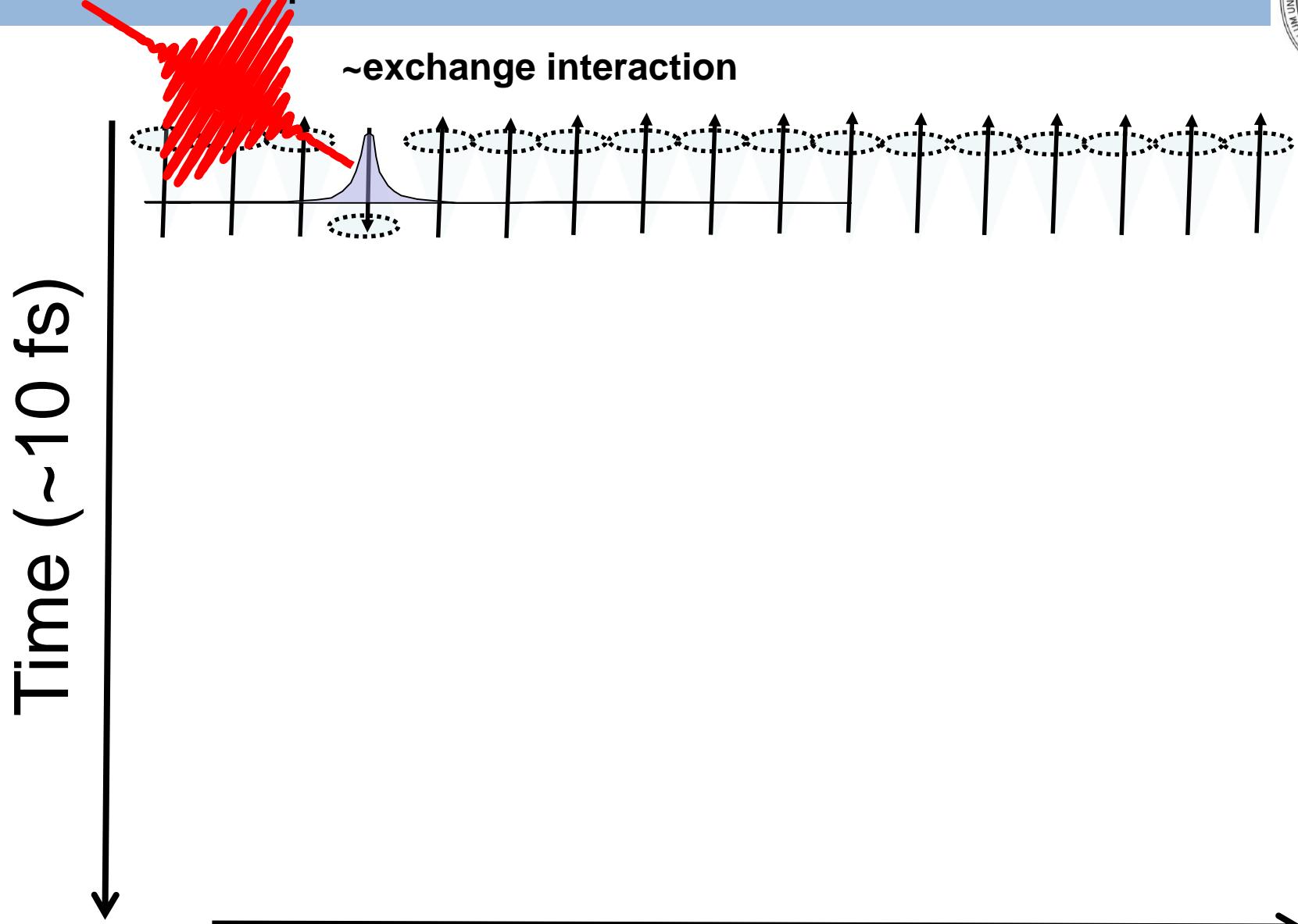
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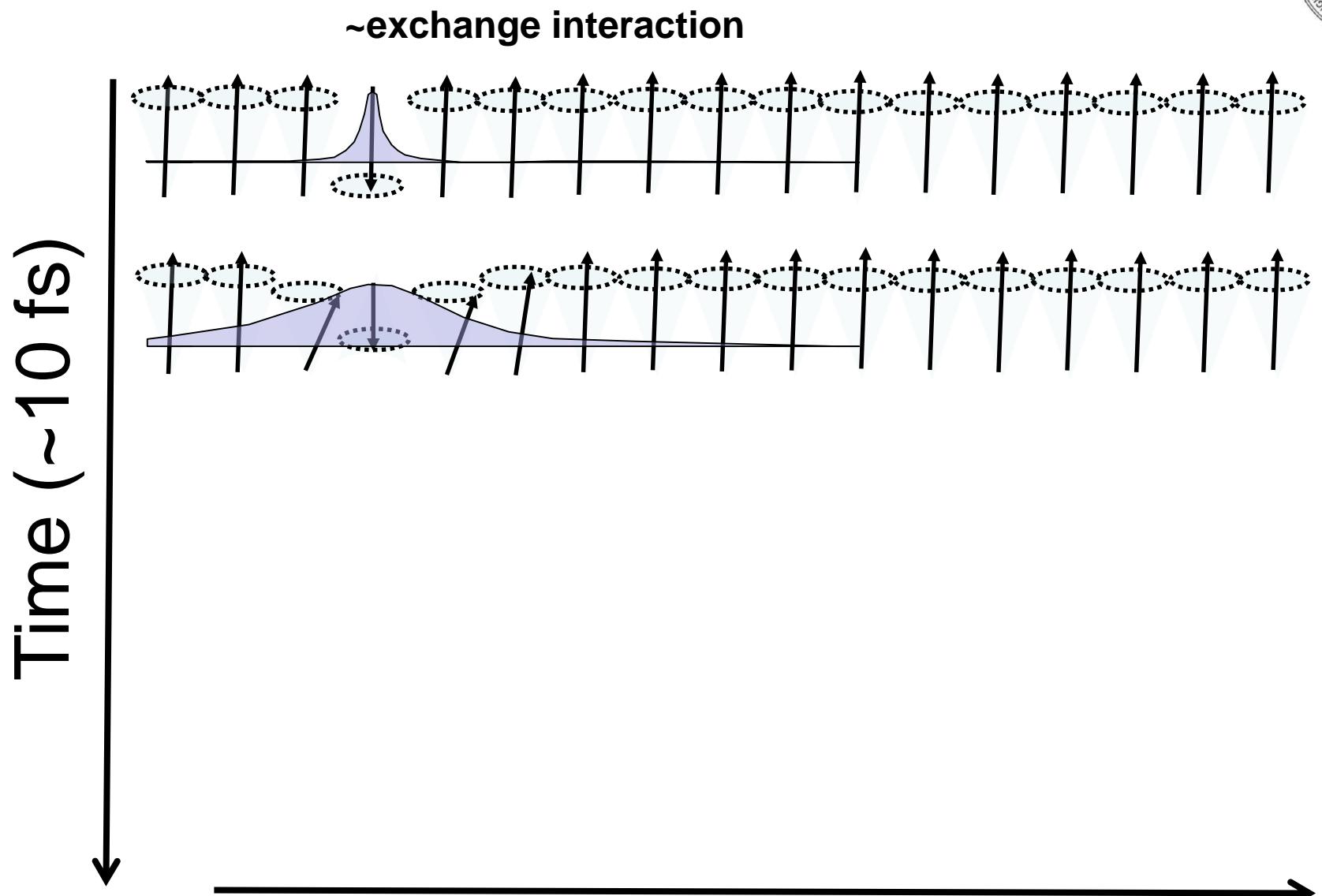


# Ultrafast: spins





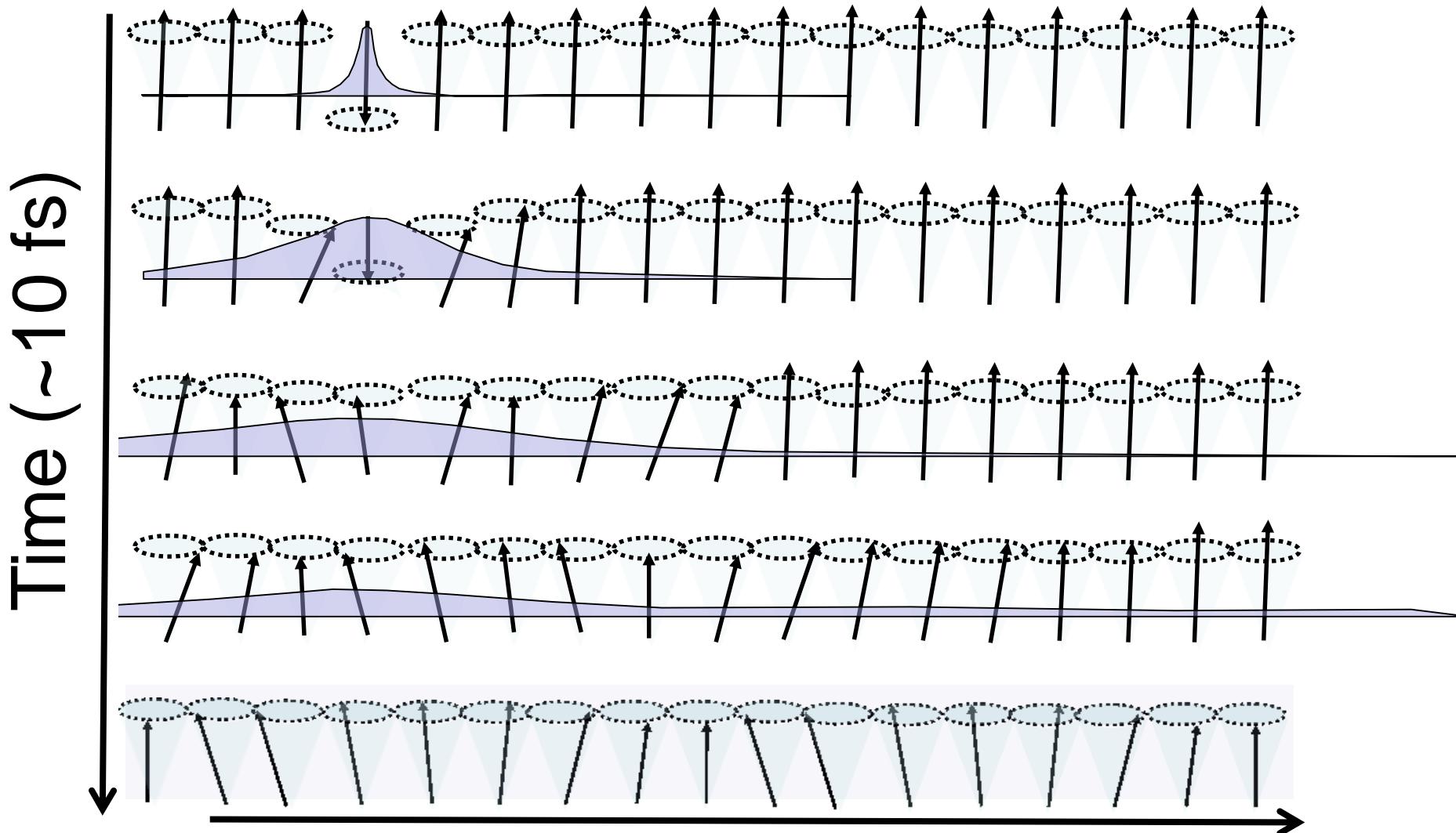
# Ultrafast: spins





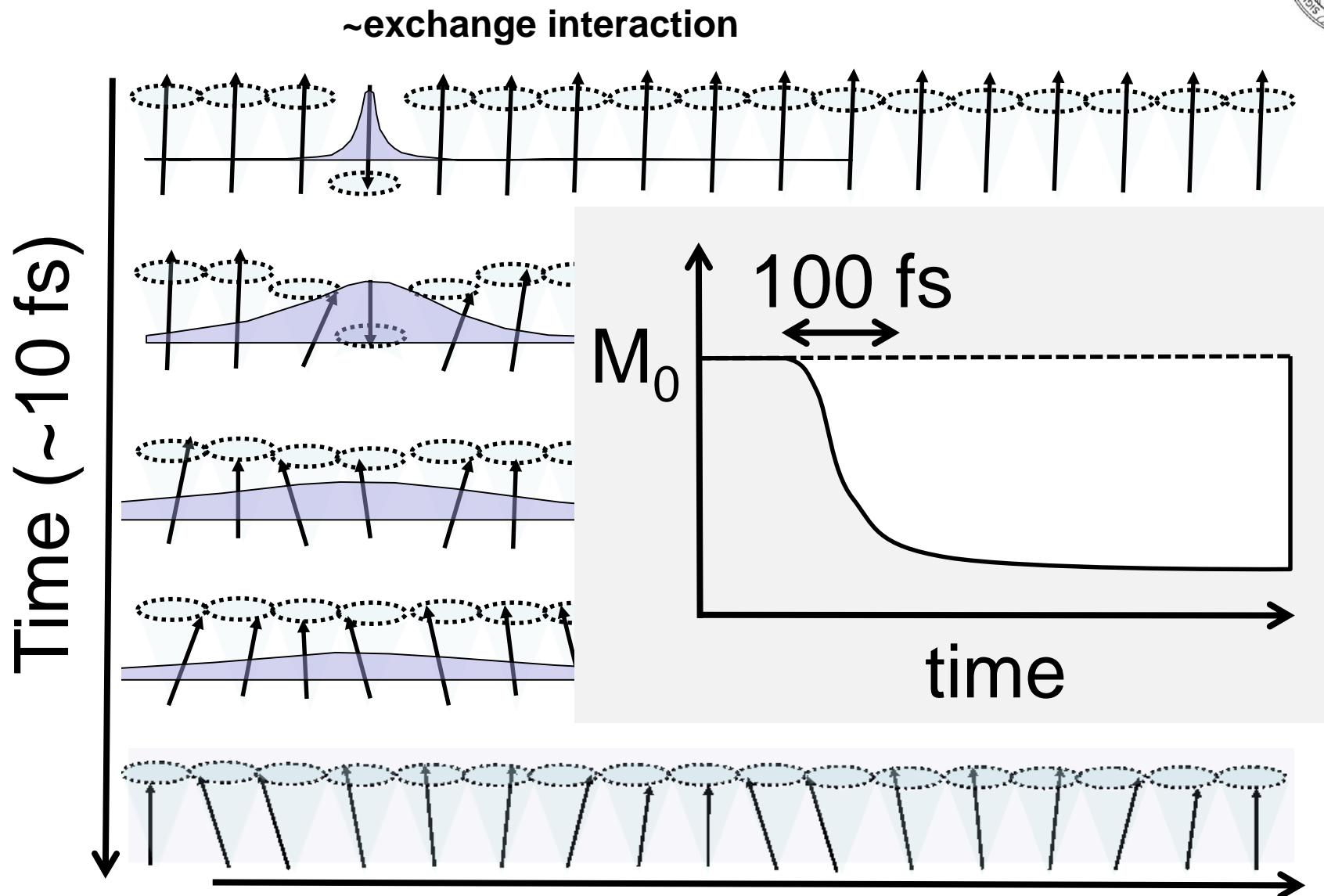
# Ultrafast: spins

~exchange interaction





# Ultrafast: spins

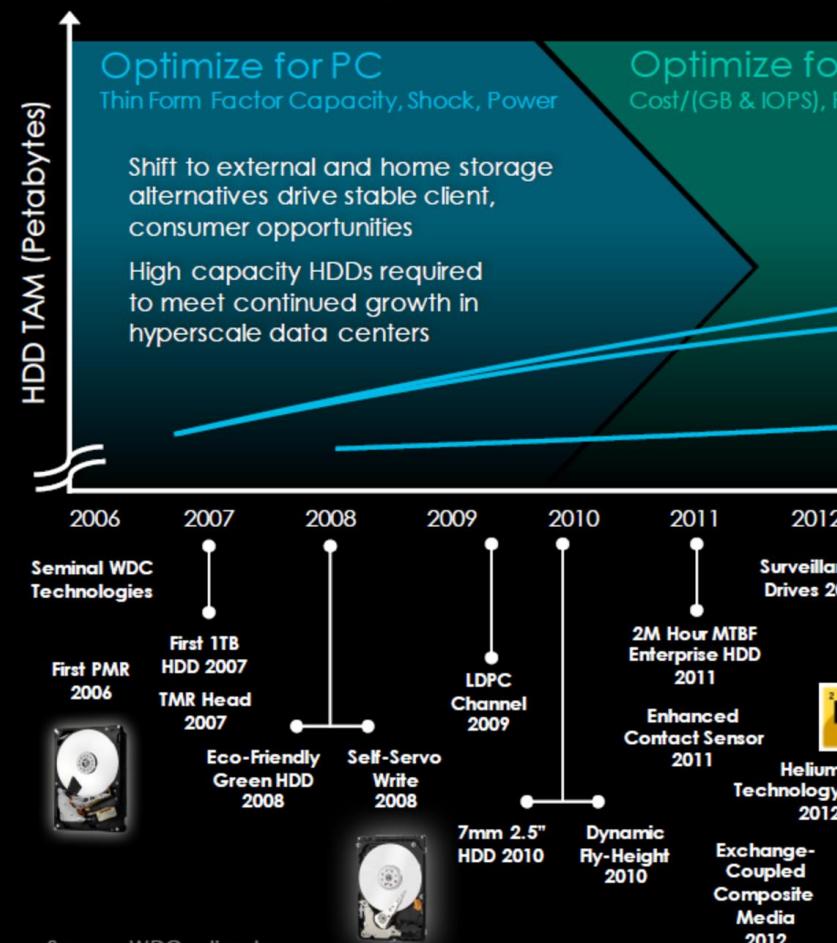


- Hard disc at fs speed



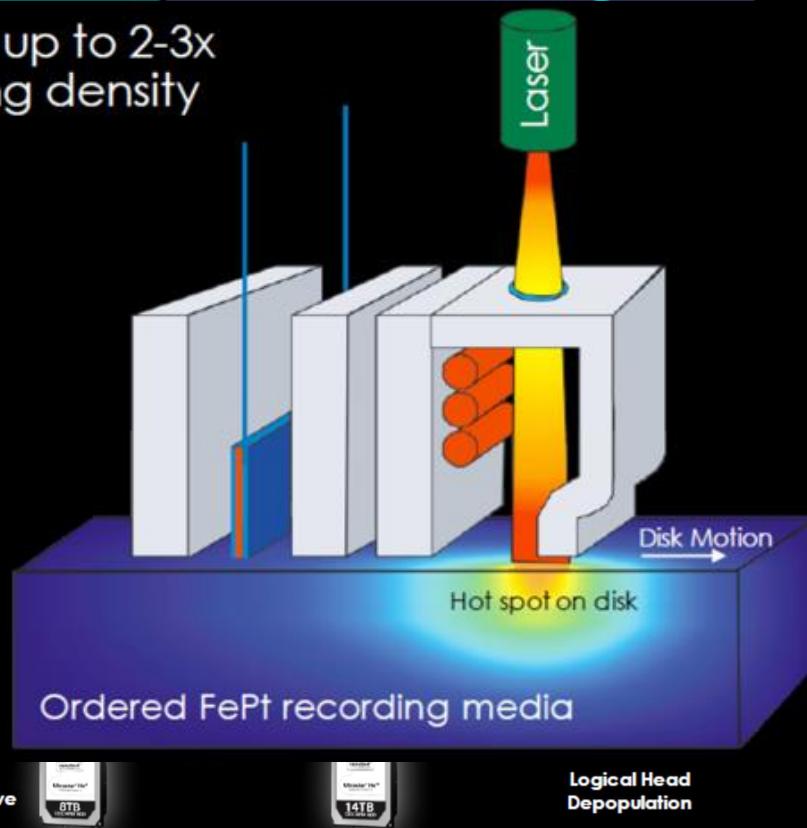
Interior of a  
hard disc

# Rich Variety of Solutions for the World's Data Storage Needs

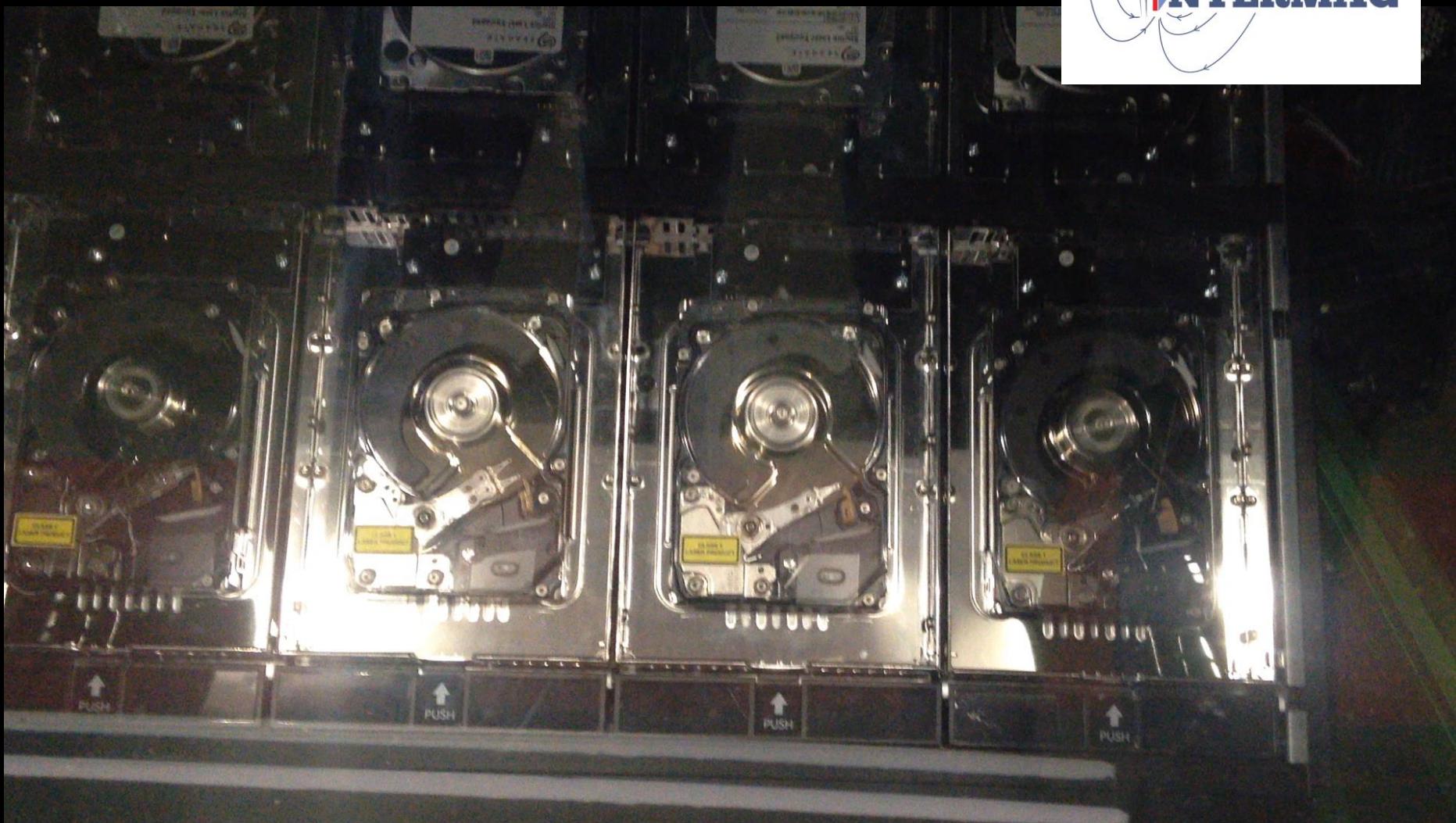


Western Digital®

- Enables up to 2-3x recording density



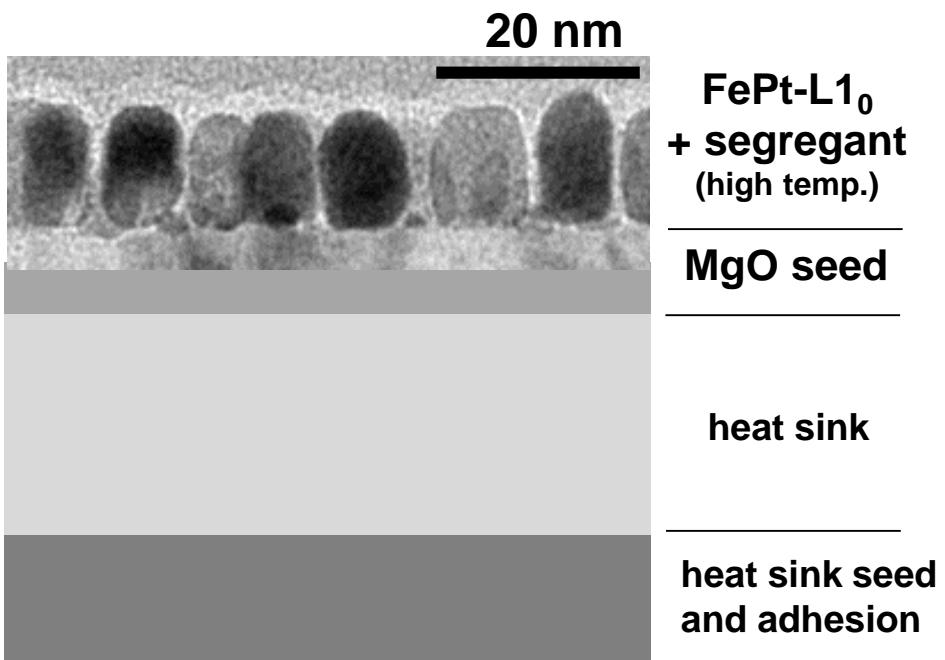
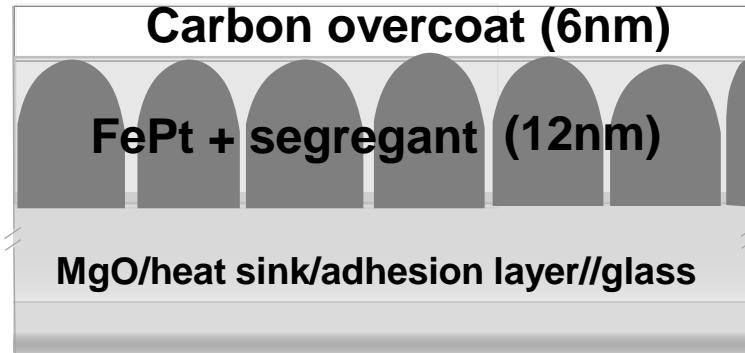
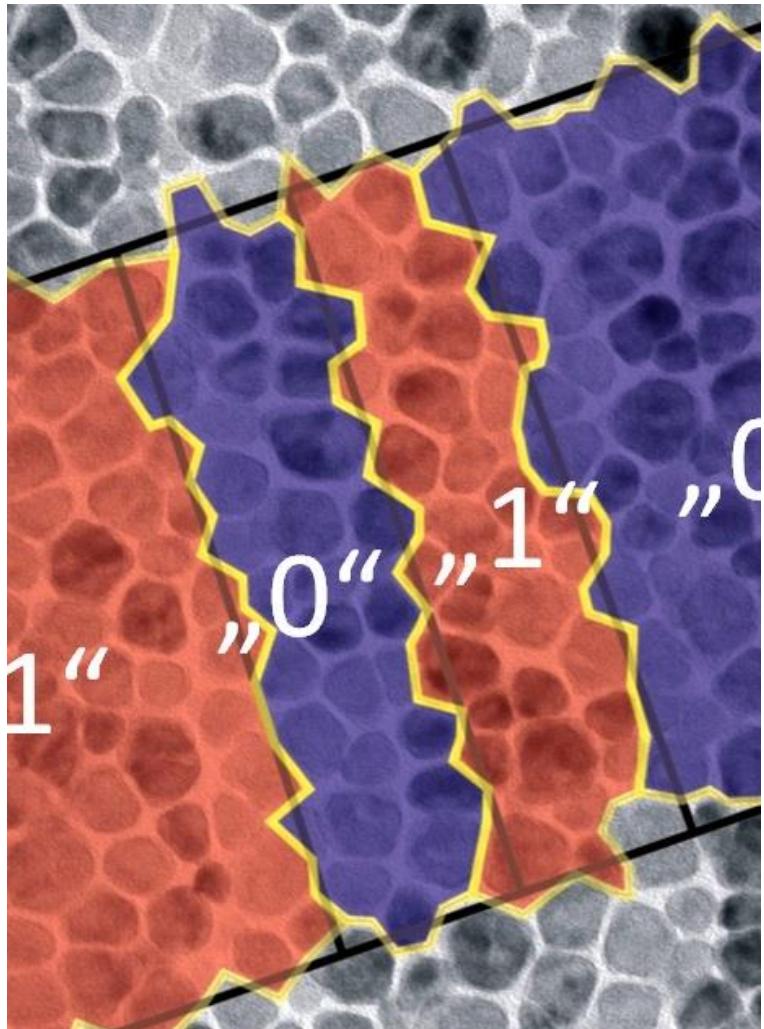
# Live demonstration HAMR Seagate server at Intermag Dublin



# Hard disc at ultrafast speed



Western Digital Corporation HAMR media,  $H_c = 4T$  at room temperature

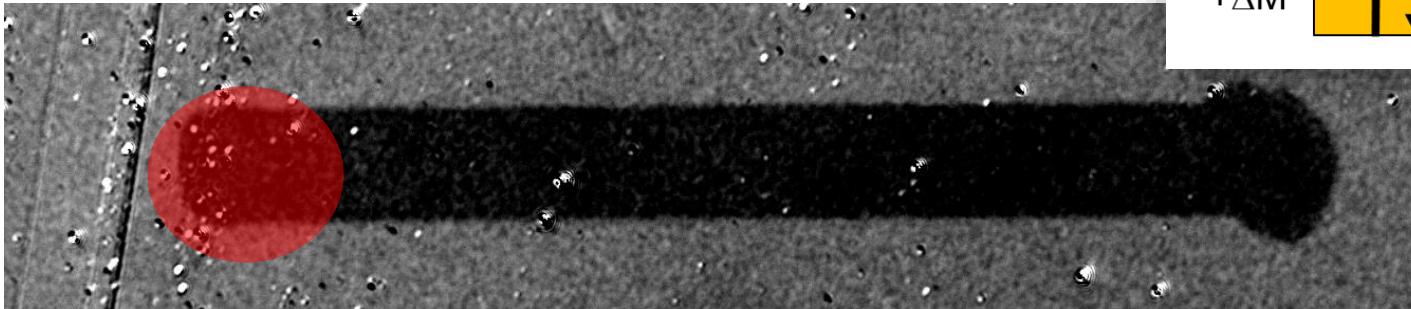


# FePt optical writing a storage media

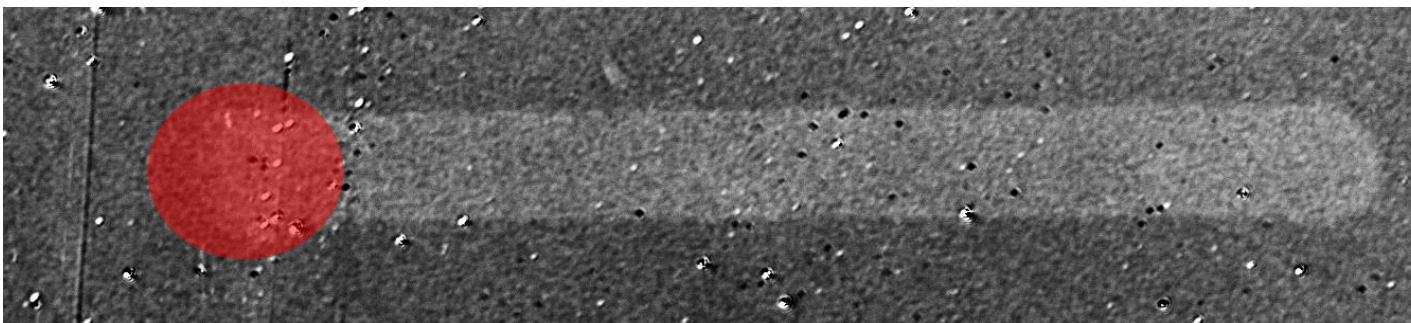


Writing using the helicity of light:

$\sigma^-$



$\sigma^+$



12.7 mJ/cm<sup>2</sup> (10 mW)

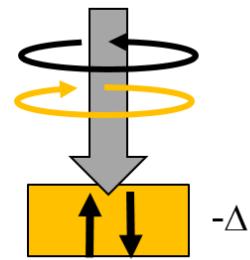
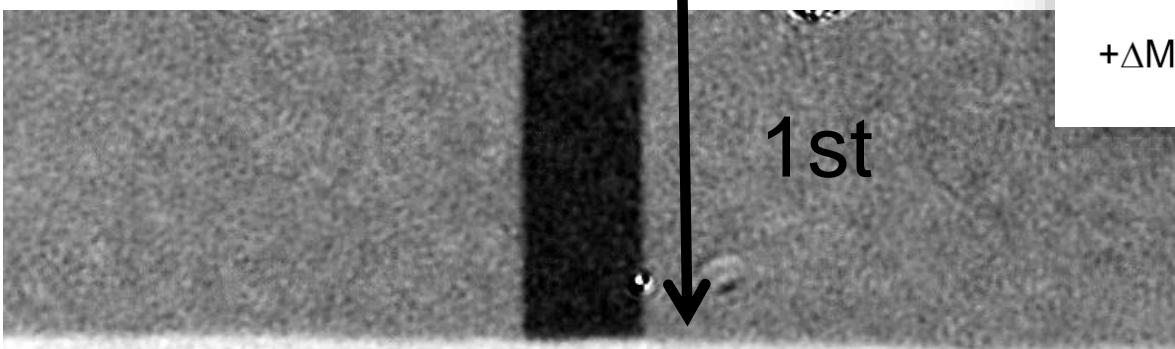
Magneto-optical contrast (Kerr microscopy)  
FePt (AgCu) granular recording media



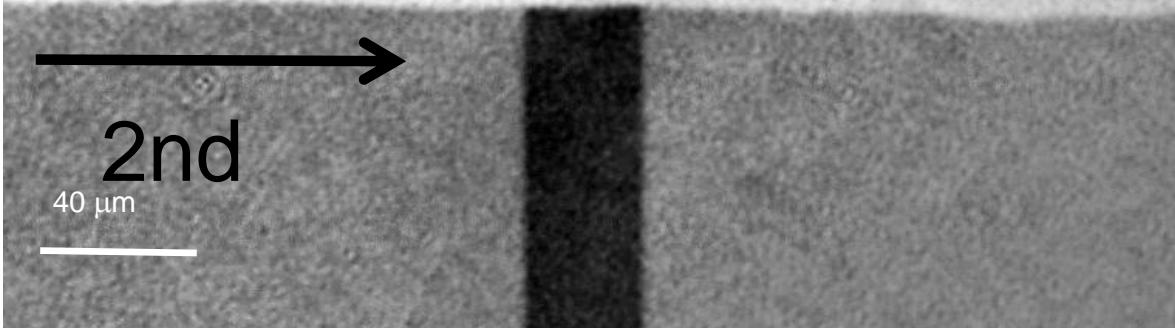
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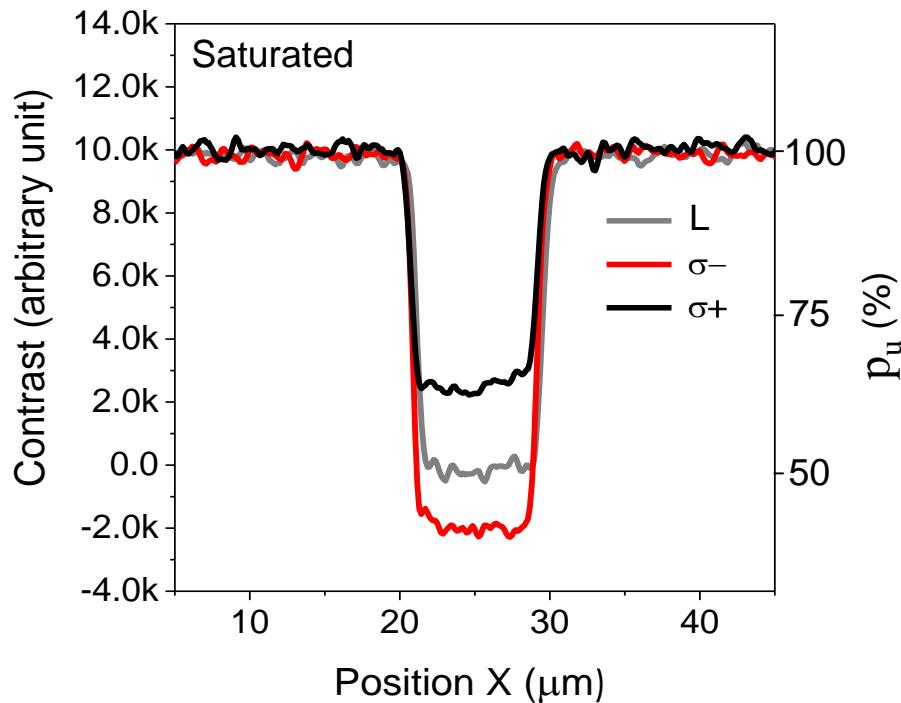
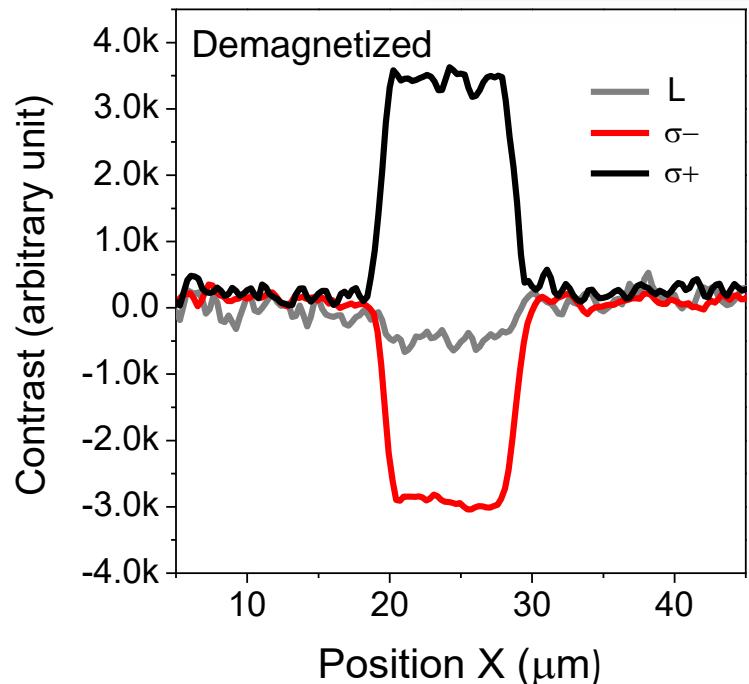
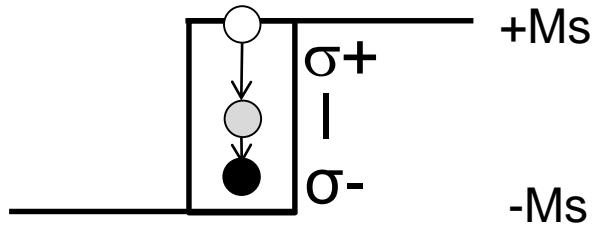
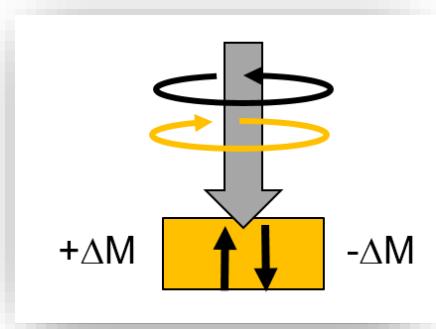
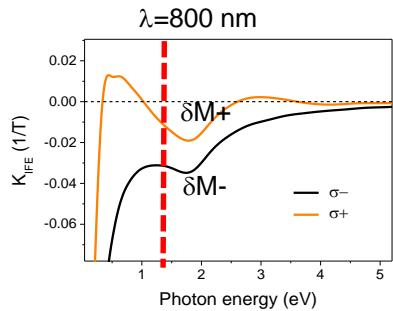
Beamwaist 15 μm

FePt (AgCu) granular recording media



# FePt optical writing a storage media

Starting with magnetization 100%  $M_s$ , saturated case:



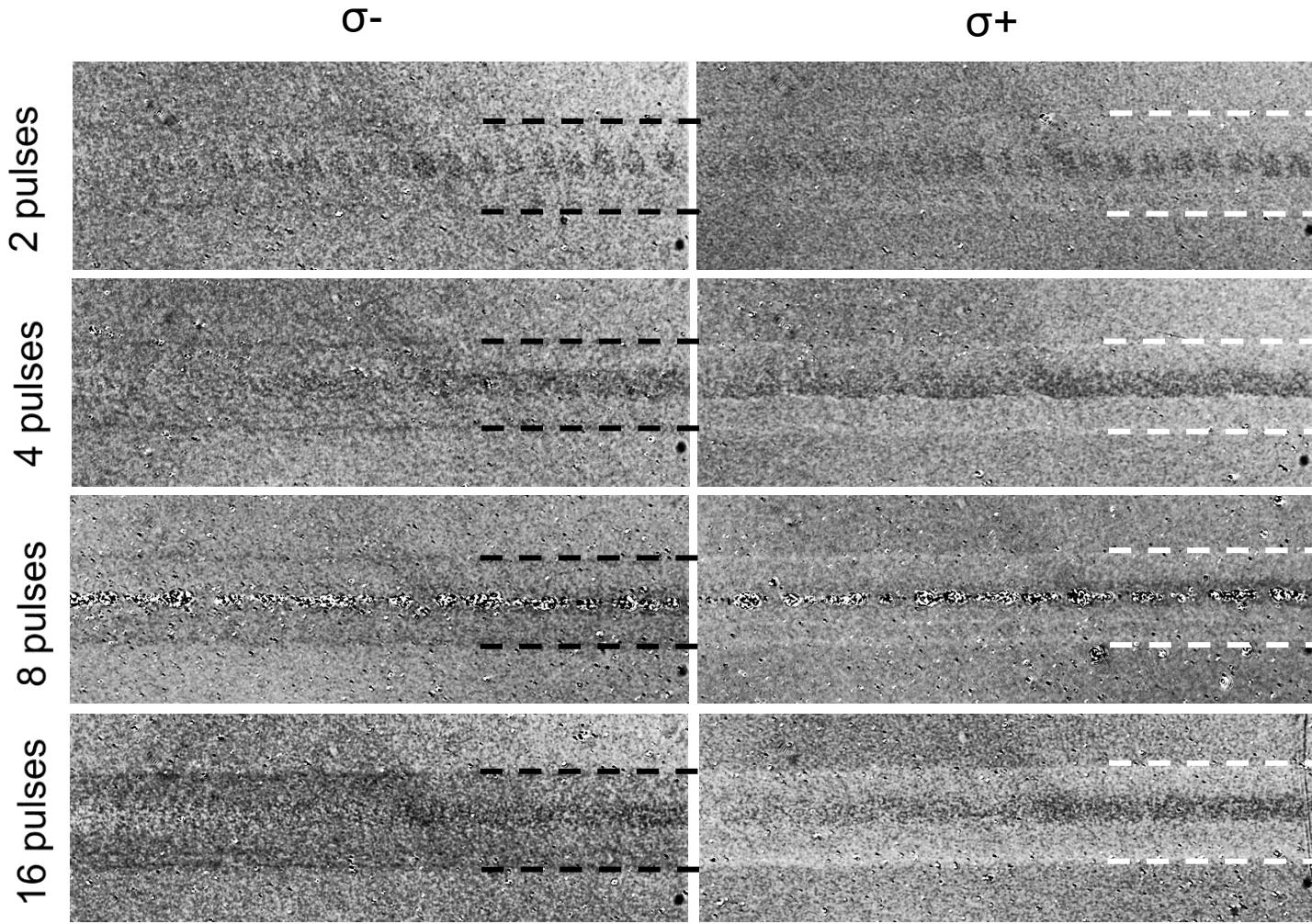
No full writing! What is the mechanism?

15 mW (13.2 mJ/cm<sup>2</sup> per pulse)

# FePt optical writing a storage media



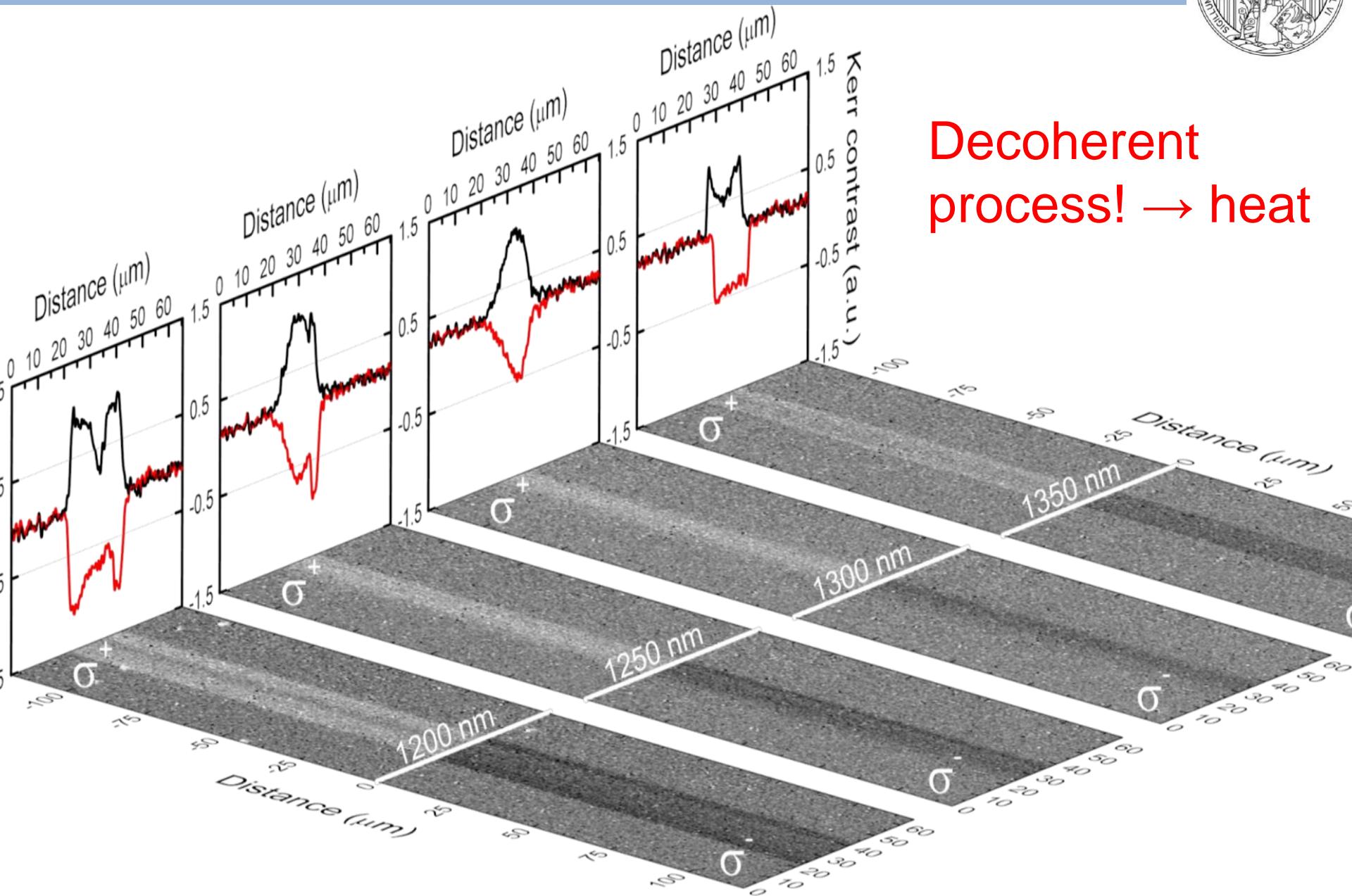
- FePt (AgCu) granular recording media:  
single/ multiple pulse writing



5 mW (30 mJ/cm<sup>2</sup> per pulse)



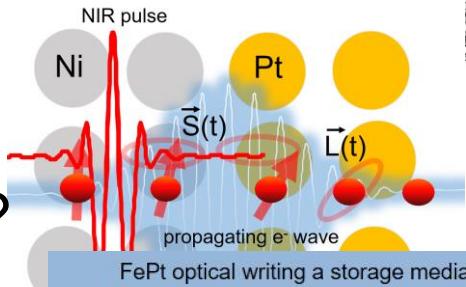
# FePt optical writing a storage media



# Outline



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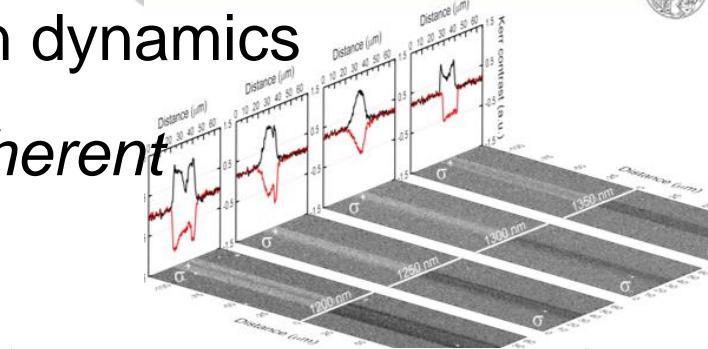
- The nature of femtosecond spin dynamics

- THz spintronic emitter – *noncoherent*

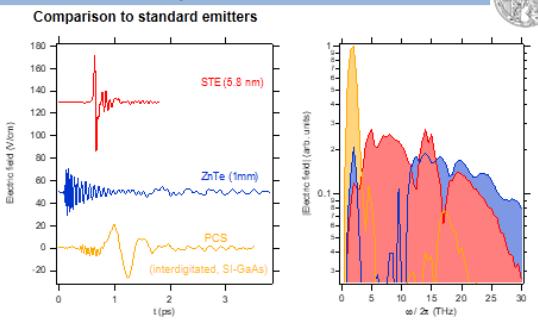
- Topological Insulators

- Lightwave electronics - coherent

- Summary

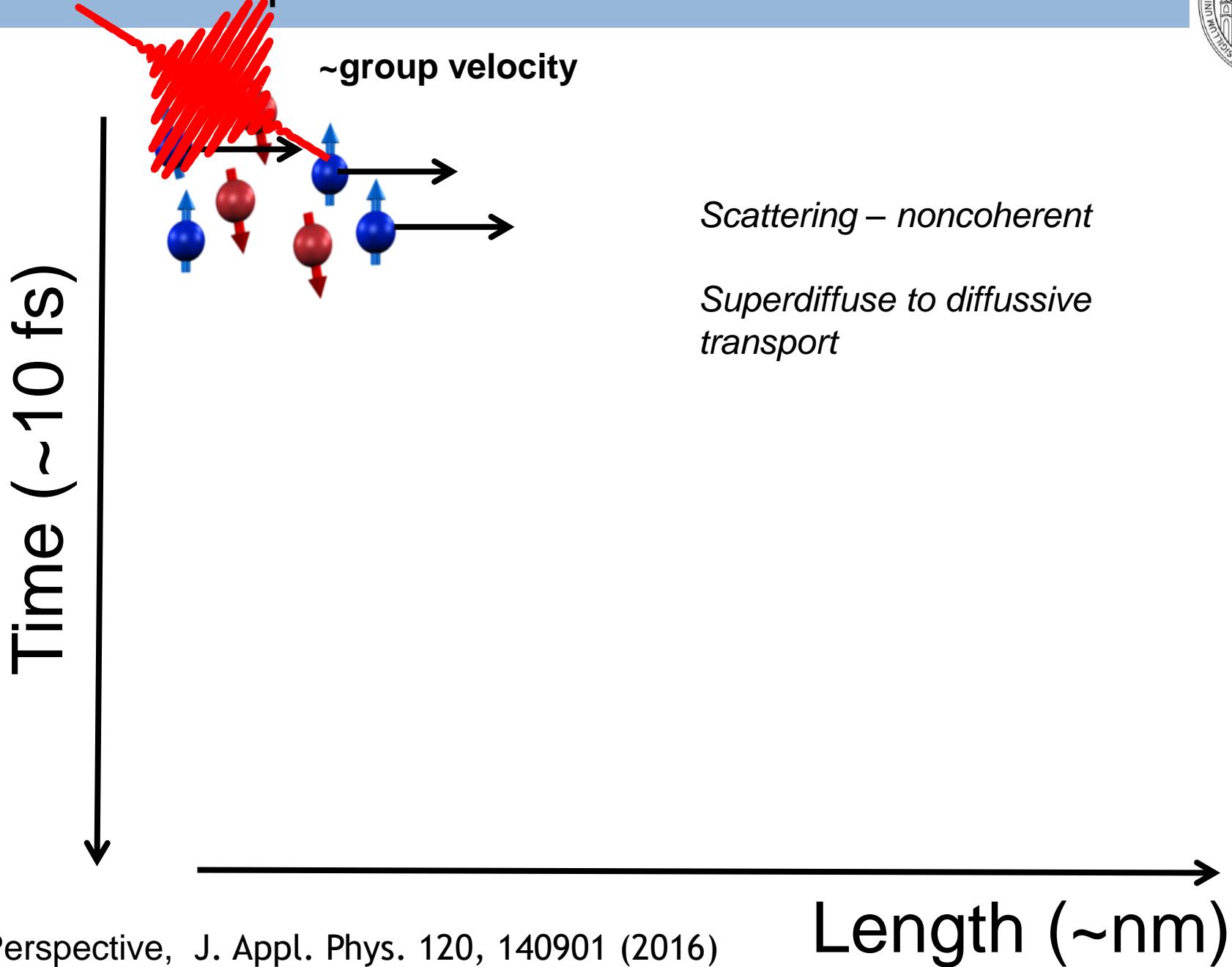


Spintronic THz emitter

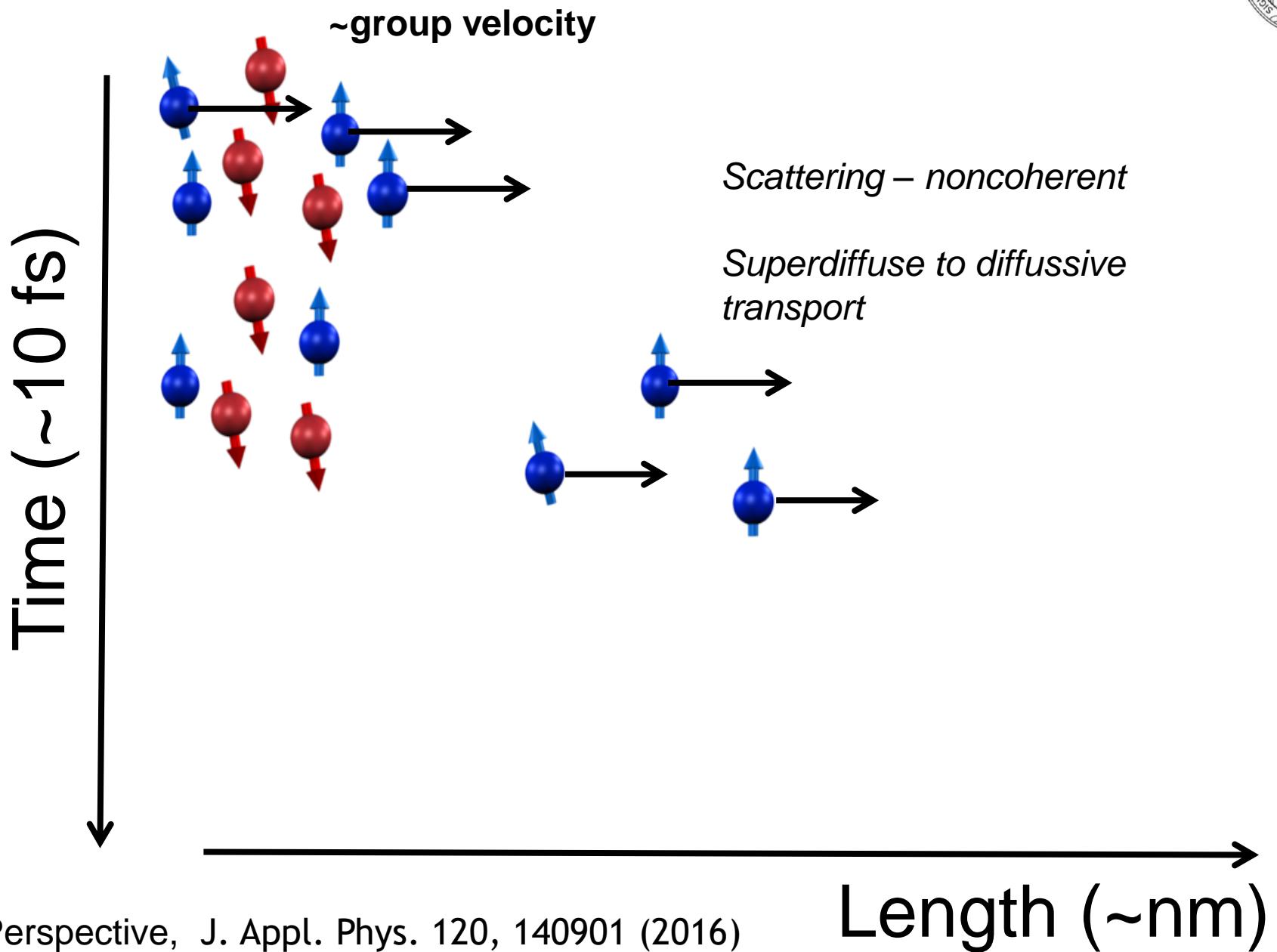


T. Seifert, et al. Nature Photonics (2016)

# Ultrafast: spins

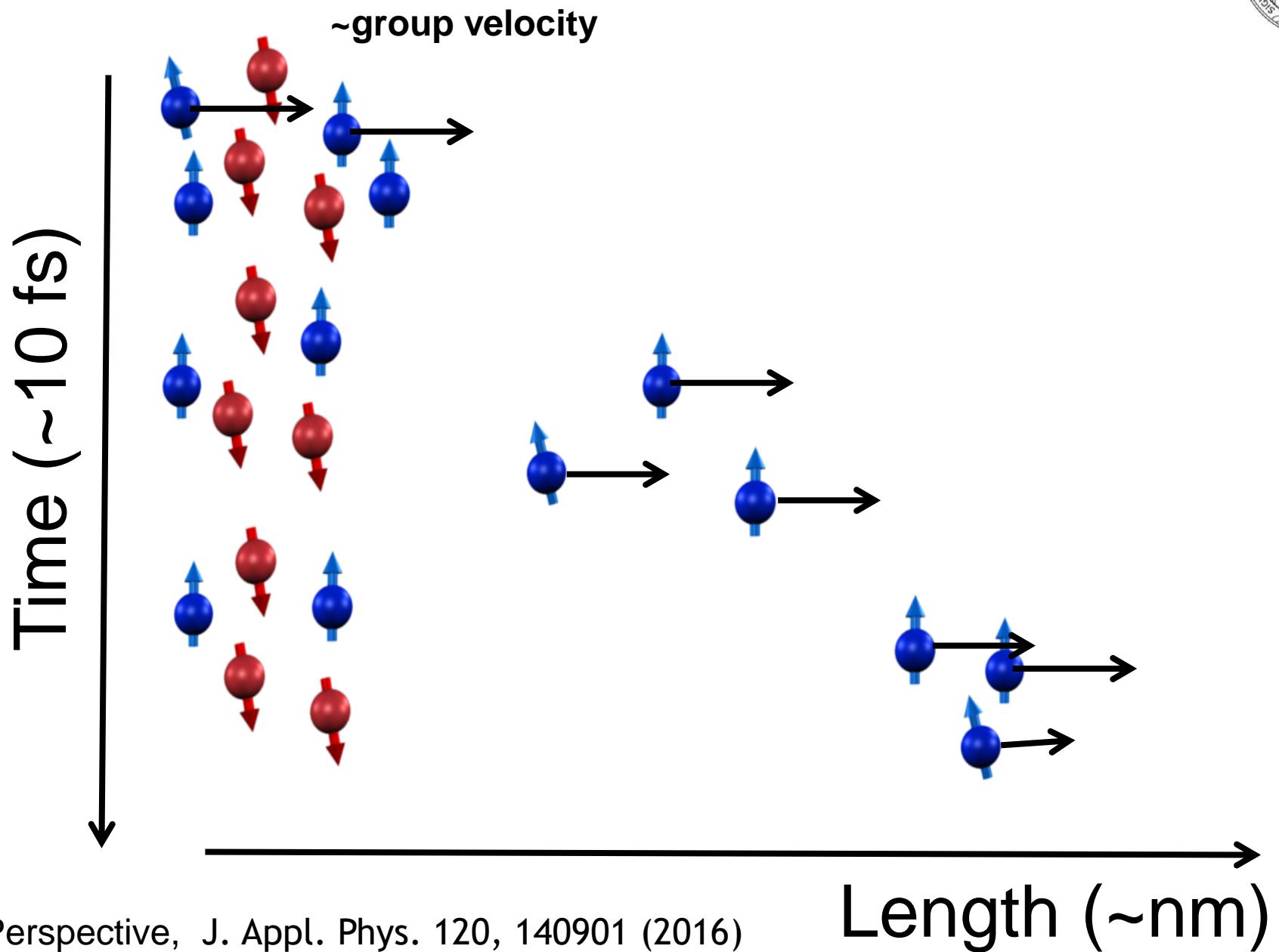


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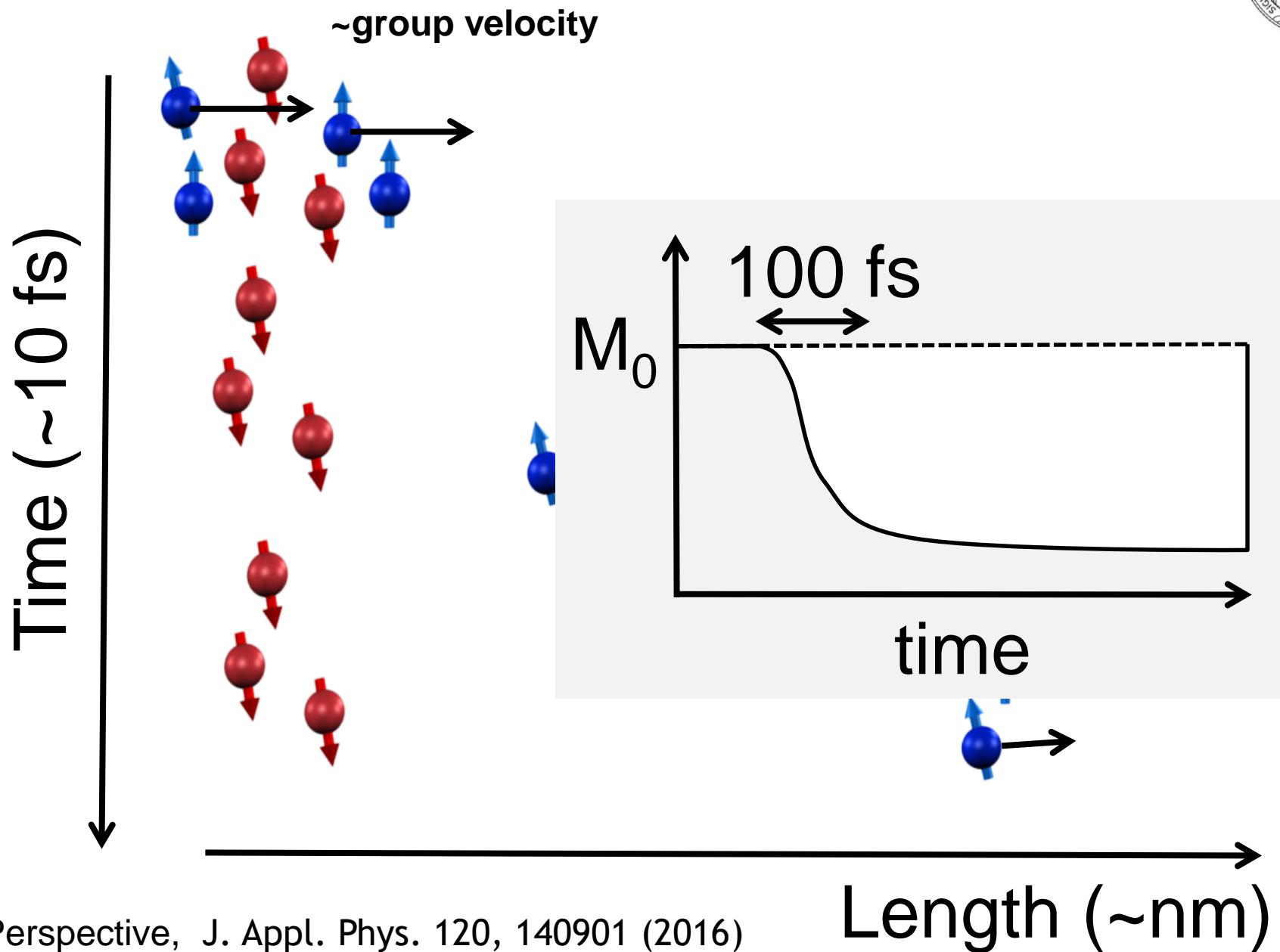




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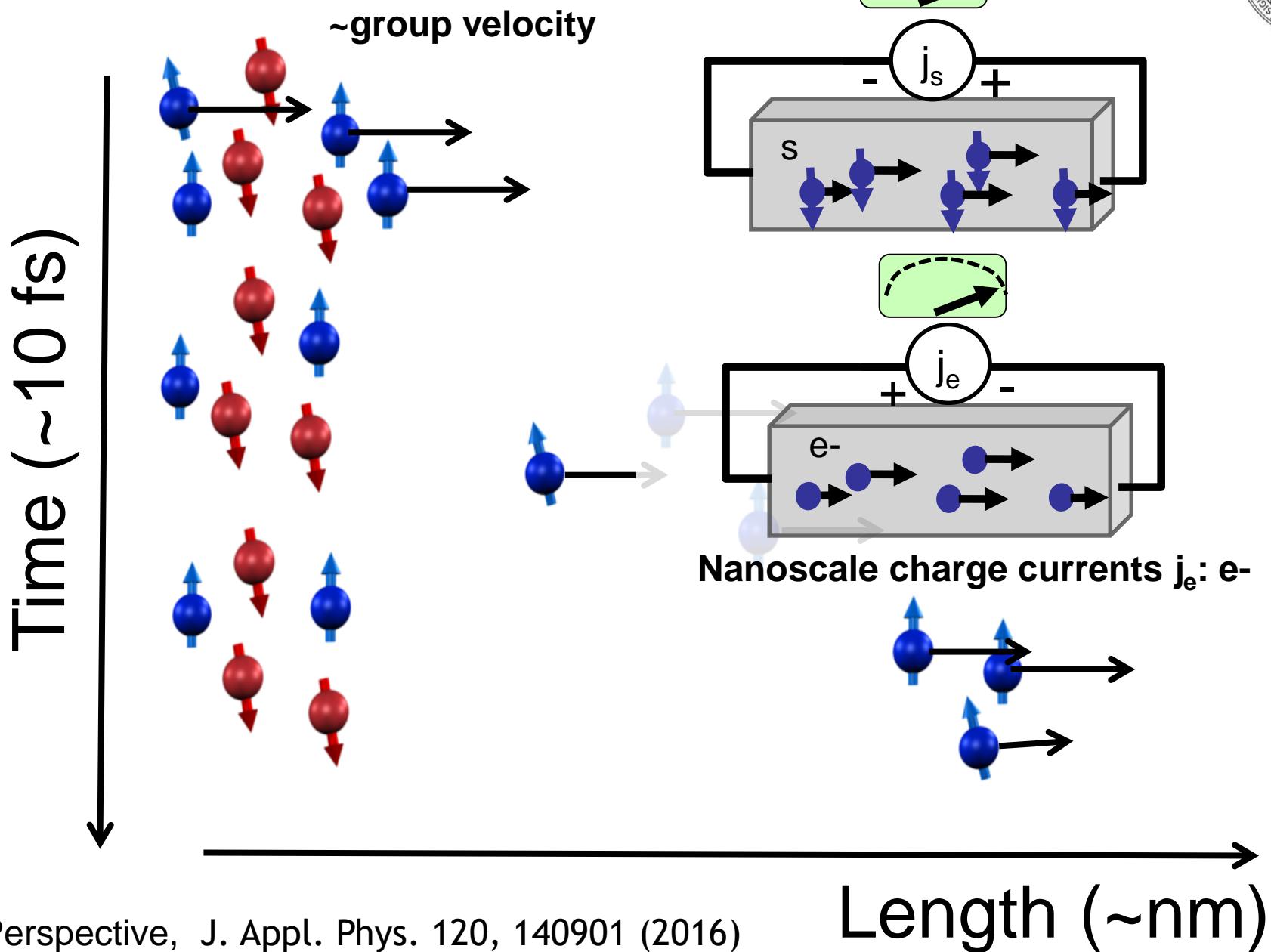


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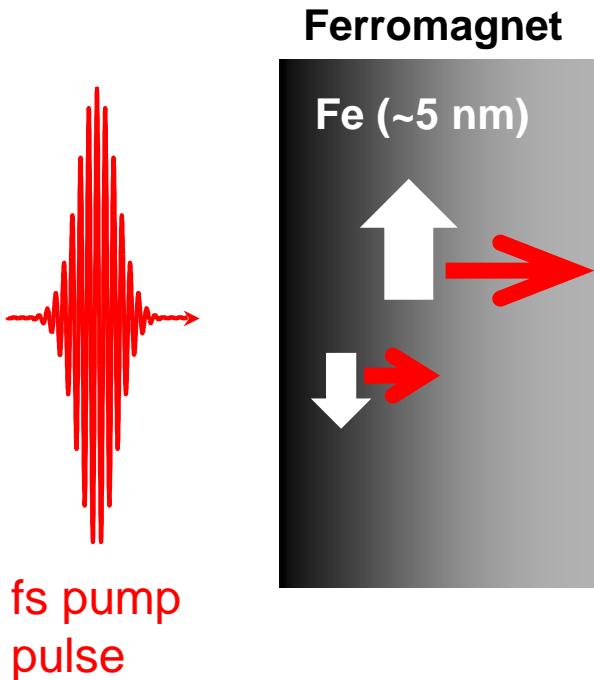
# Ultrafast: spins

## Nanoscale spin currents $j_s: m_s$





# Spintronic THz emitter



**Pump pulse excites  $\uparrow$  and  $\downarrow$  electrons**

$\uparrow$ :  $d \rightarrow sp$  bands  $\Rightarrow$  become fast

$\downarrow$ :  $d \rightarrow d$  bands  $\Rightarrow$  stay slow

Oppeneer *et al.*, PRL (2010)

$\Rightarrow$  Pump launches spin-polarized current

Melnikov *et al.*, PRL (2011)

Rudolf *et al.*, NatComm (2012)

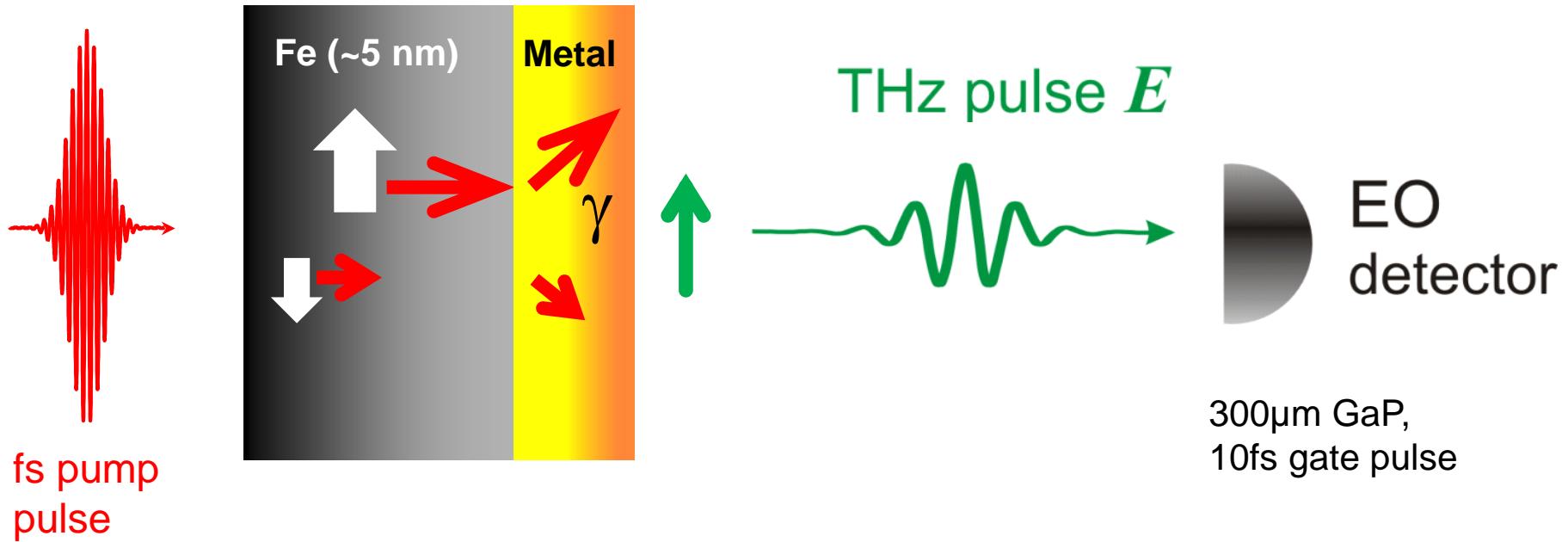
Turgut *et al.*, PRL (2013)

**How to detect the spin current?**

**Idea:** convert spin current into charge current



# Spintronic THz emitter



⇒ Measure THz emission from photoexcited FM/NM bilayers

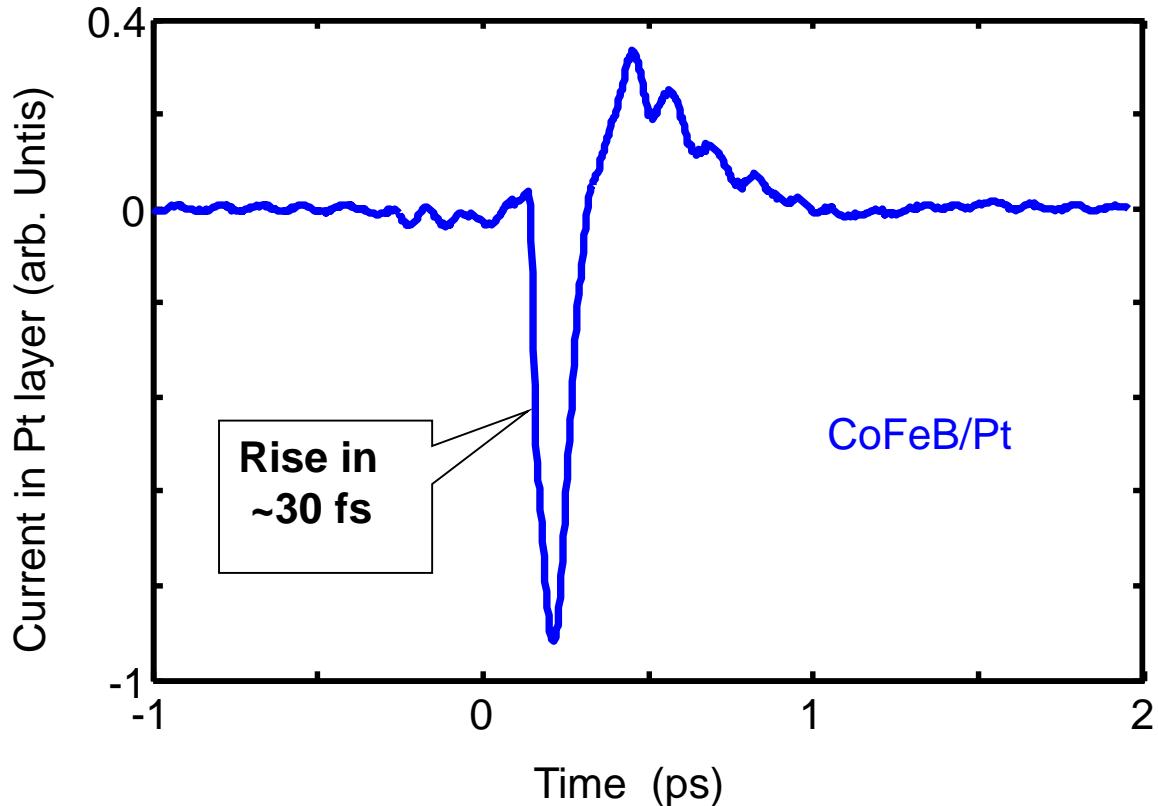
**Note:** just used a pulsed laser oscillator (10 fs, 80 MHz)

Kampfrath, Battiato, Oppeneer, Wolf, Freimuth, Mokrousov, Münzenberg *et al.*, Nature Nanotech. 8, 256 (2013)

# Spintronic THz emitter



## THz Pt sheet current



Reveals spin transport dynamics with 10 fs resolution



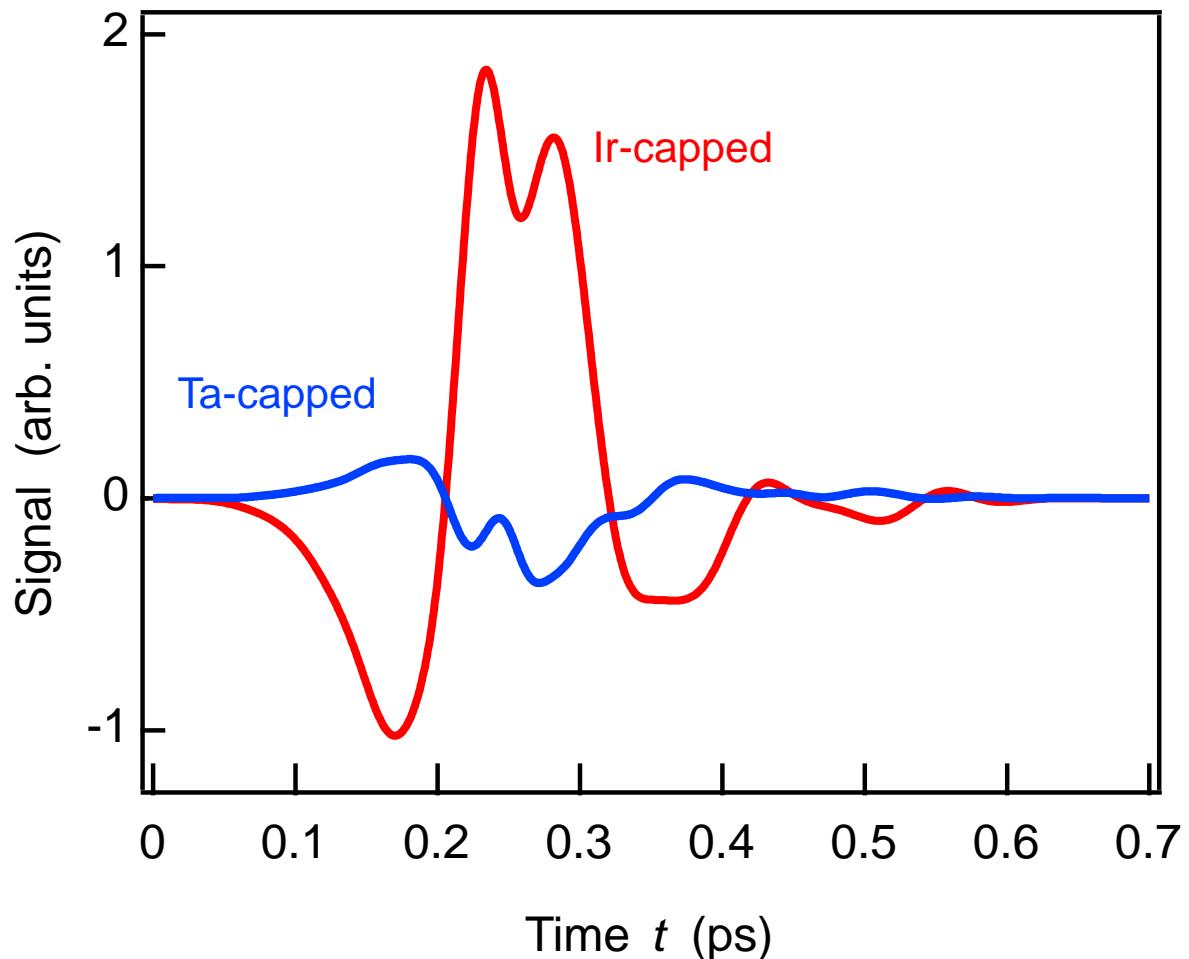
# Spintronic THz emitter

Idea:

Vary nonmagnetic cap layer

Ta vs Ir:

Opposite spin Hall angles, Ir larger

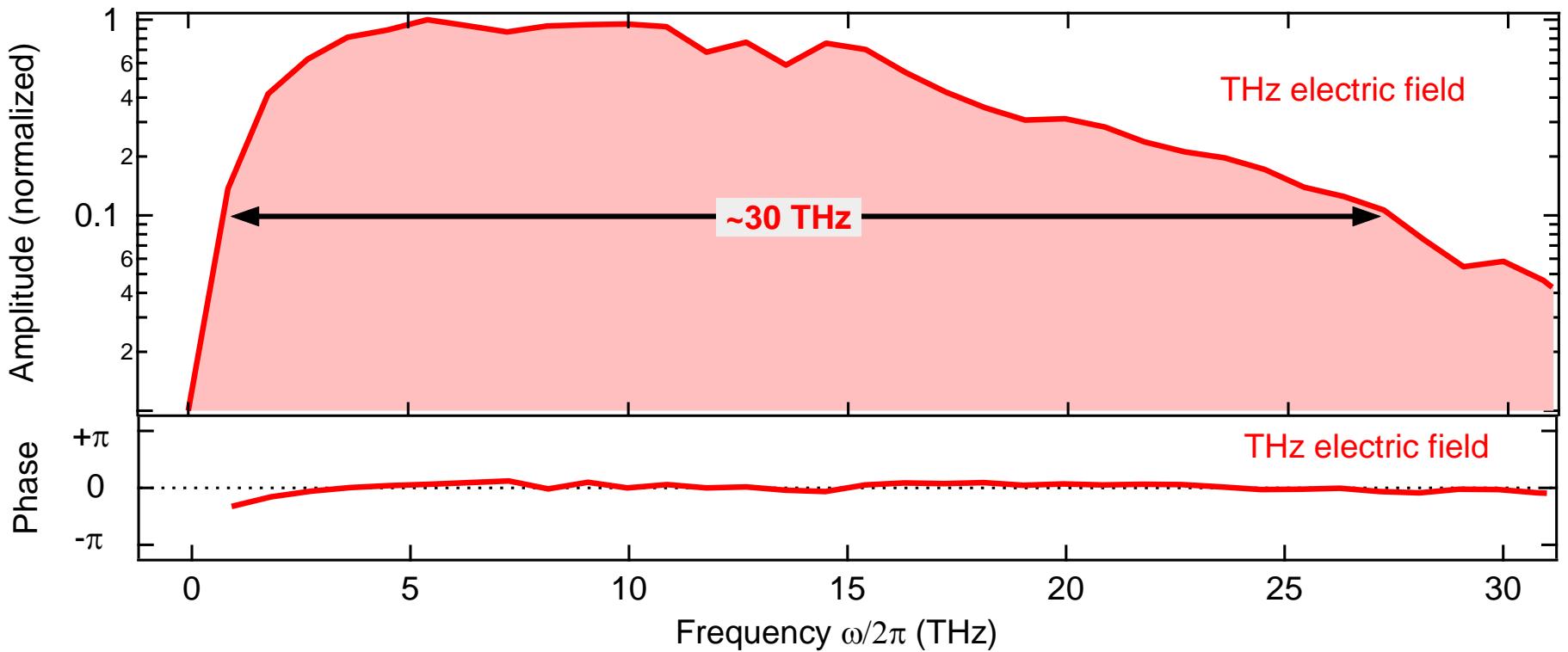


Behavior consistent  
with ISHE scenario?

# Spintronic THz emitter



Fourier transform of time-domain data yields spectrum

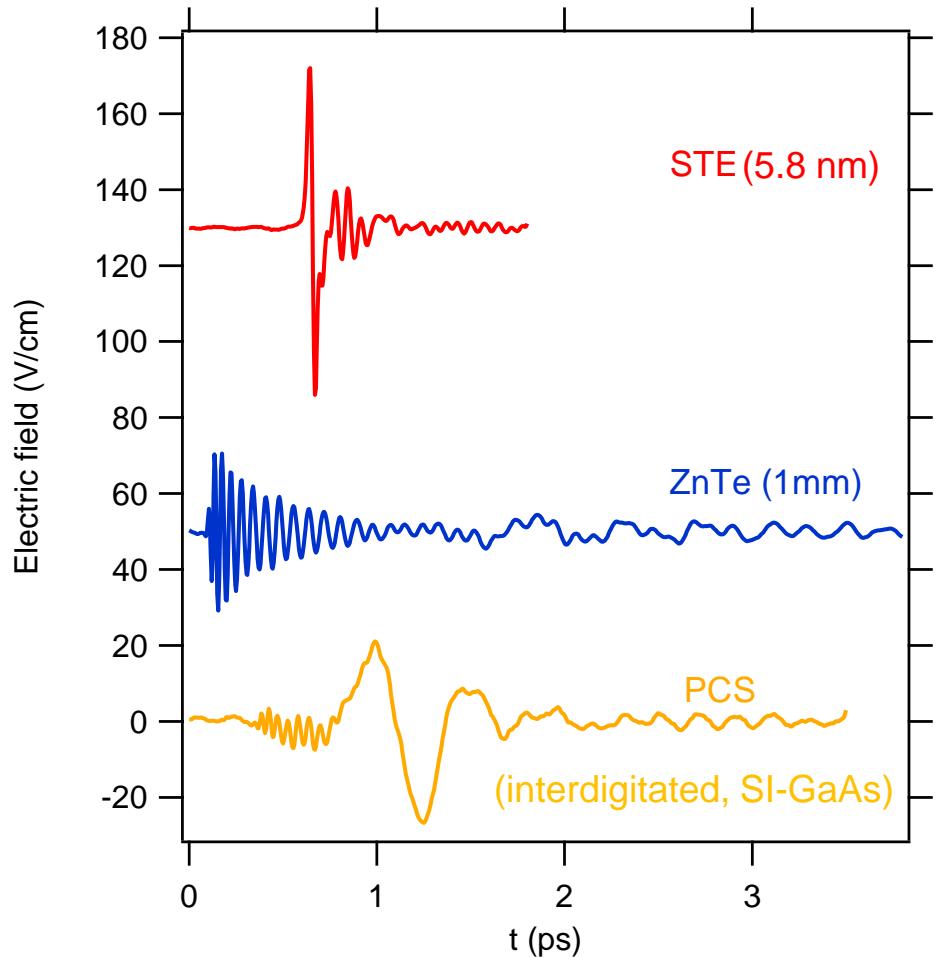


Gap-free emission from 1-30 THz



# Spintronic THz emitter

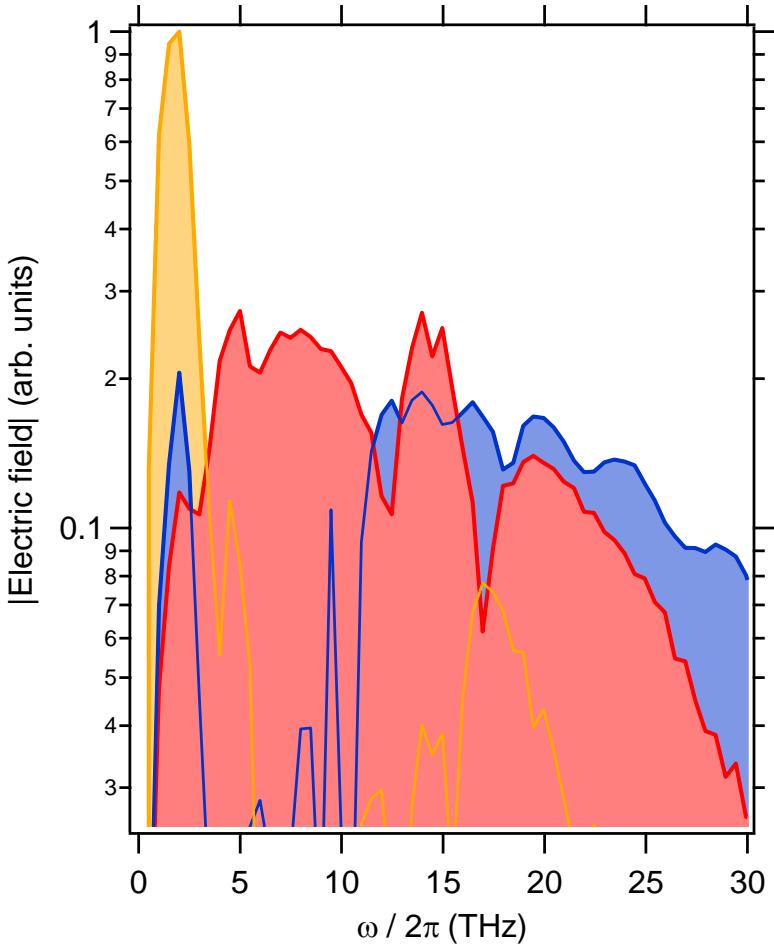
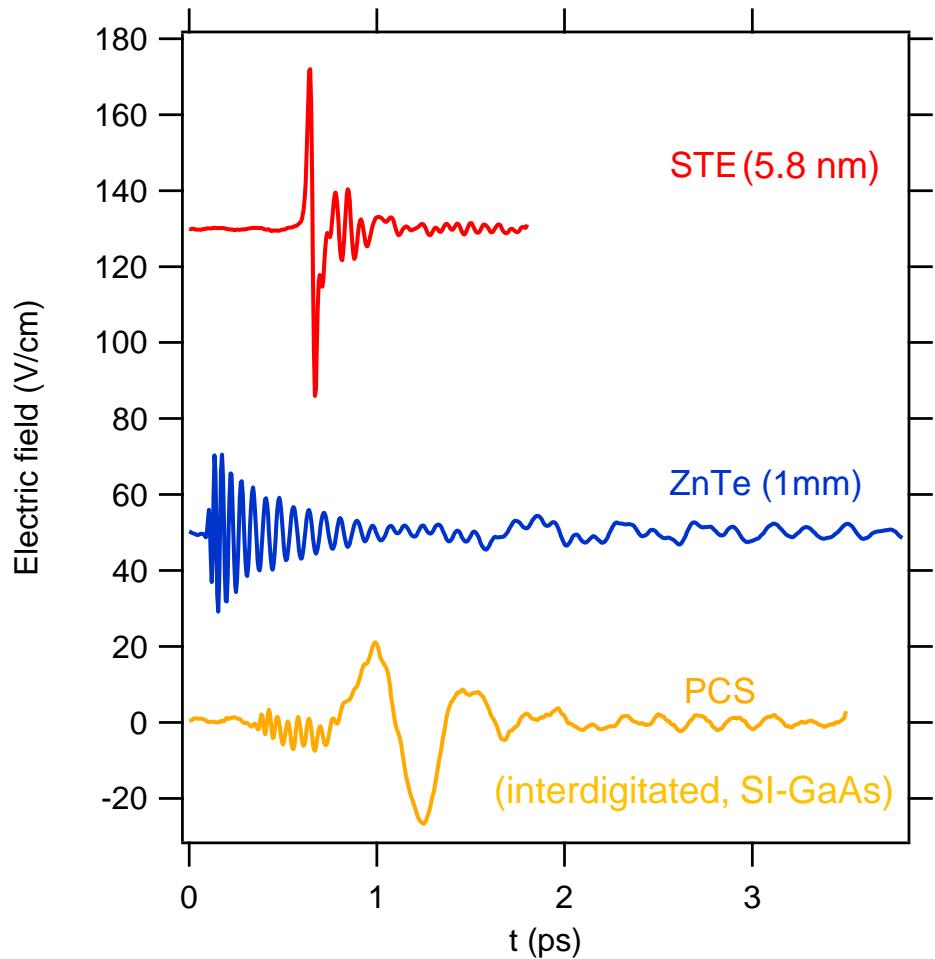
## Comparison to standard emitters





# Spintronic THz emitter

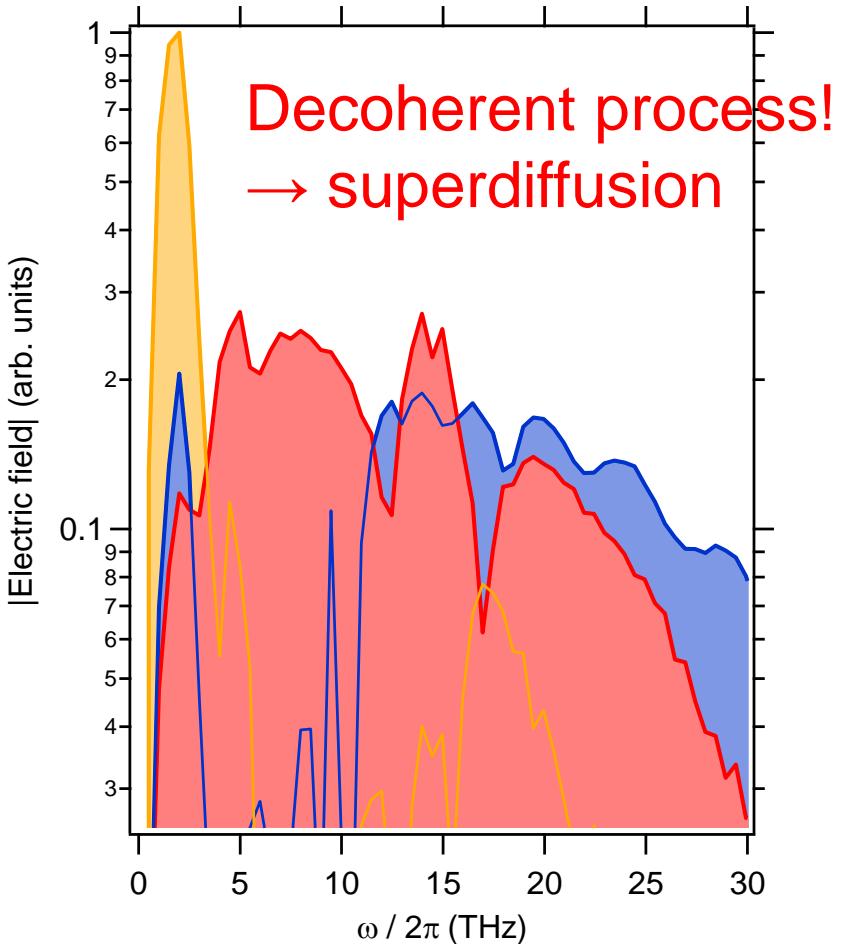
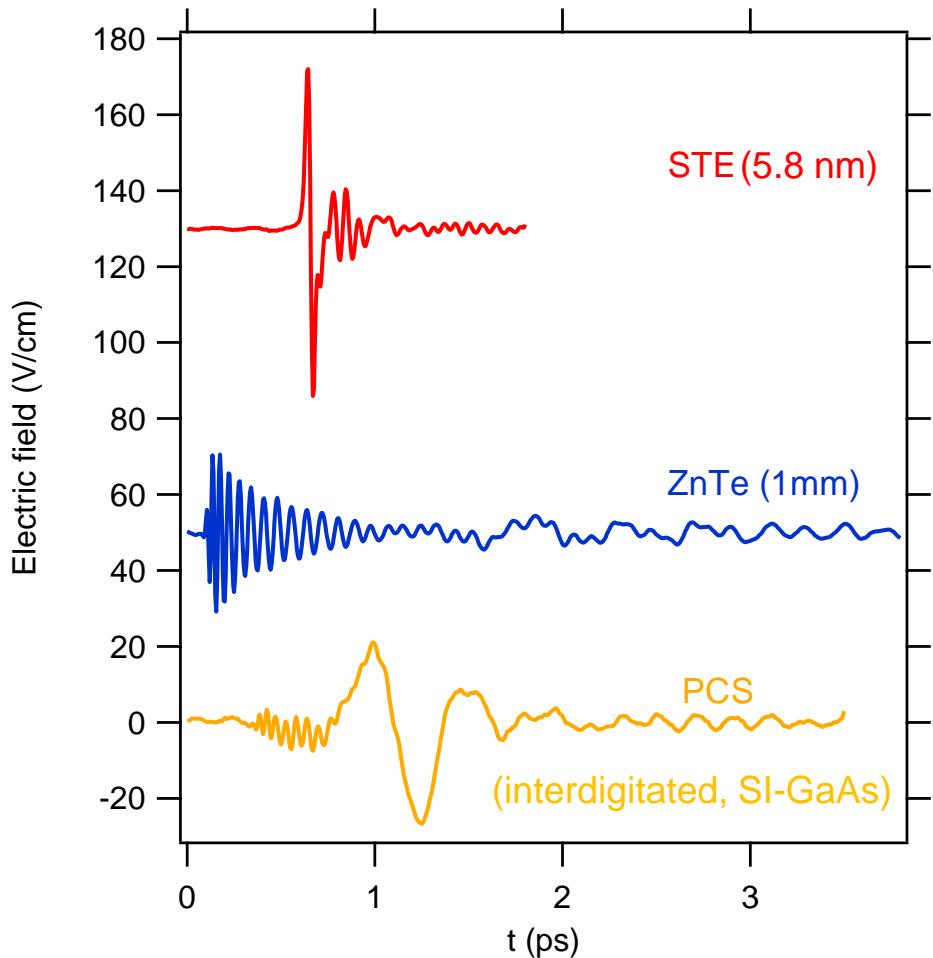
## Comparison to standard emitters





# Spintronic THz emitter

## Comparison to standard emitters



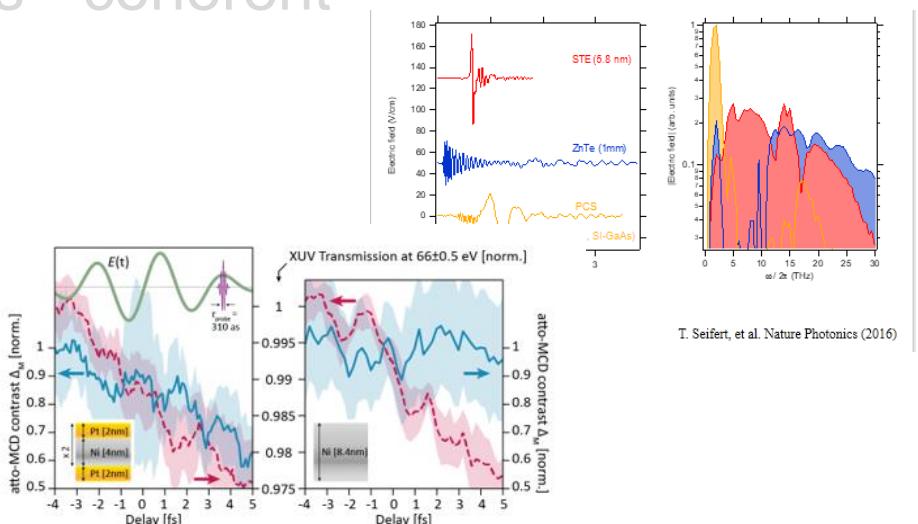
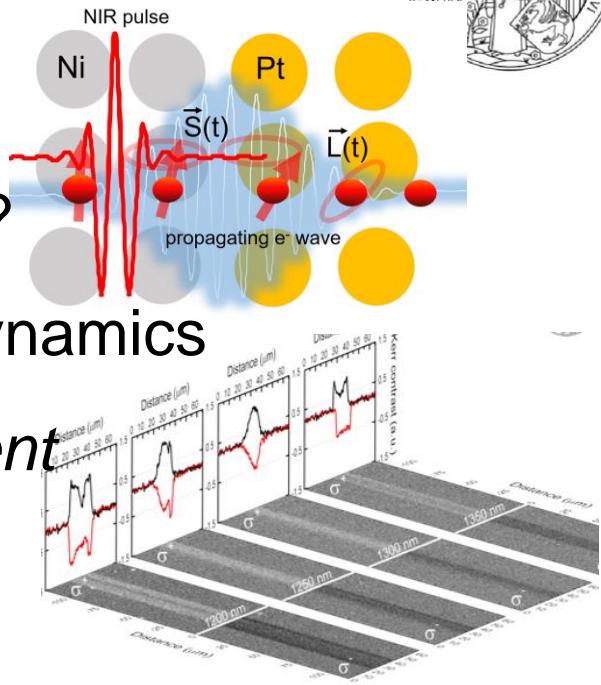
Spintronic metallic emitter outperforms standard emitters over large frequency intervals

Robust, low cost, scalable, easy to handle and flexible

# Outline



- A coherent attosecond spintronics?
- The nature of femtosecond spin dynamics
- THz spintronic emitter - *noncoherent*
- Topological Insulators
- Lightwave electronics - coherent
- Summary



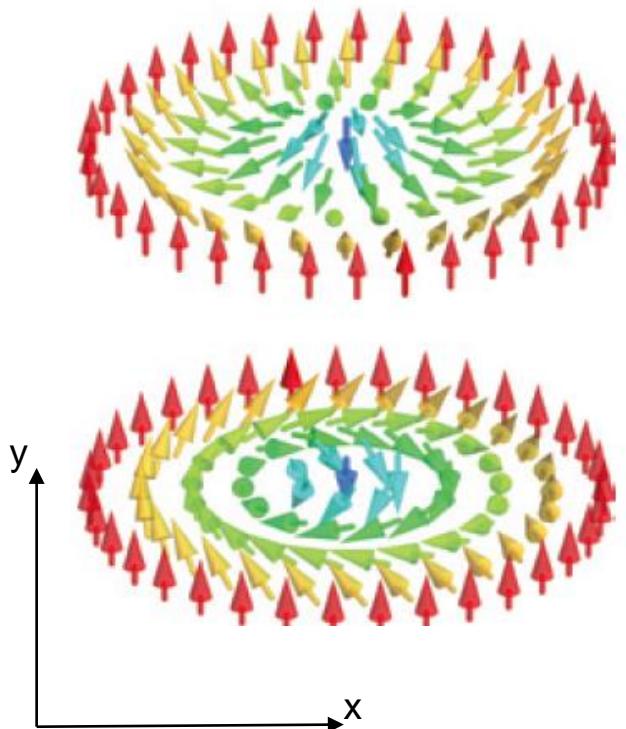
T. Seifert, et al. Nature Photonics (2016)

# Topological matter



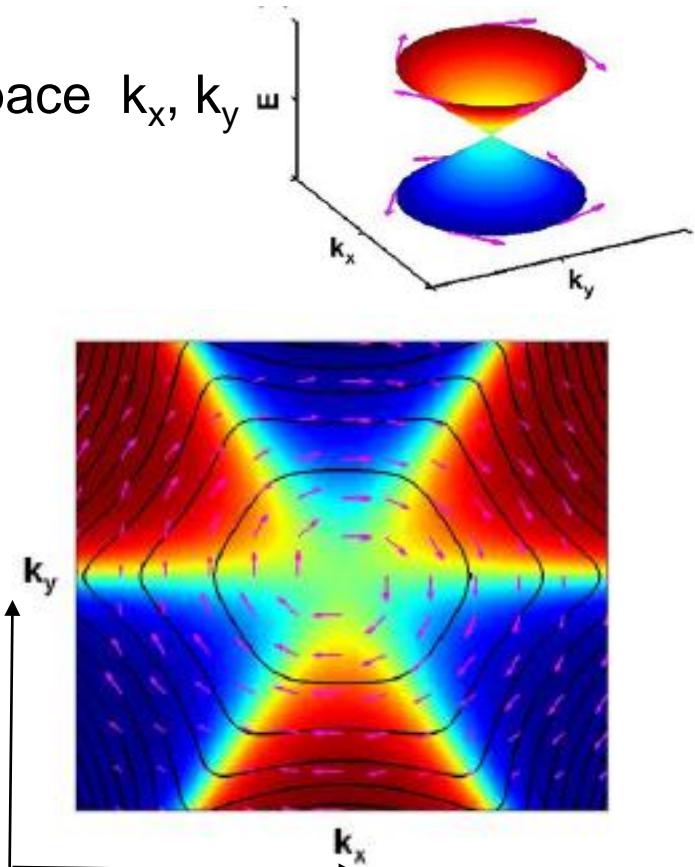
## Skyrmions

Real space  $x, y$



## Topological Insulators

Reciprocal space  $k_x, k_y$



From A. Fert, V. Cros, and J. Sampaio, Nat. Nano. 8, 152–156 (2013)

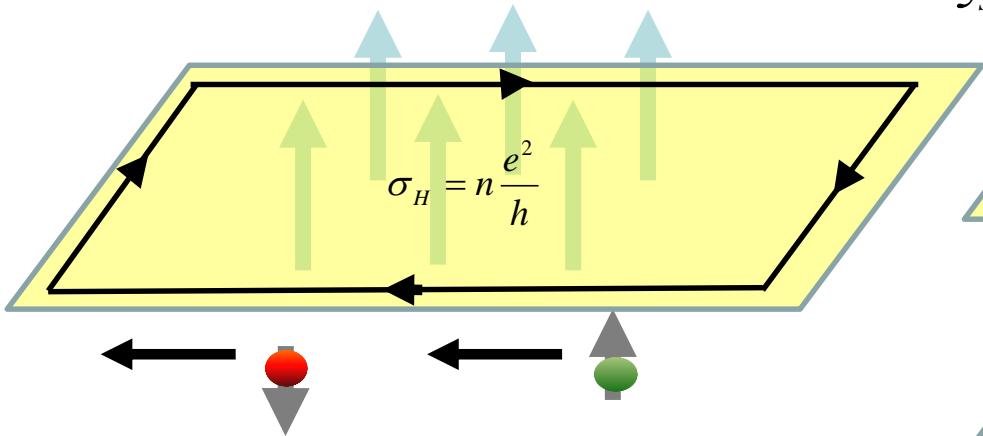
Nature Phys. 5 (2009) 438-42  
Phys. Rev B 82 (2010) 045122

# Topological matter



Large B field

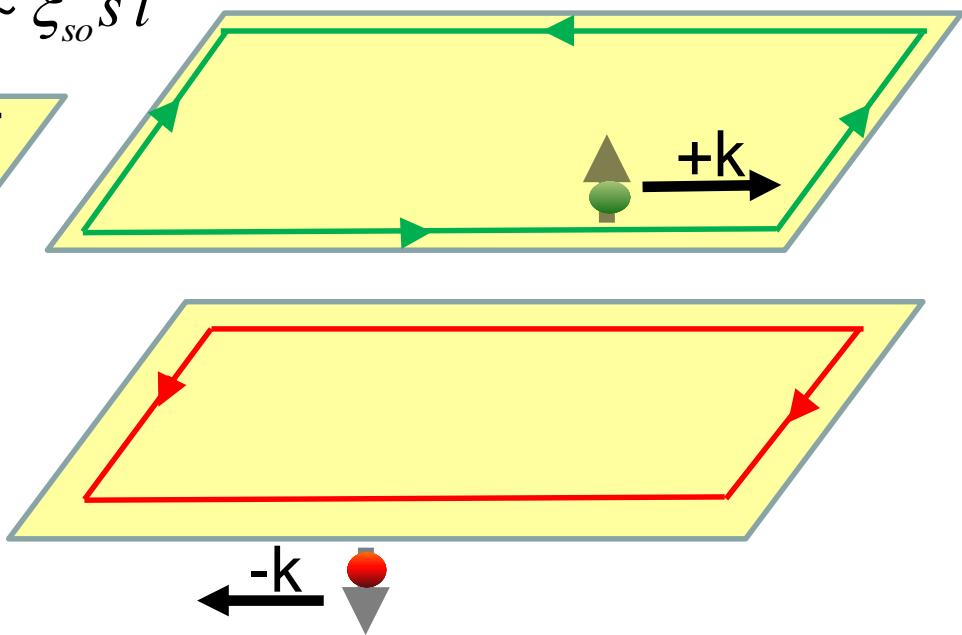
$$E \sim \vec{s} \cdot \vec{B}$$



Cyclotron resonance, in 2D materials quantum Hall effect and edge states

Internal spin-orbit field

$$E \sim \xi_{so} \vec{s} \cdot \vec{l}$$



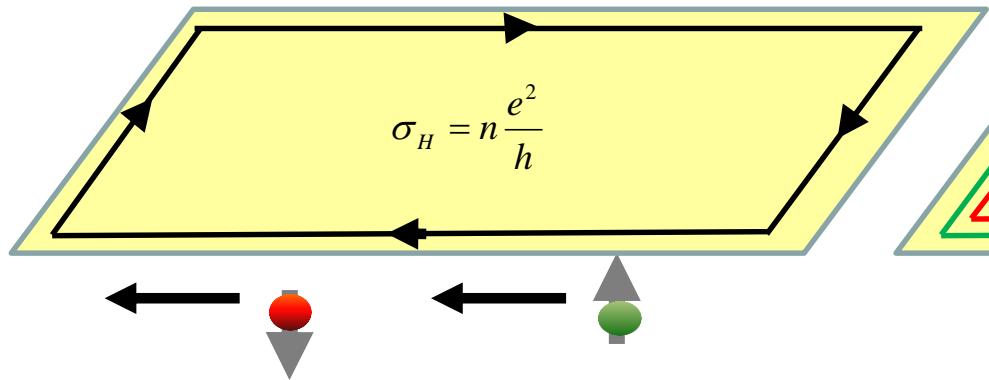
Large spin-orbit coupling leads to cyclotron orbit

# Topological matter



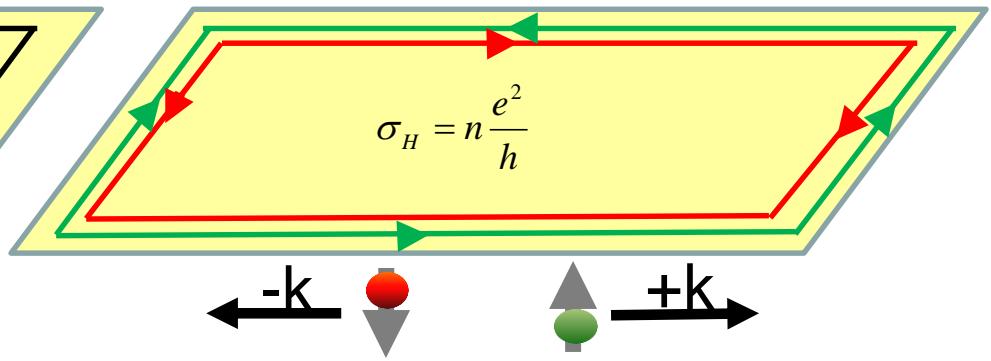
Large B field

$$E \sim \vec{s} \cdot \vec{B}$$



Internal spin-orbit field

$$E \sim \xi_{so} \vec{s} \cdot \vec{l}$$



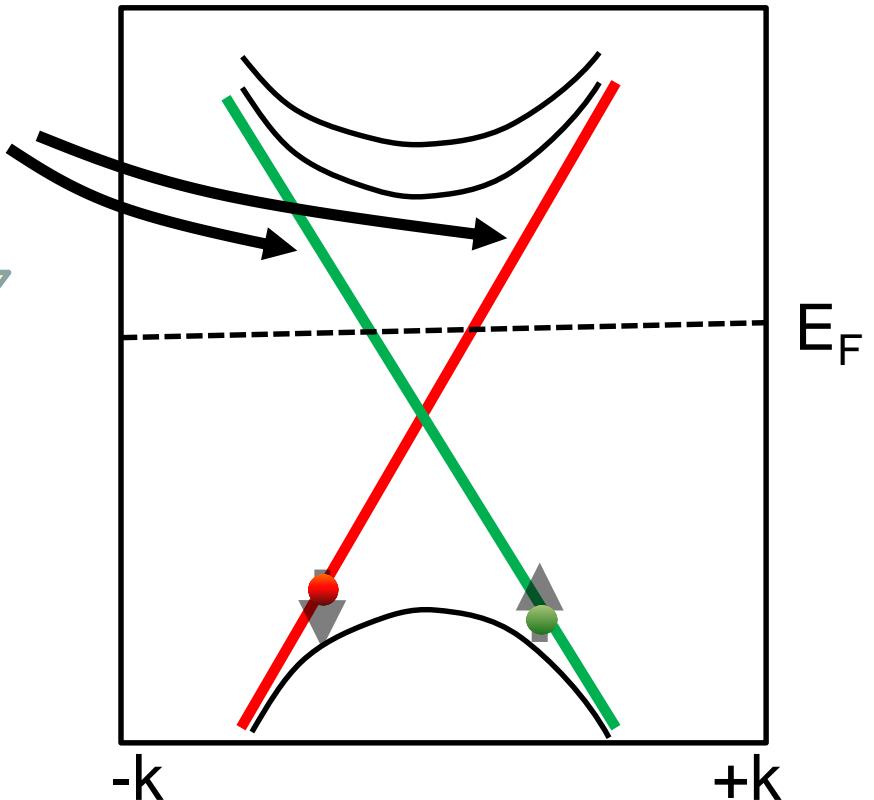
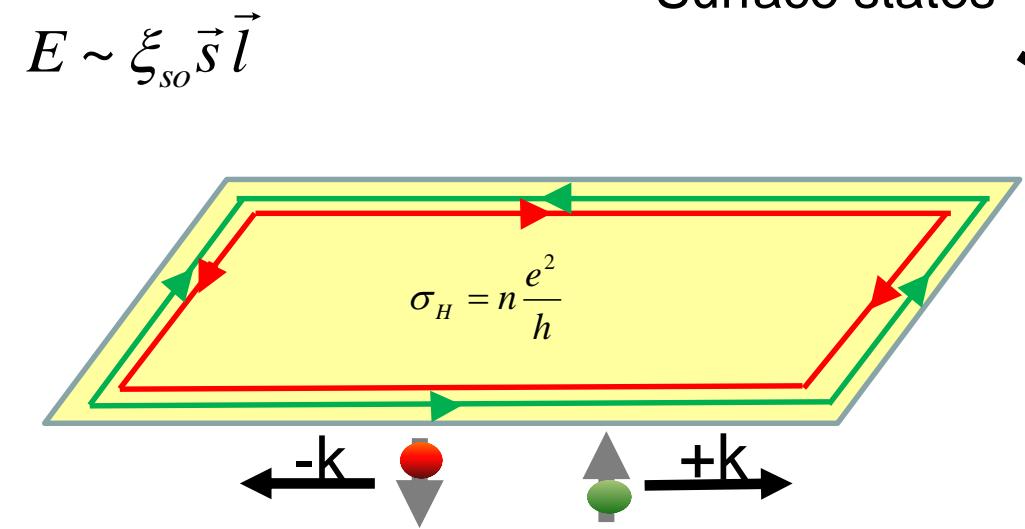
Quantum Hall effect and edge states

Spin-momentum locking, spin polarized edge states

# Topological matter

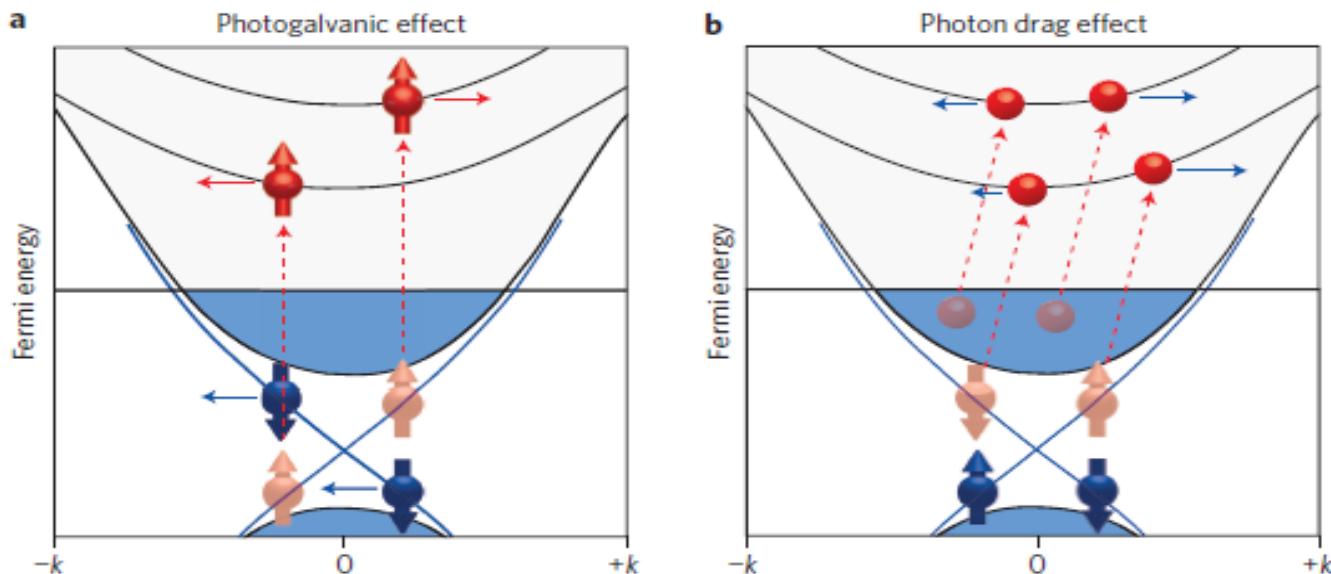
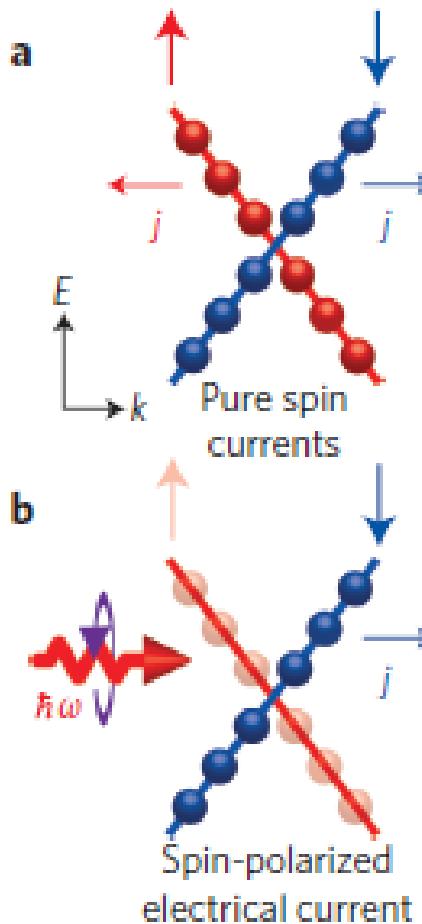
Topological insulator

Schematic bands



- Spin-momentum locking of the surface state
- Generation of spin currents driven by light

# Photoinduced currents in topological insulators



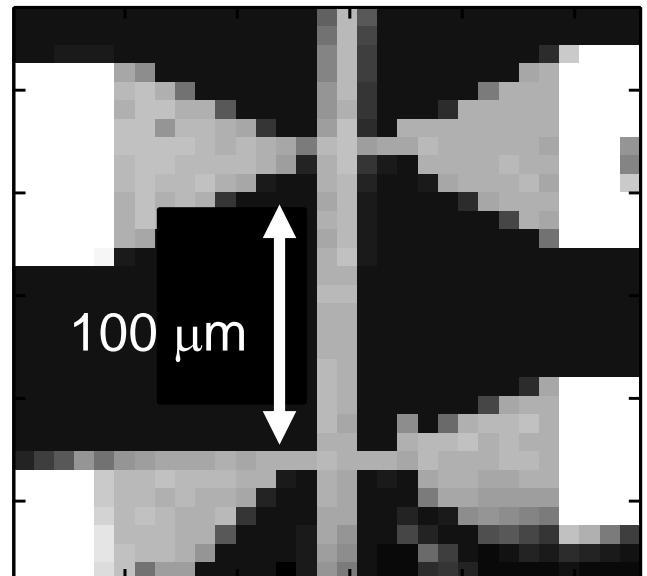
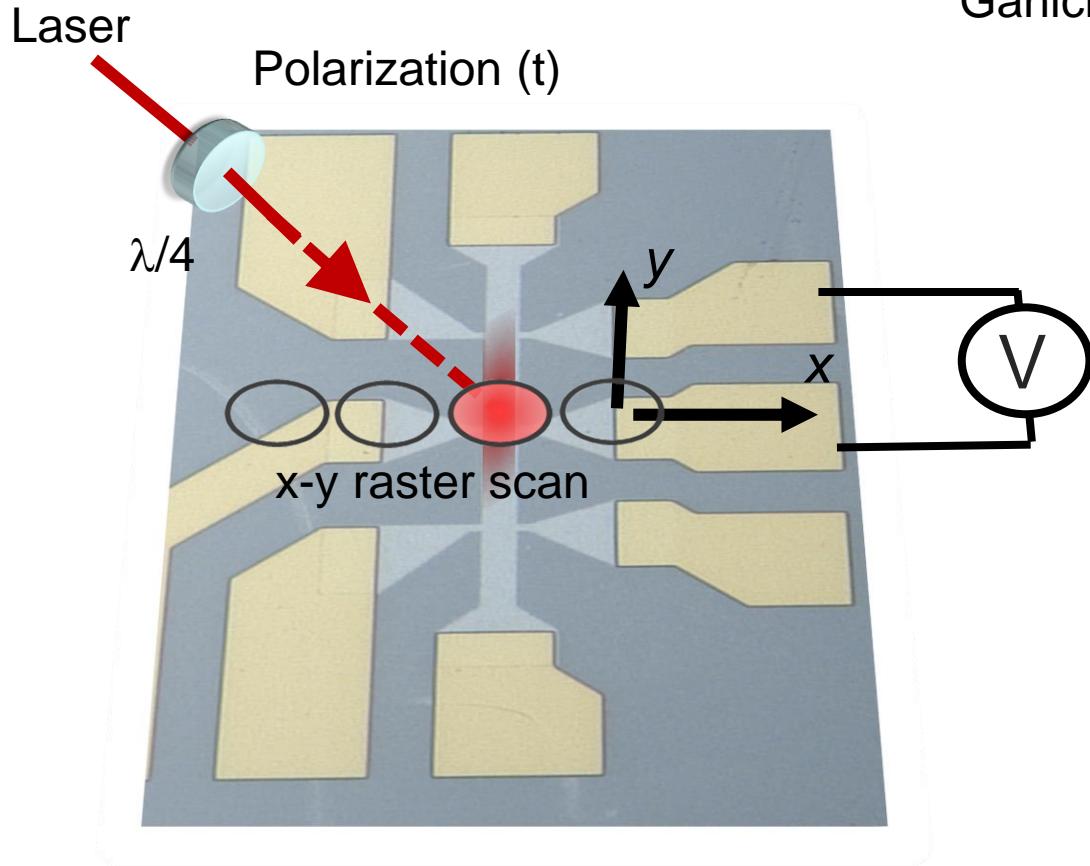
- [1] Control over topological insulator photocurrents with light polarization; J. W. McIver, D. Hsieh, H. Steinberg, P. Jarillo-Herrero and N. Gedik; NATURE NANOTECHNOLOGY LETTERS: DOI: 10.1038/NNANO.2011.214

# Photoinduced currents in topological insulators



- Circular photovoltaic effect

J. W. McIver, et al. Nature Nano.(2011).  
Ganichev et al. Nature (2002).

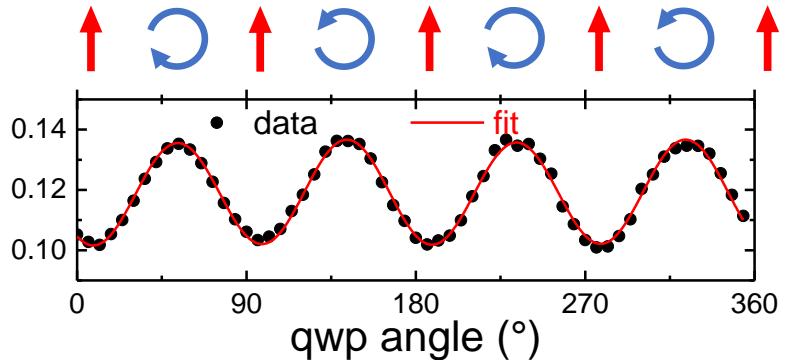
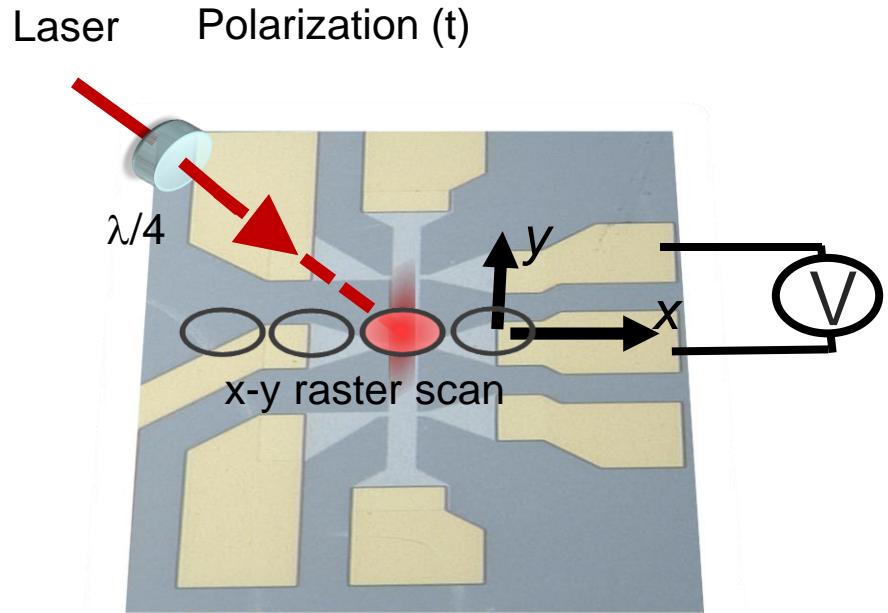


Reflectivity map  
scan area

- 3D topological insulator intrinsic doping, 16 nm  
 $(\text{Bi}_{0.57}\text{Sb}_{0.43})_2\text{Te}_3$

# Topological matter

- Circular photogalvanic effect C, thermovoltage D



Ganichev model ( $\theta$ =pump helicity):

$$U(\theta) = \frac{C \sin(2\theta) + L_1 \sin(4\theta)}{+ L_2 \cos(4\theta) + D}$$

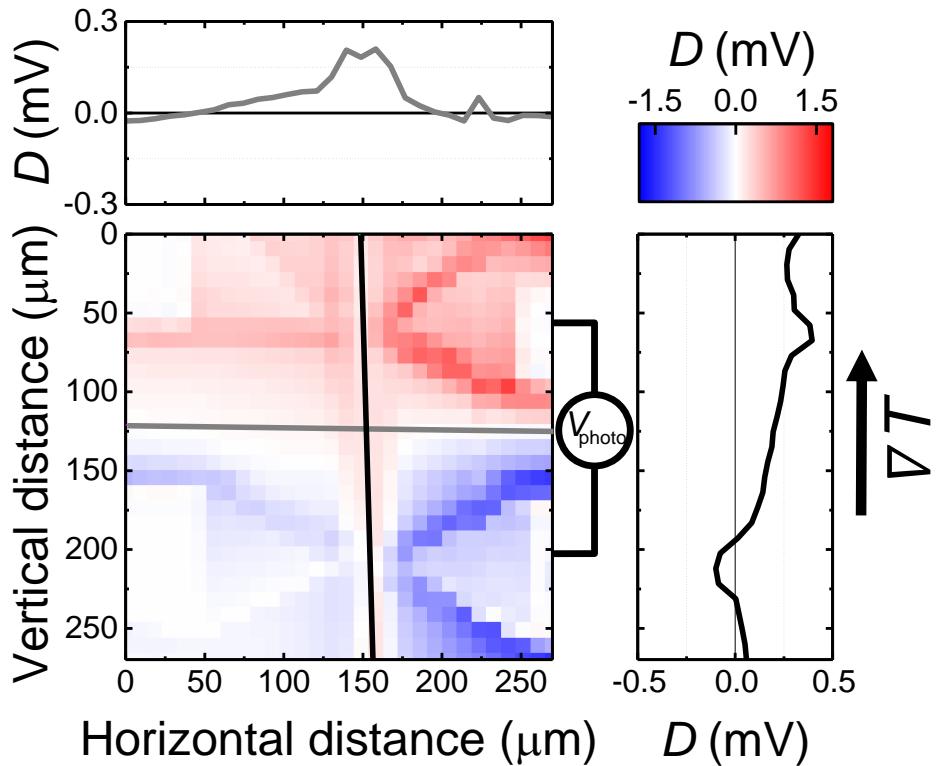
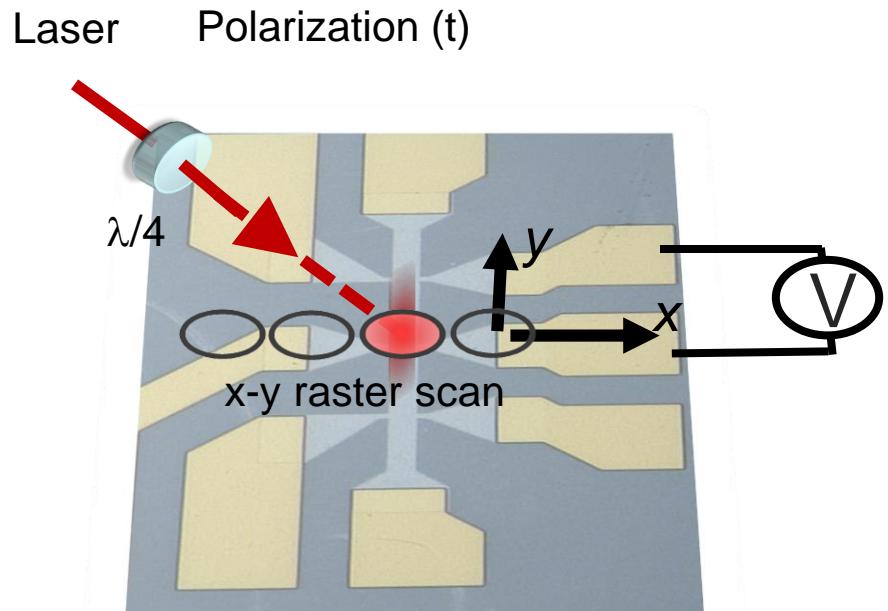
Separation of the different contributions.

Seebeck effects  
(thermal)

Photoinduced currents

# Topological matter

- Circular photogalvanic effect C, thermovoltage D



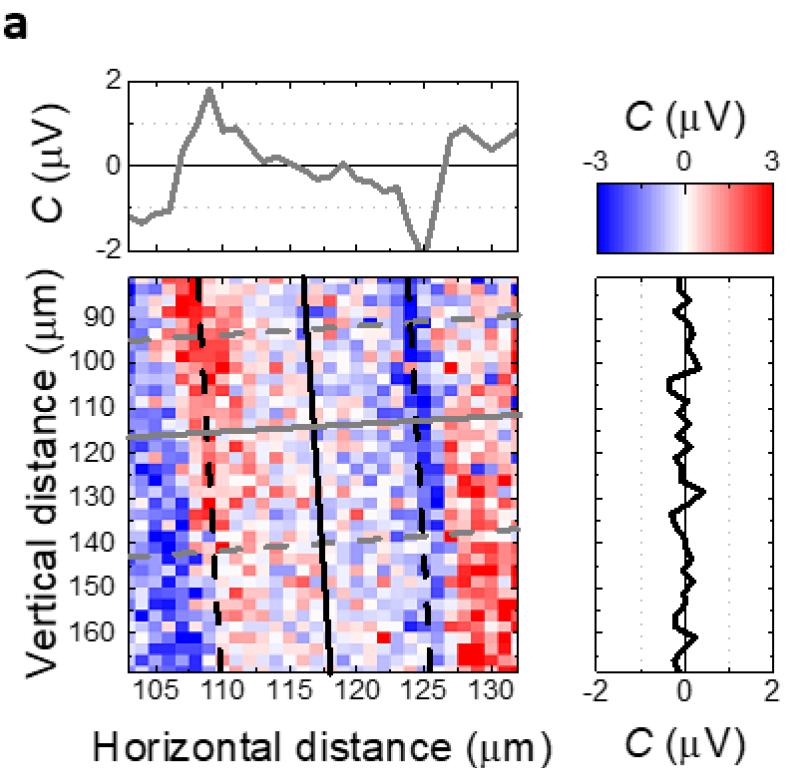
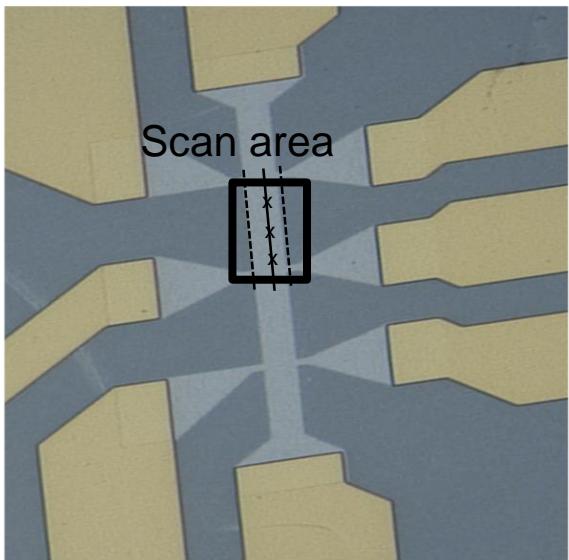
- 3D topological insulator intrinsic doping, 16 nm  
 $(\text{Bi}_{0.57}\text{Sb}_{0.43})_2\text{Te}_3$

# Topological matter



- Circular photovoltaic effect C, thermovoltage D

Detail: Hall bar



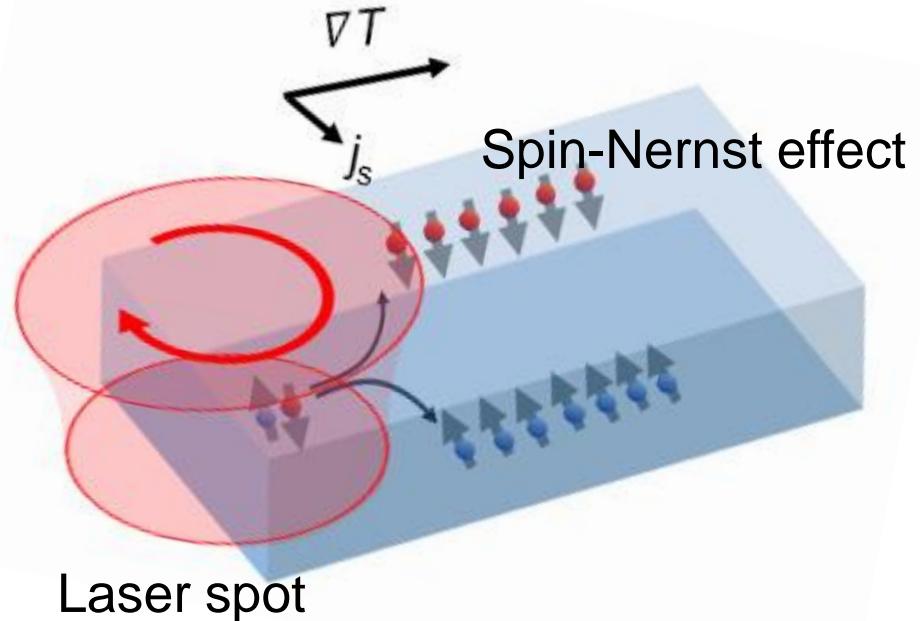
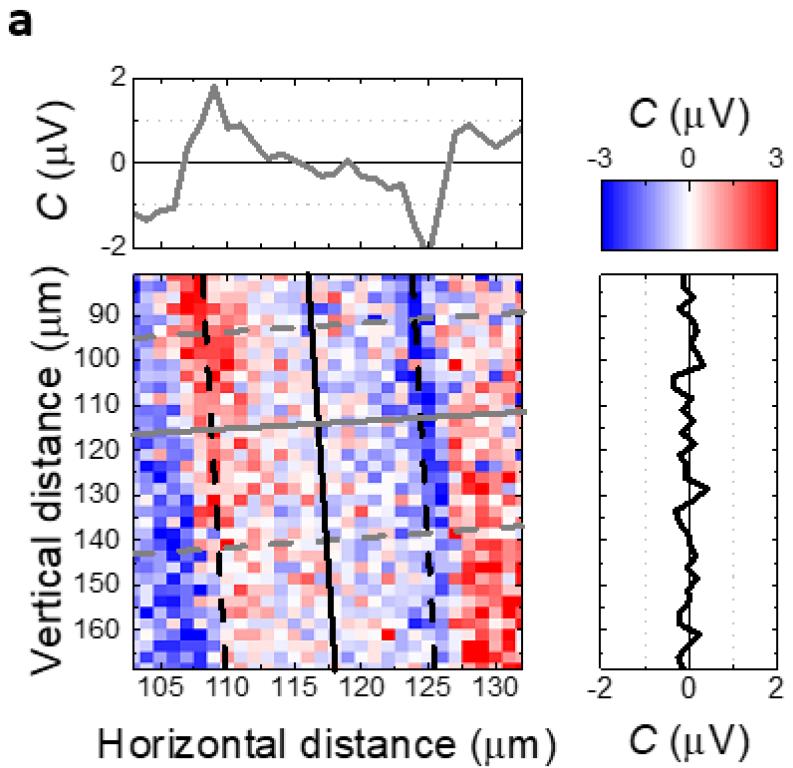
- Spin accumulation by Spin-Nernst effect

# Topological matter



- Circular photogalvanic effect C, thermovoltage D

Detail: Hall bar

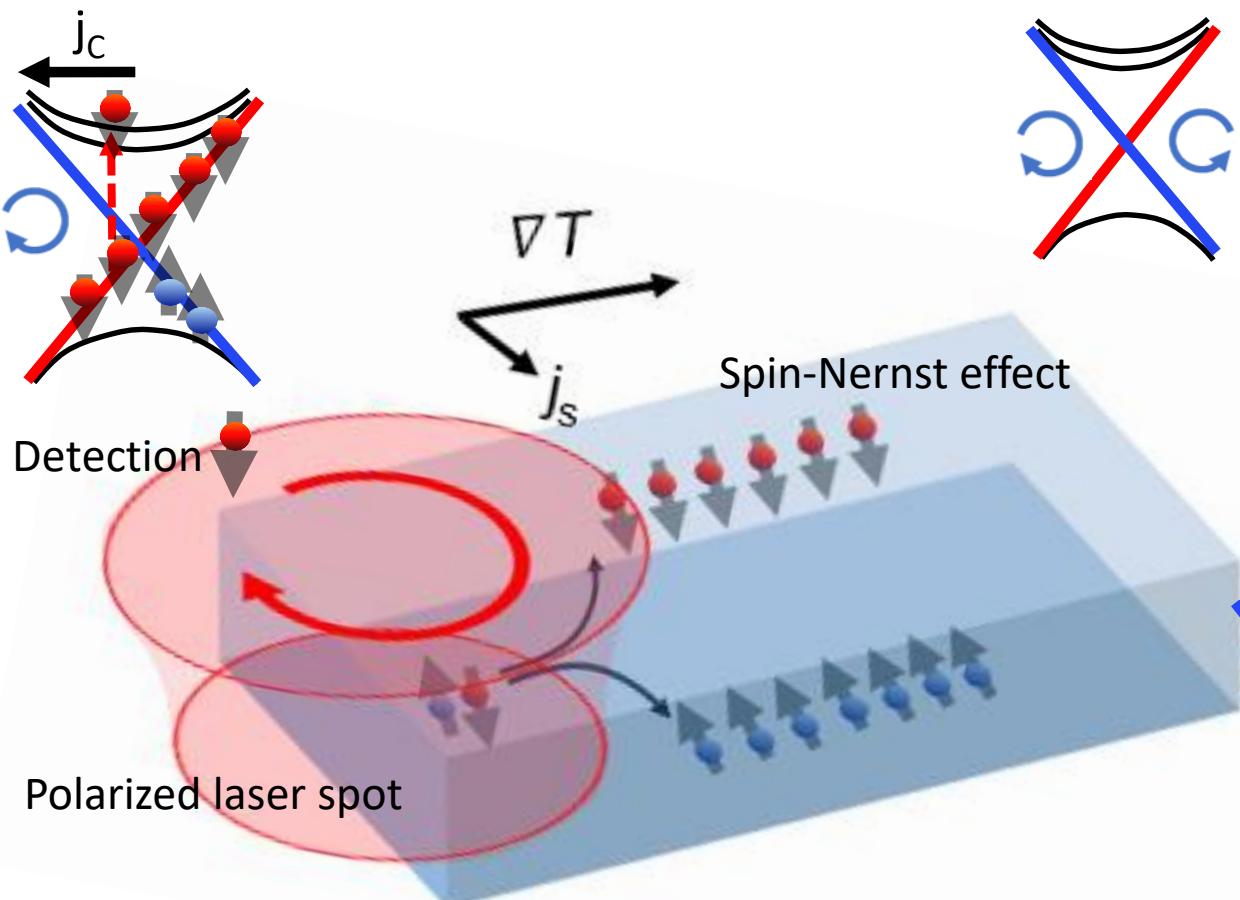


- Spin accumulation by Spin-Nernst effect

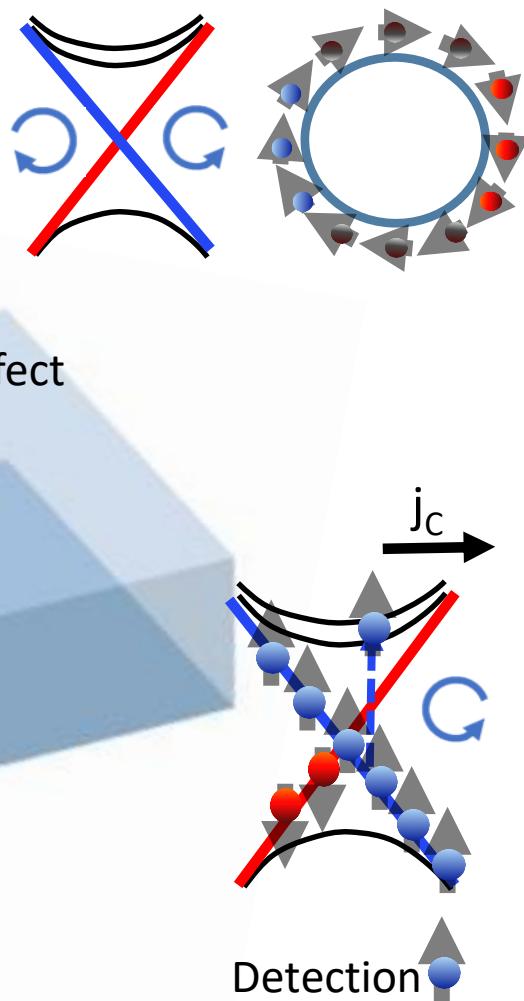
# Topological matter



Spin-orbit driven spin-accumulation



Spin-sensitive detection by photo-current blocking

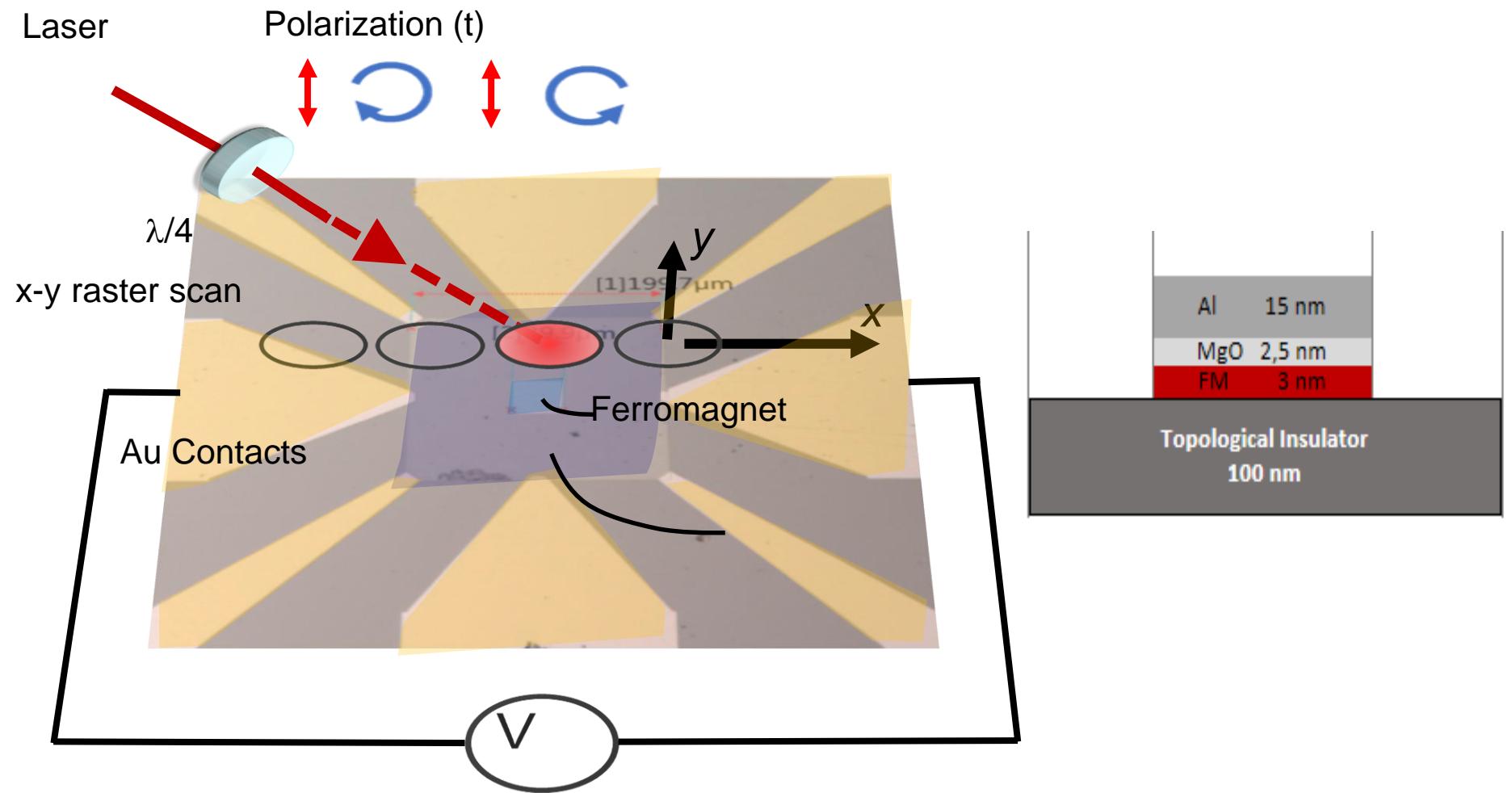


See also S. Meyer et al. Nature Materials (2017).

# Topological matter



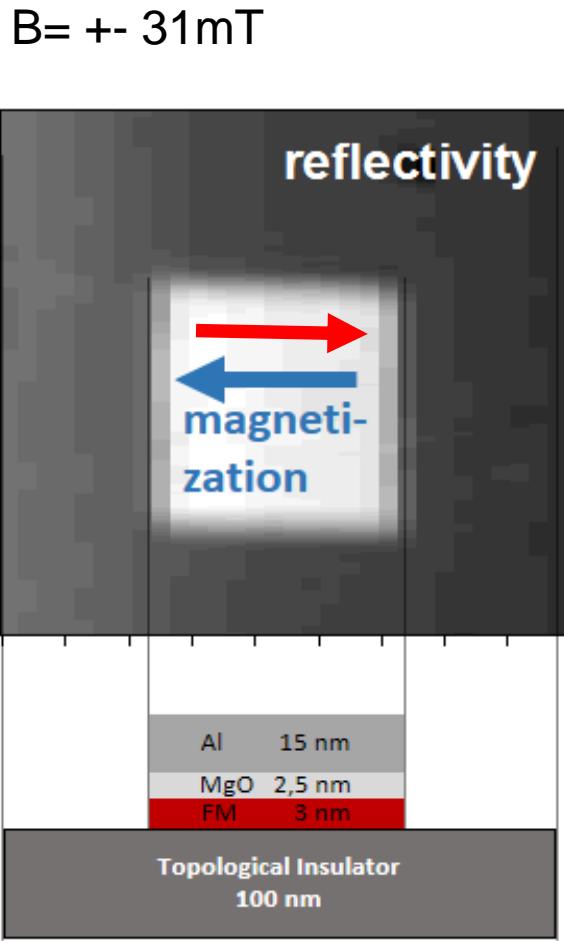
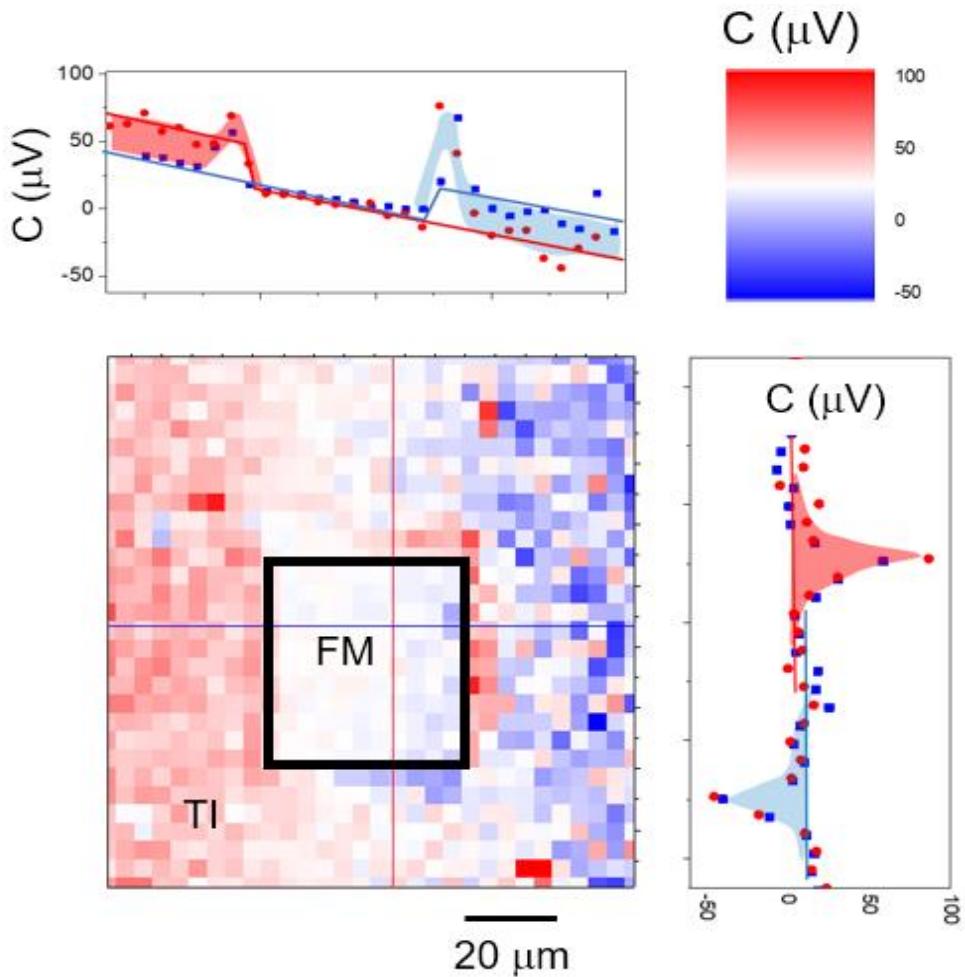
## Ferromagnet/ Topological insulator hybrid structures



# Topological matter

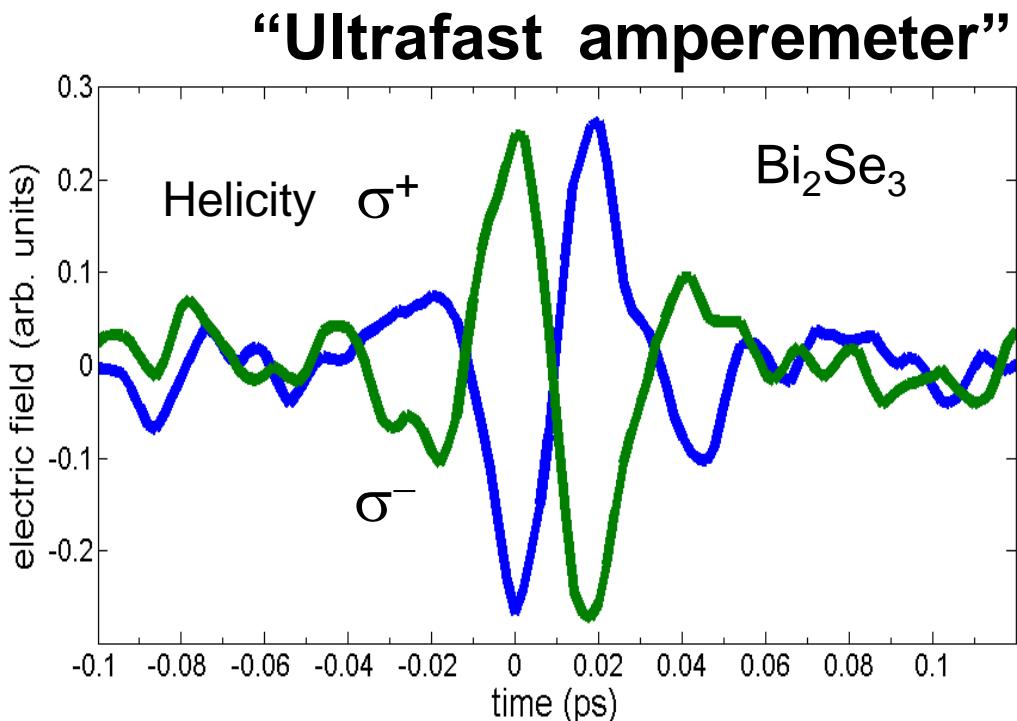
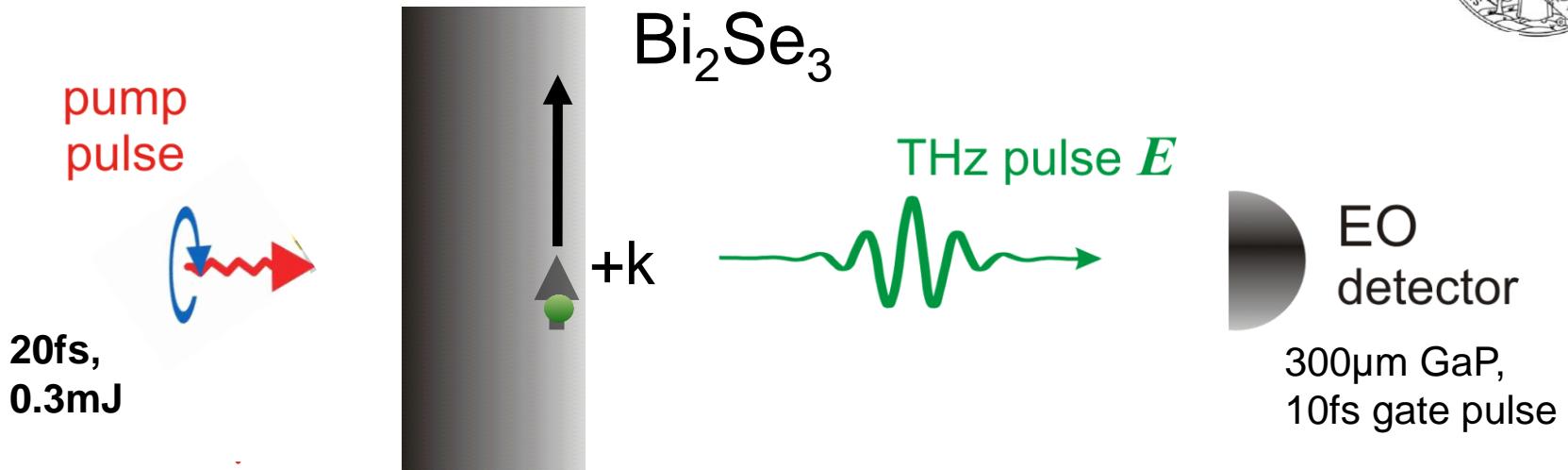


- Circular photovoltaic effect C with ferromagnetic structure





# Spins driven in Topological Insulators

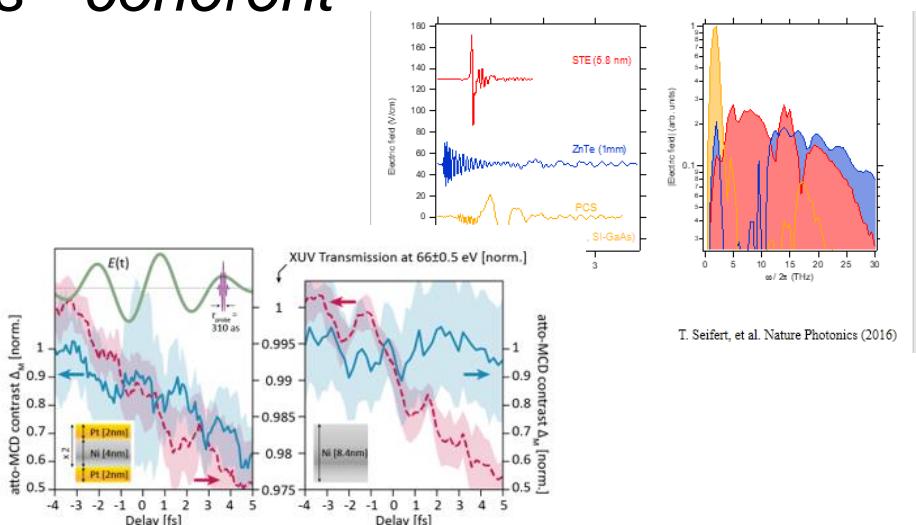
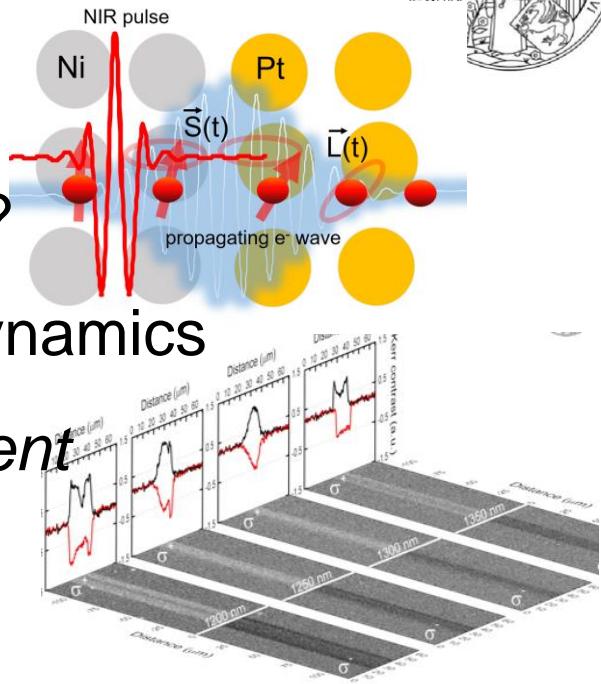


- THz emission is polarization dependent
- Full story here:  
L. Braun, et al., Nature Comm. 7, 13259 (2016).

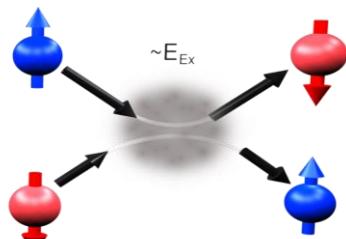
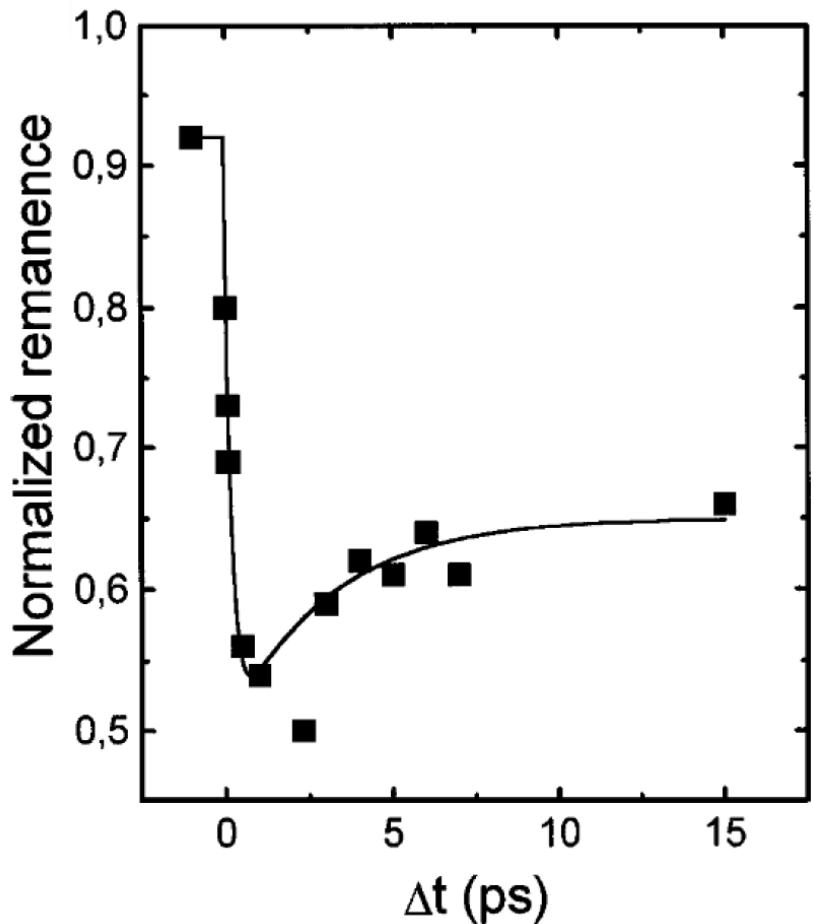
# Outline



- A coherent attosecond spintronics?
- The nature of femtosecond spin dynamics
- THz spintronic emitter – *noncoherent*
- Topological Insulators
- Lightwave electronics - *coherent*
- Summary



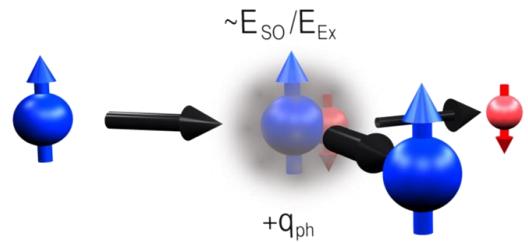
# Lightwave Spintronics – Attosecond dynamics



Metal	Exchange	Spin-orbit
Fe	52 fs	50 fs
Co	80 fs	52 fs
Ni	380 fs	48 fs

Laser-induced **demagnetization**

Beaurepaire et al. PRL 76, 4250 (1996)



# Family tree of models for ultrafast spin dynamics

## Electronic description

### Excitation

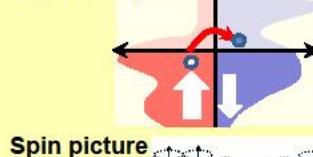


### Spin-scattering



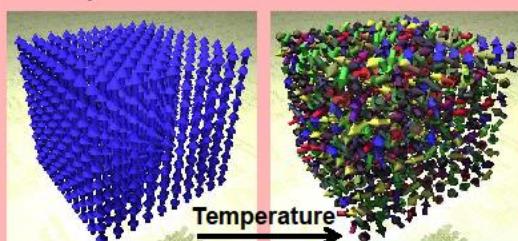
### Transition region

#### Electronic picture

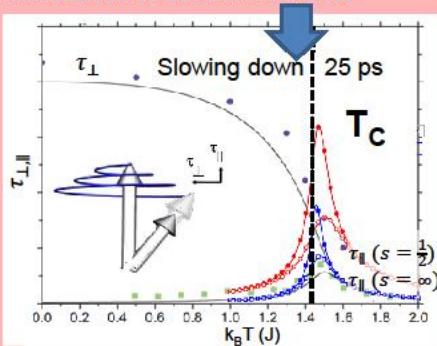


#### Spin projected description

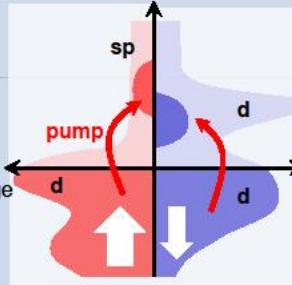
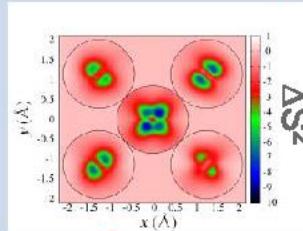
##### Thermal spin ensemble: atomistic



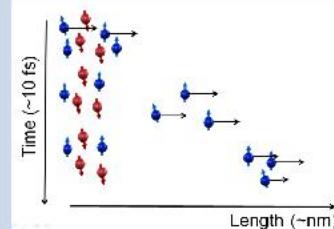
##### Landau-Lifshitz Bloch and M3TM



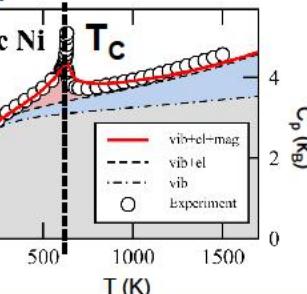
## Time dependent density functional theory (TDDFT)



### Spin diffusion



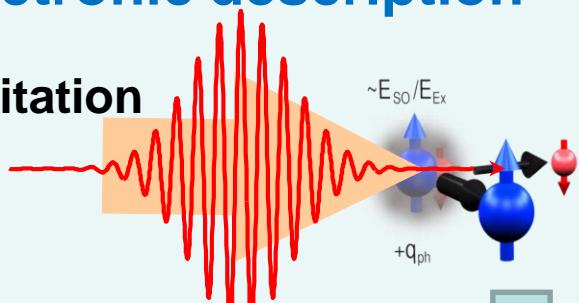
## Specific heat picture



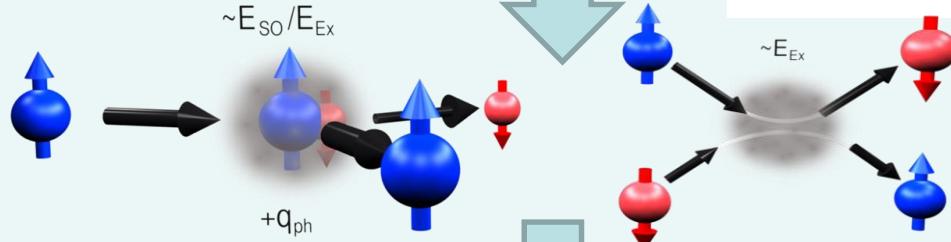
# Family tree of models for ultrafast spin dynamics

## Electronic description

### Excitation

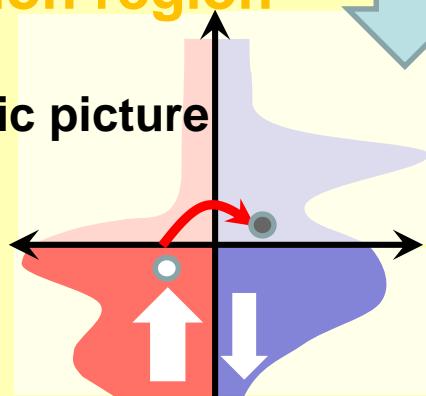


### Spin-scattering

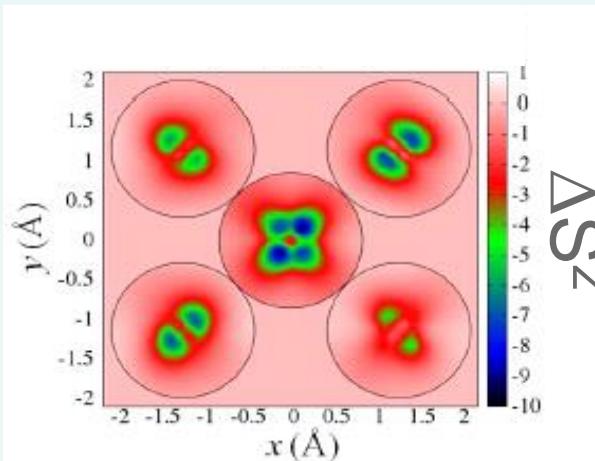
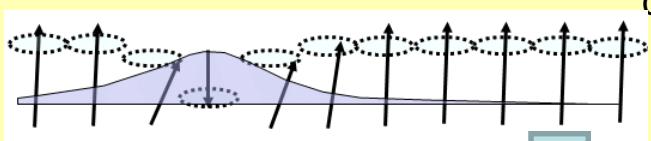


### Transition region

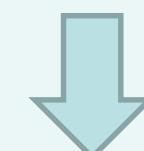
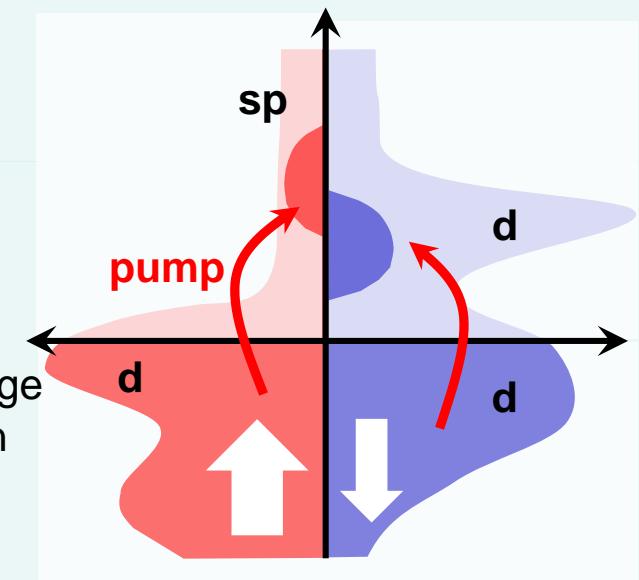
#### Electronic picture



#### Spin picture



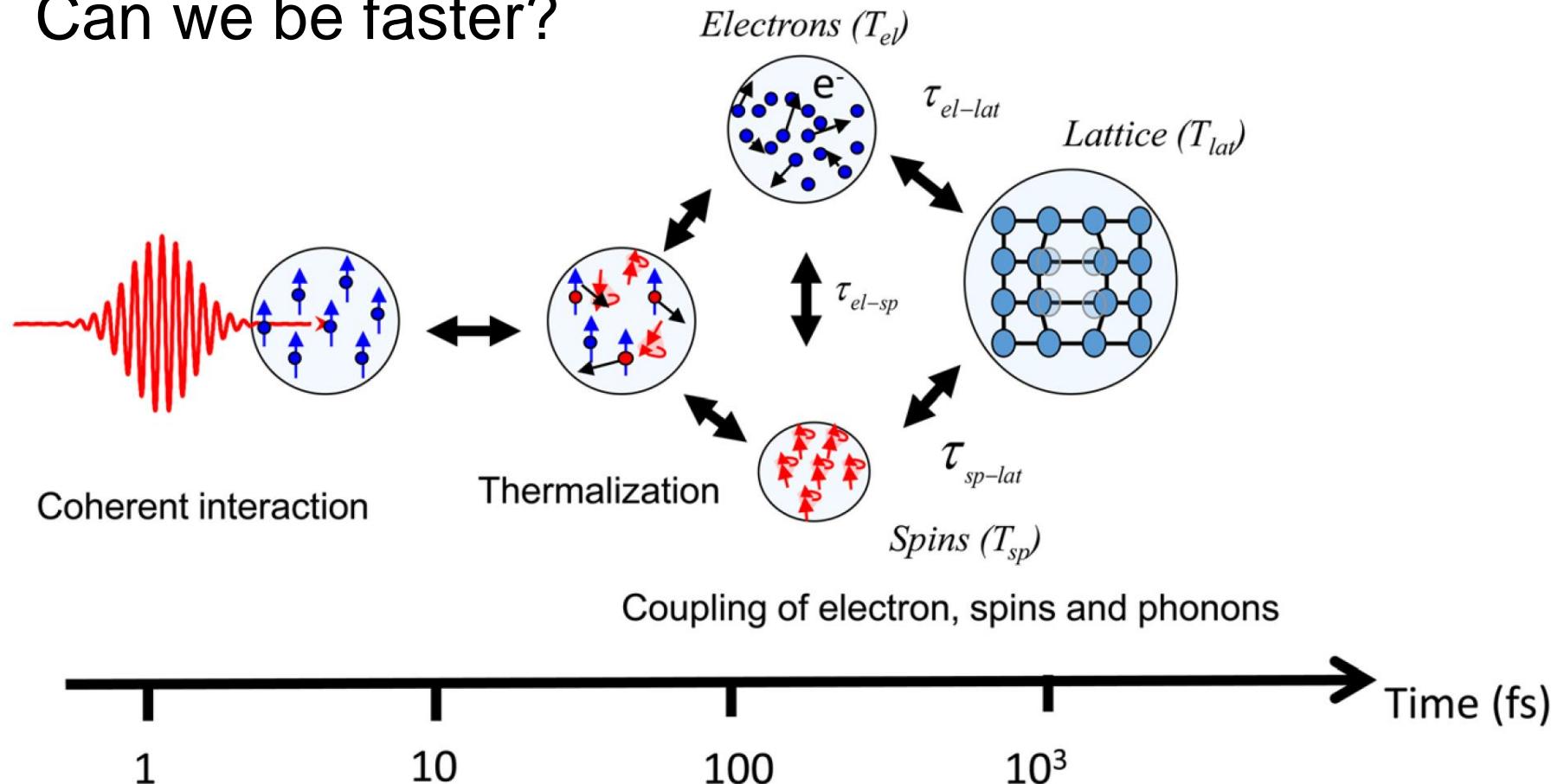
## Time dependent density functional theory (TDDFT)



# Lightwave Spintronics – Attosecond dynamics



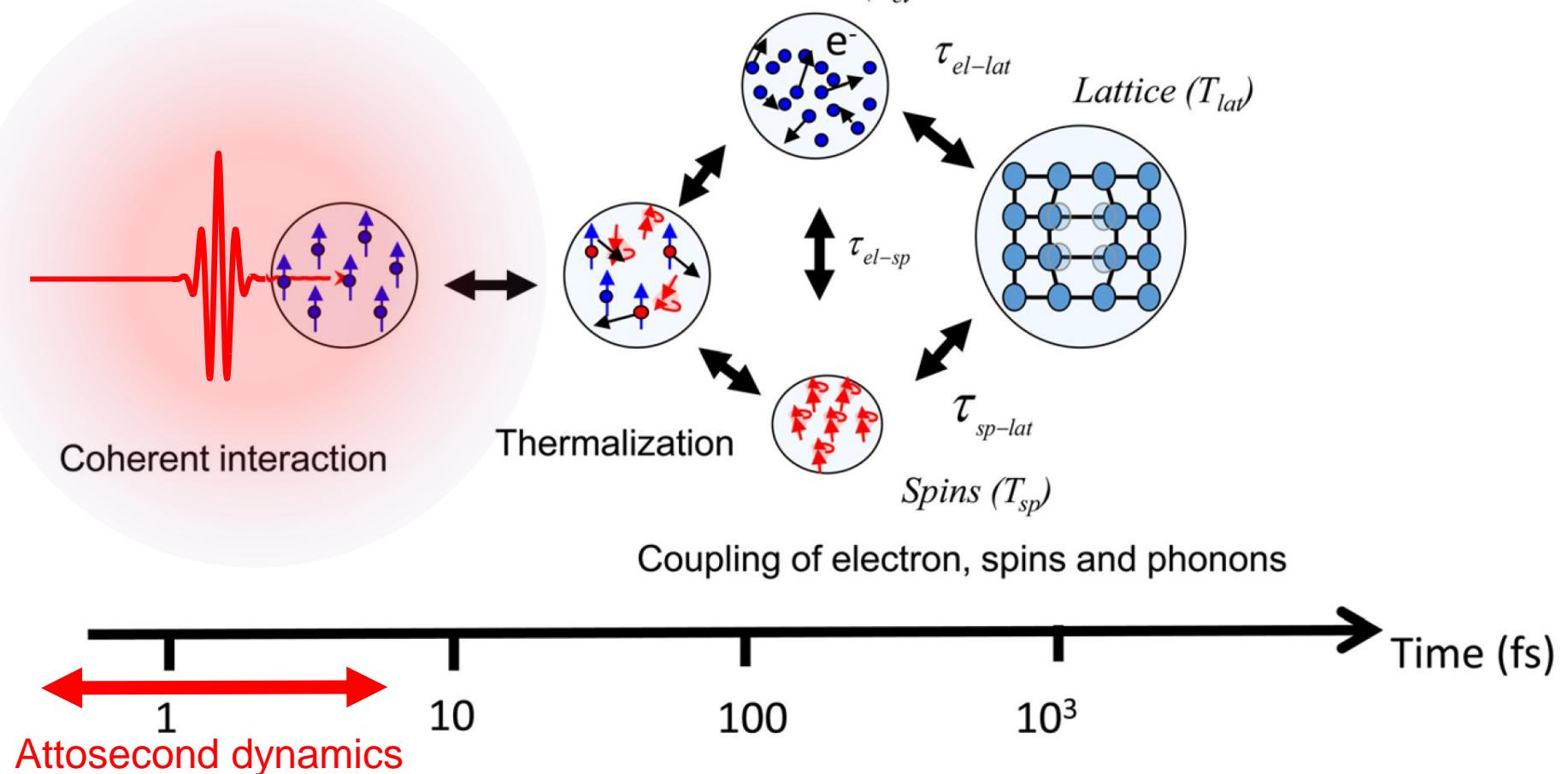
Can we be faster?



# Lightwave Spintronics – Attosecond dynamics



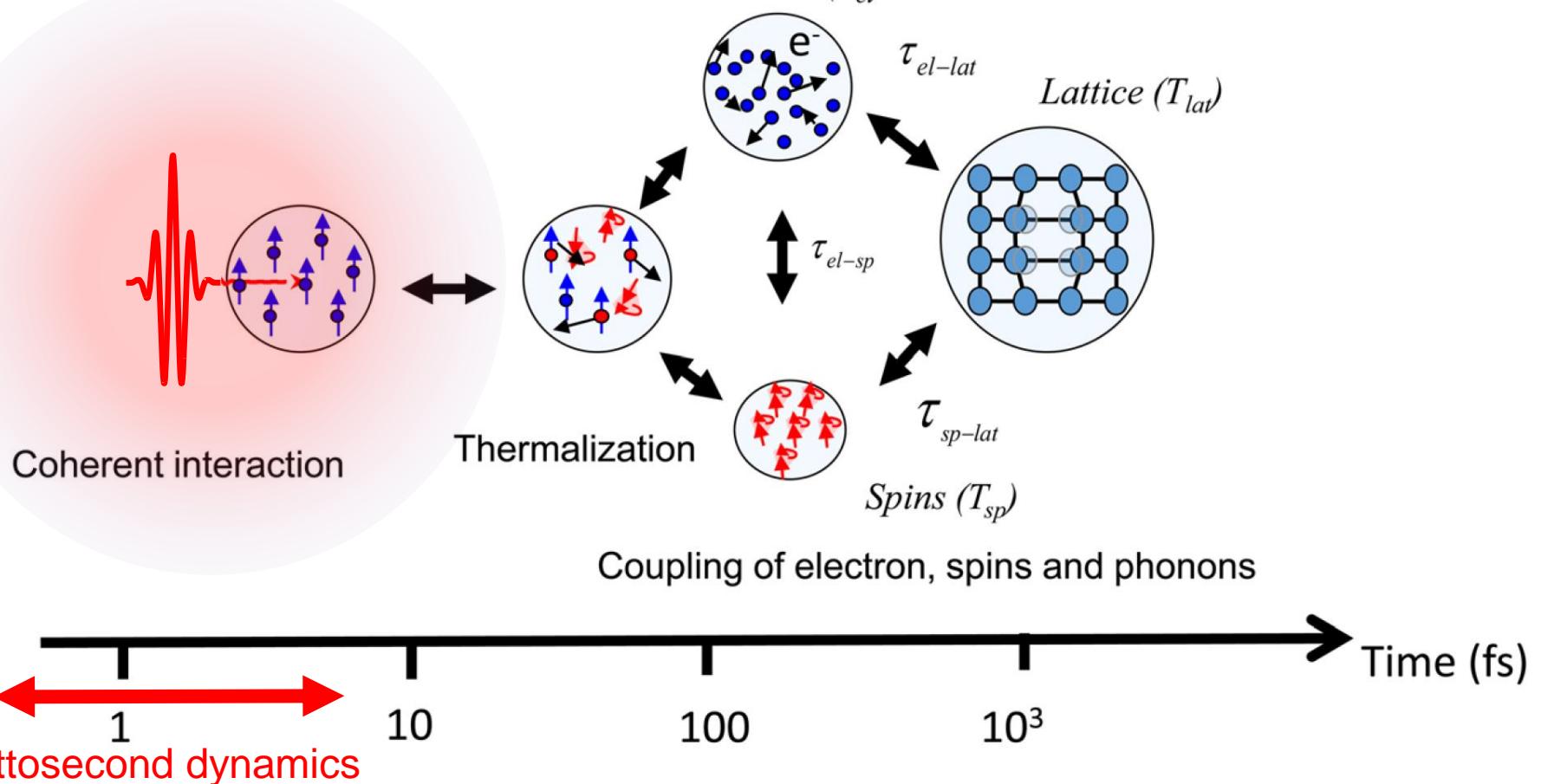
## Probe the coherence!



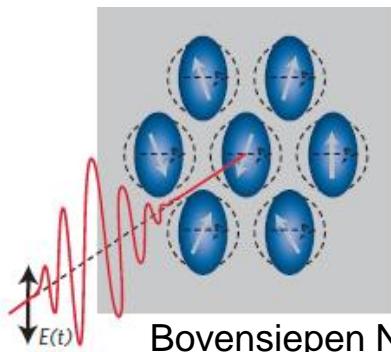
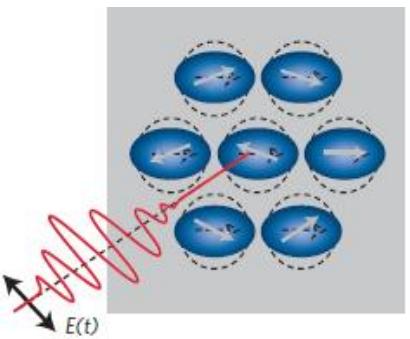
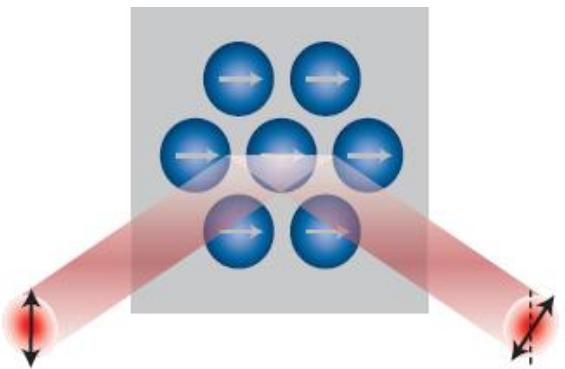
# Lightwave Spintronics – Attosecond dynamics



## Probe the coherence!

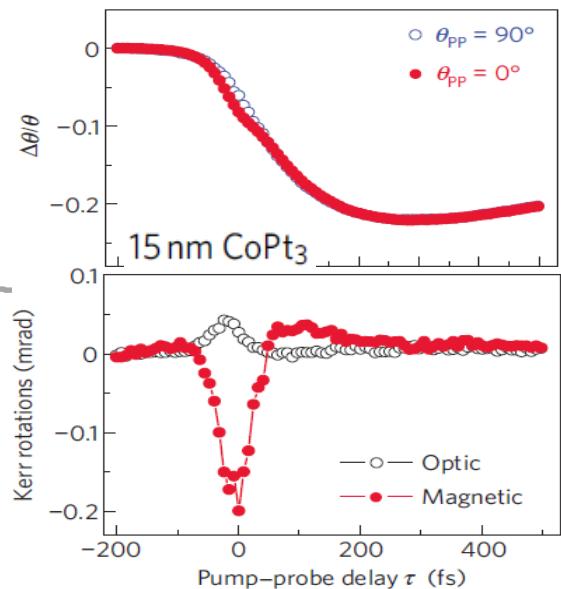
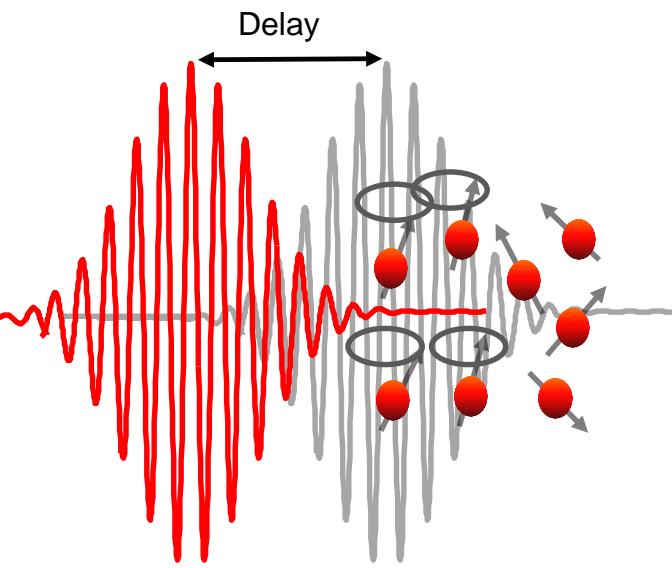


# Going sub-femtosecond: coherent effects



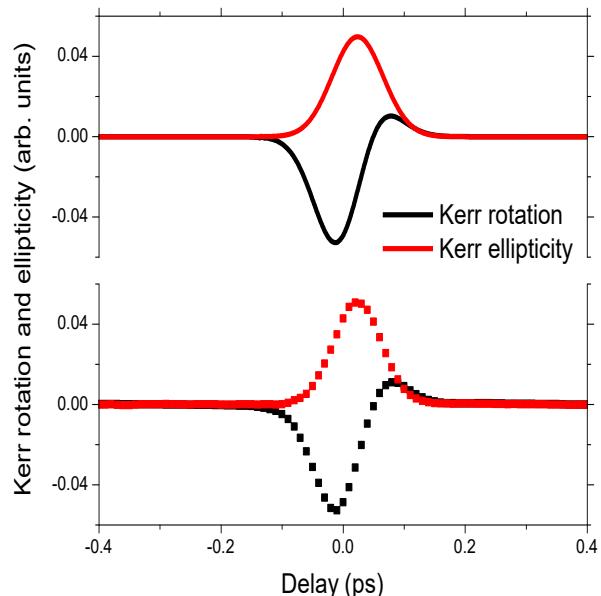
Bovensiepen Nat. Phys.

Laser cycle driven: ferromagnet  $\text{CoPt}_3$



Bigot et al. Nat. Phys.

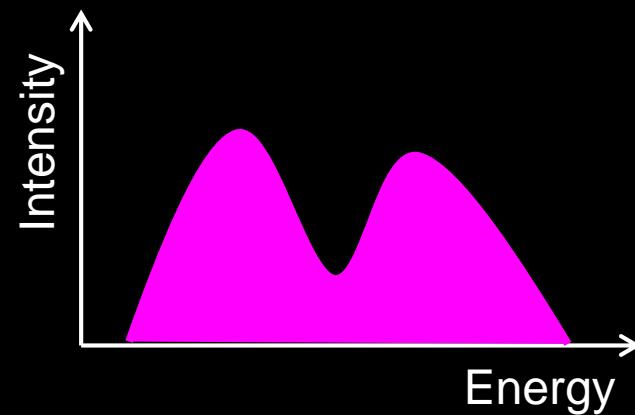
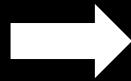
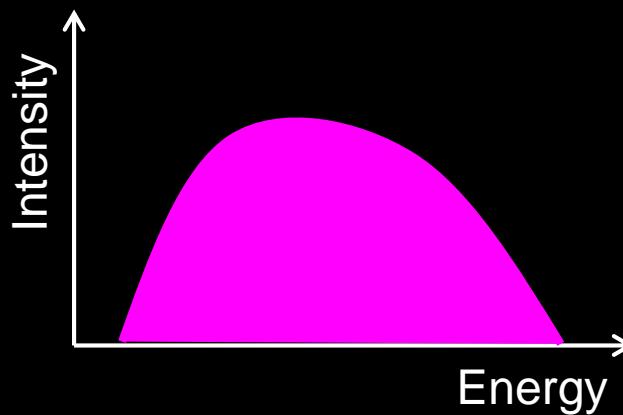
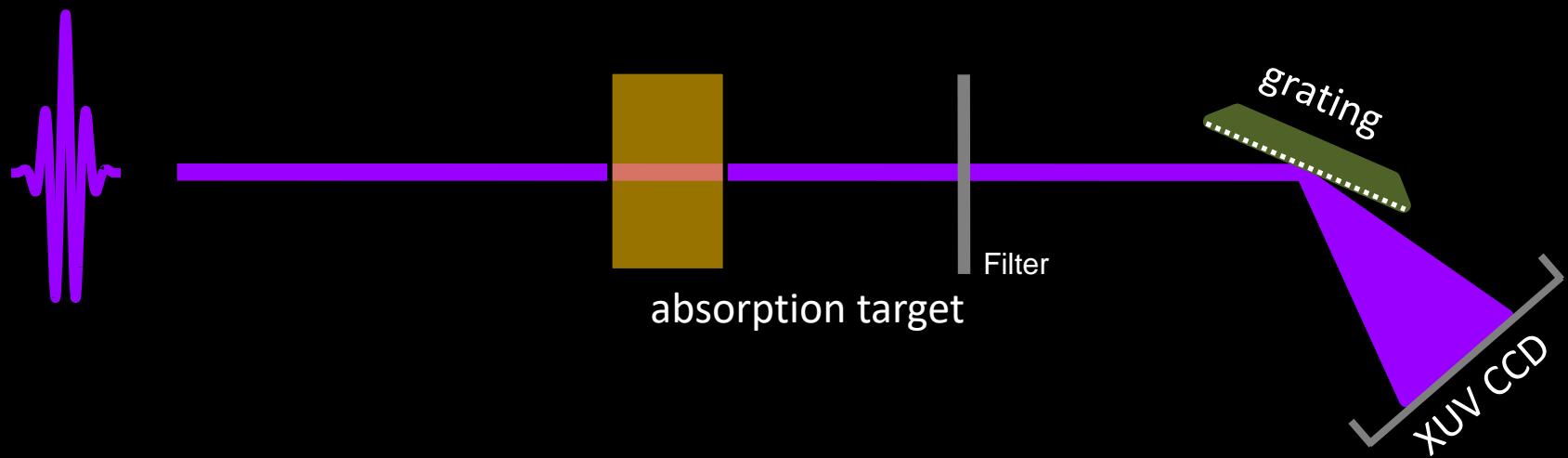
Topological insulator  $(\text{Bi}_{0.57}\text{Sb}_{0.43})_2\text{Te}_3$



Boschini et al. Sci. Rep.

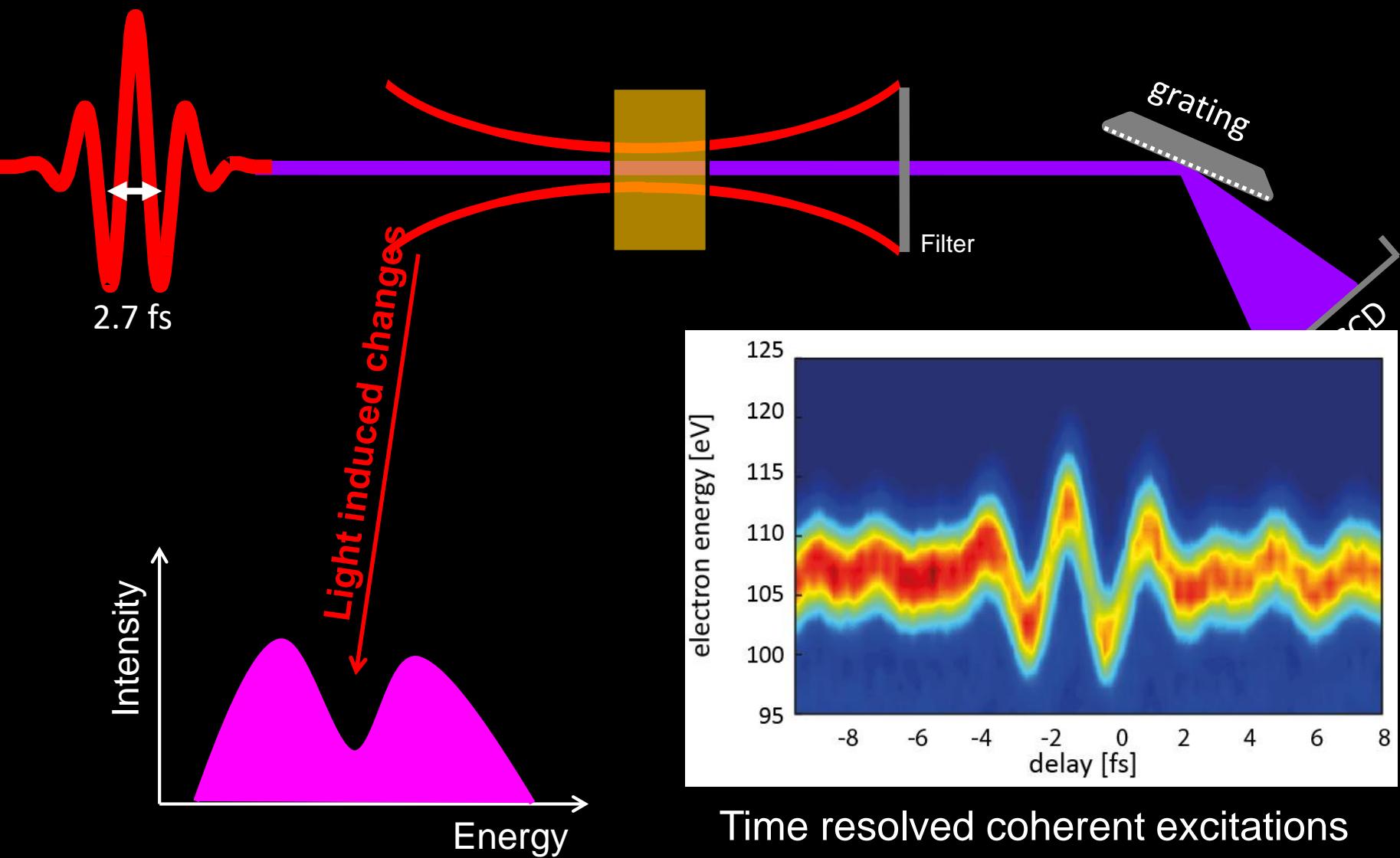
# Lightwave Spintronics – Attosecond dynamics

100-300 Attoseconds



# Lightwave Spintronics – Attosecond dynamics

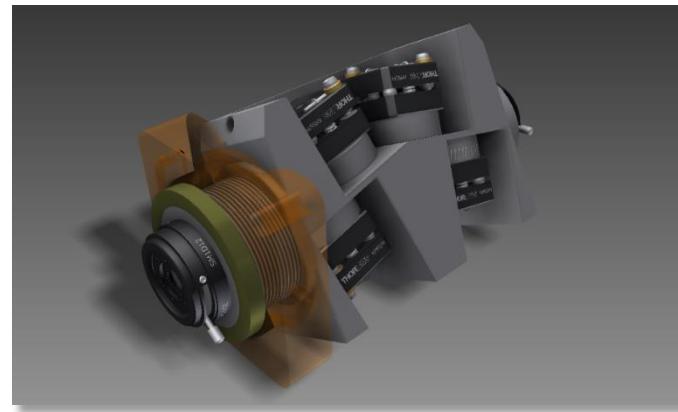
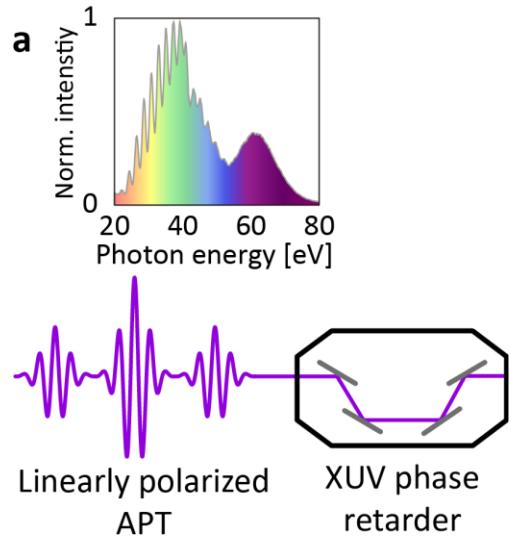
Few cycle light pulse, Pump



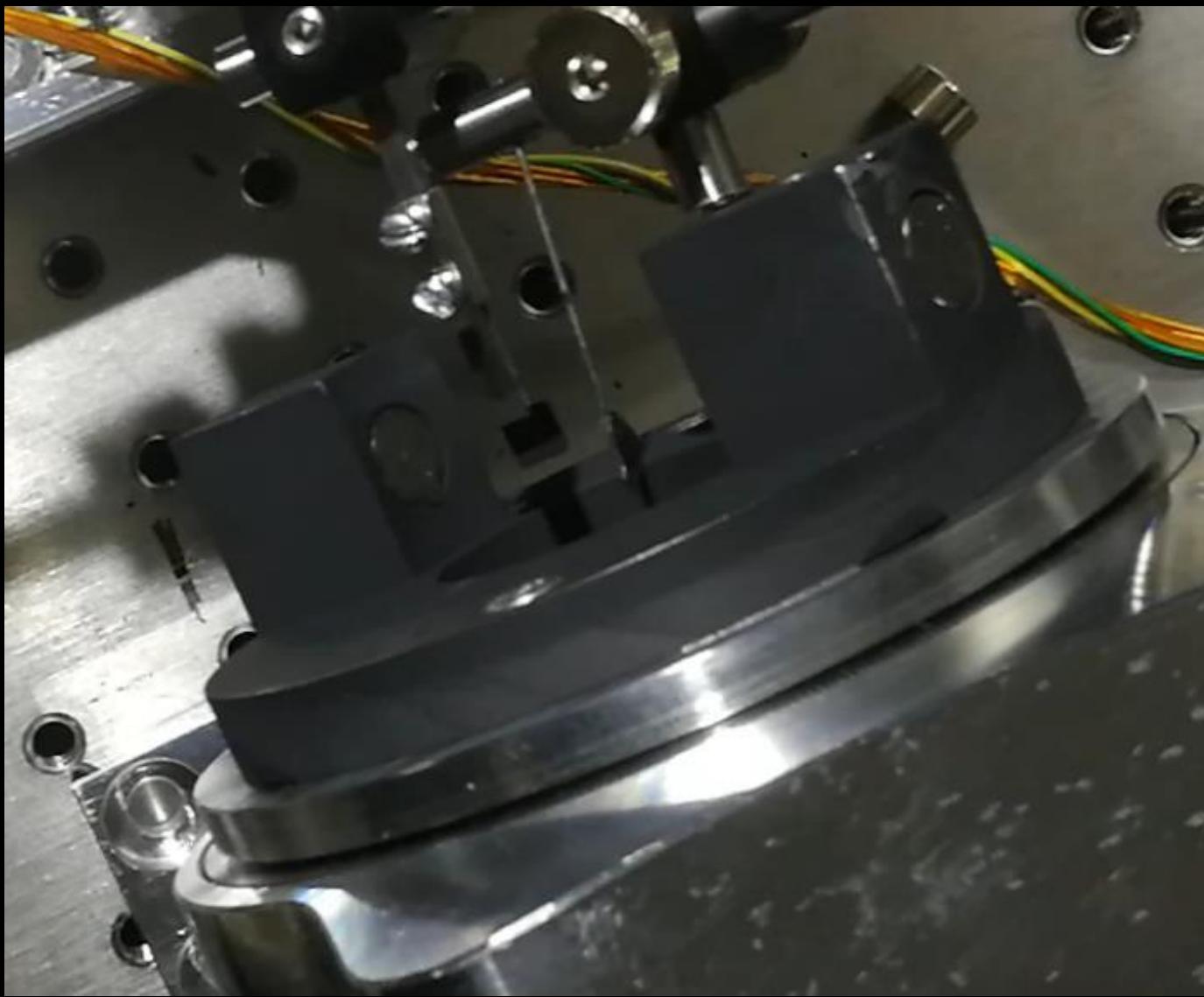
# Lightwave Spintronics – Attosecond dynamics



For ferromagnets we need attosecond x-ray dichroism:



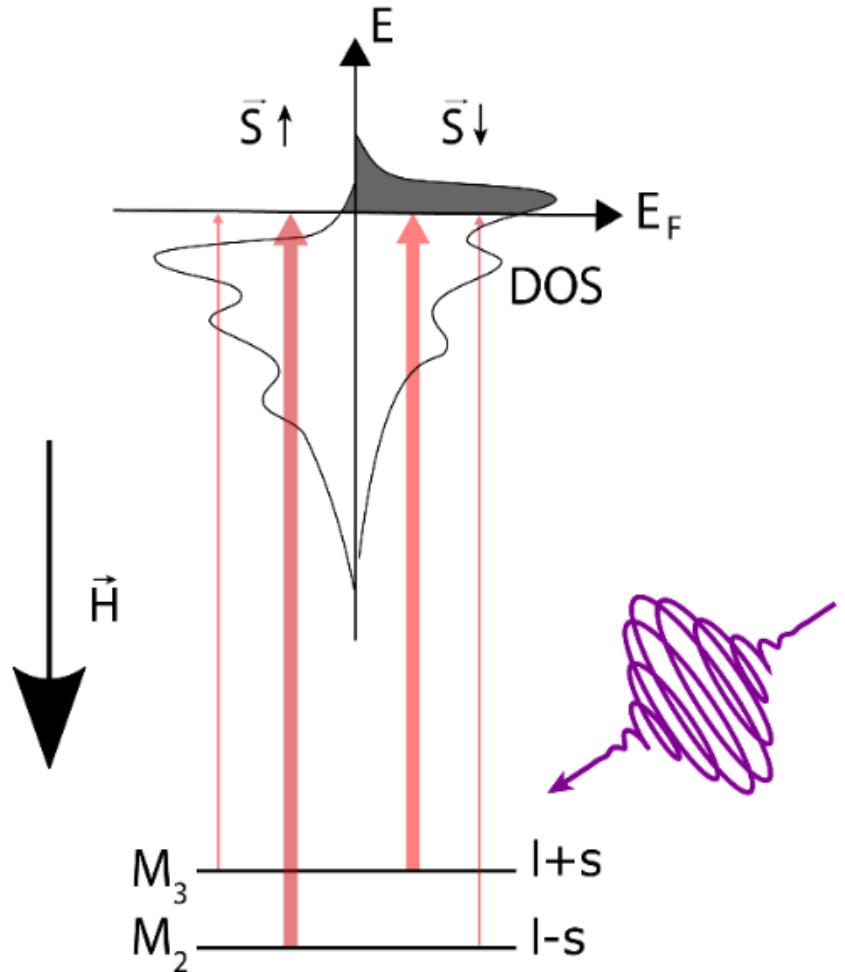
Mirror based Quarter Wave Plate (QWP)



# Lightwave Spintronics – Attosecond dynamics



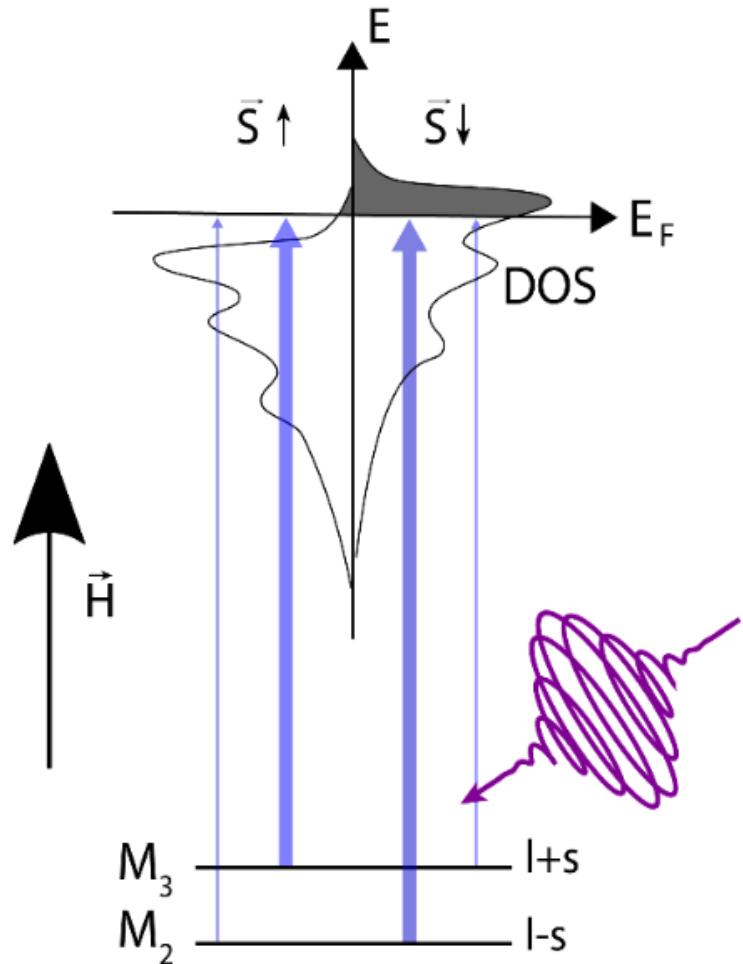
Ni M<sub>2,3</sub> edge



# Lightwave Spintronics – Attosecond dynamics



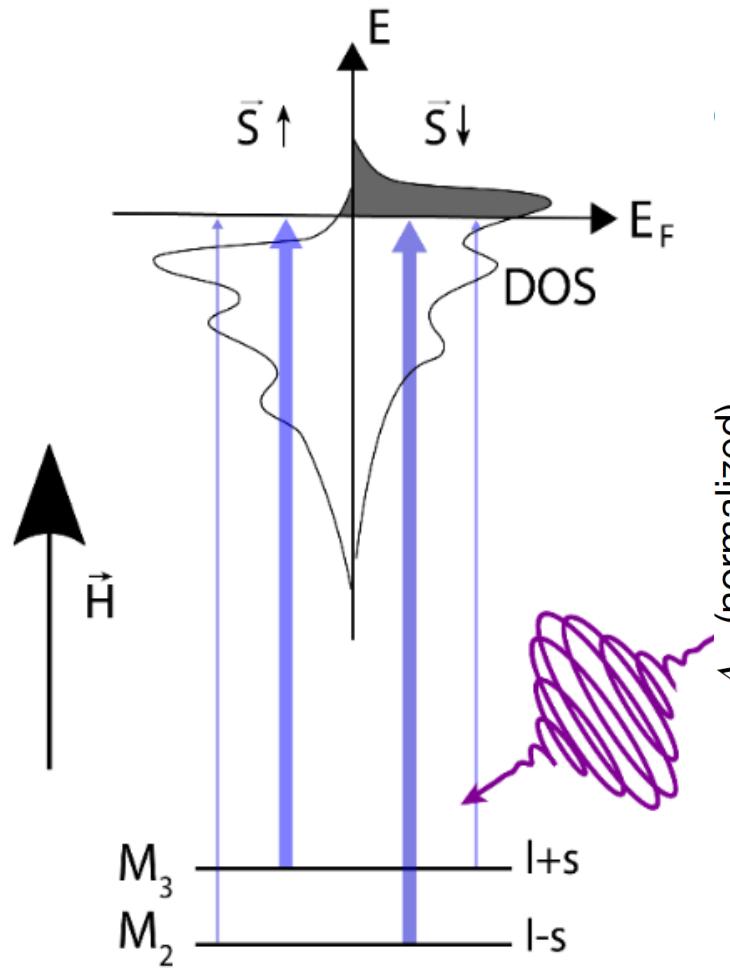
Ni M<sub>2,3</sub> edge



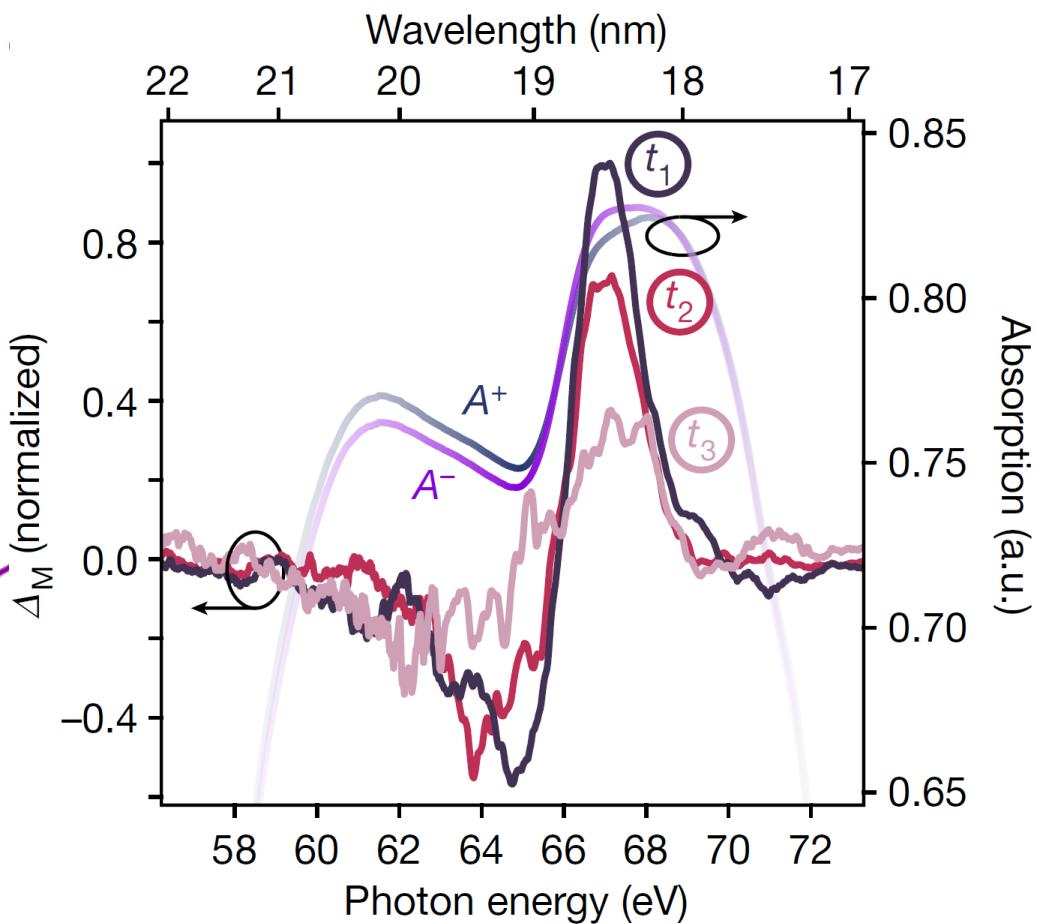
# Lightwave Spintronics – Attosecond dynamics



Ni M<sub>2,3</sub> edge



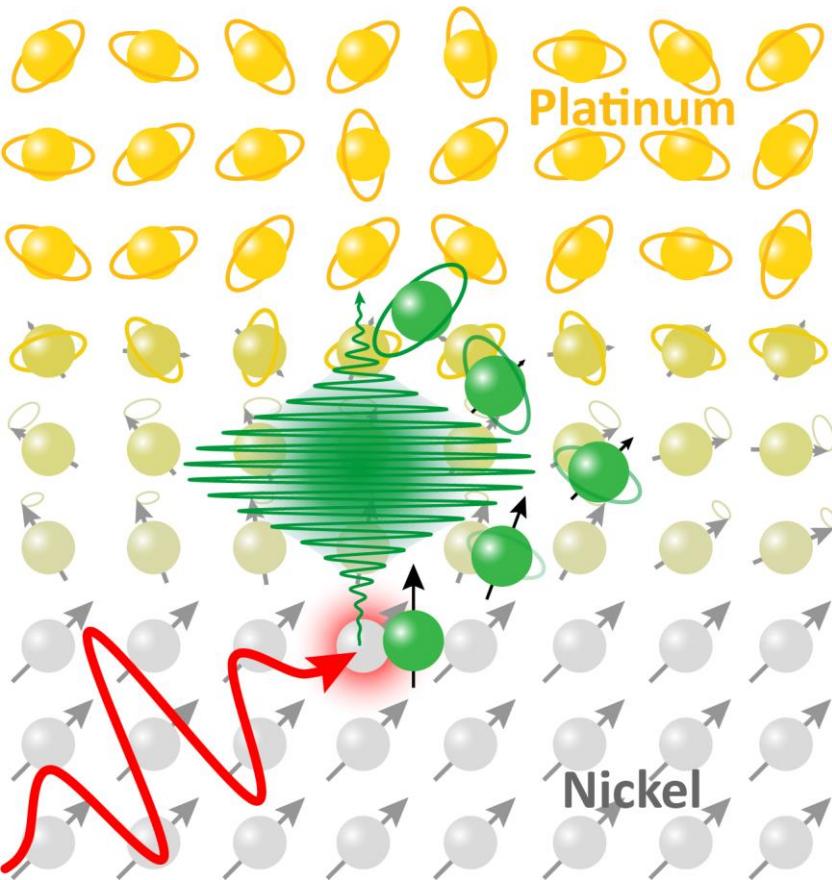
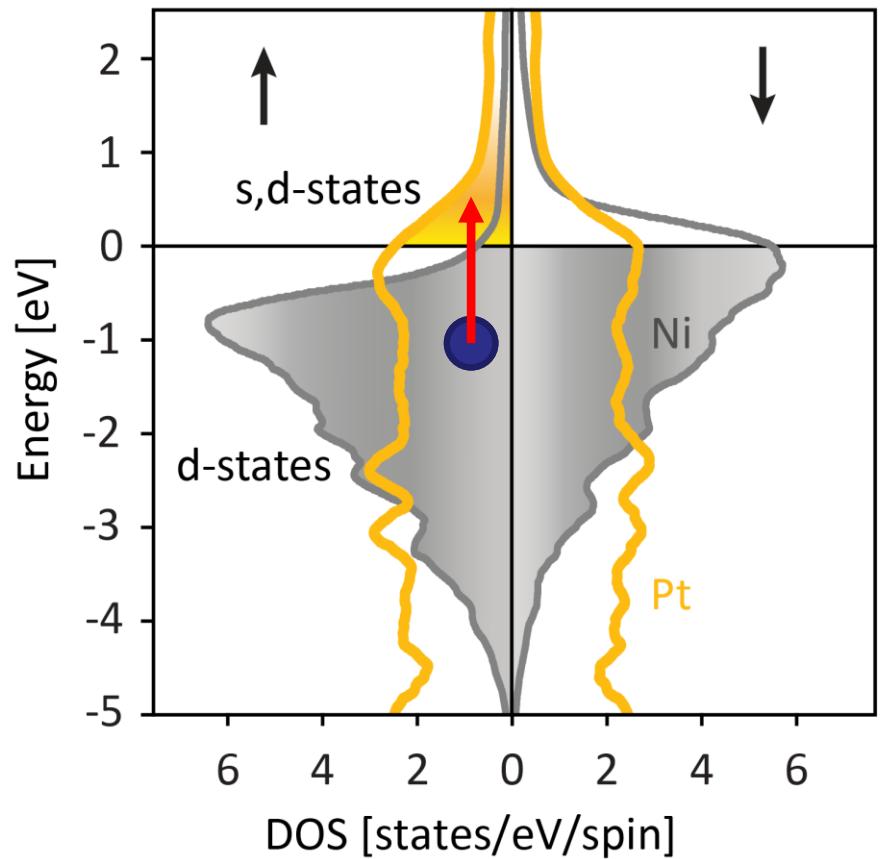
XMCD for different delay  $t_1$ ,  $t_2$ ,  $t_3$



# Lightwave Spintronics – Attosecond dynamics

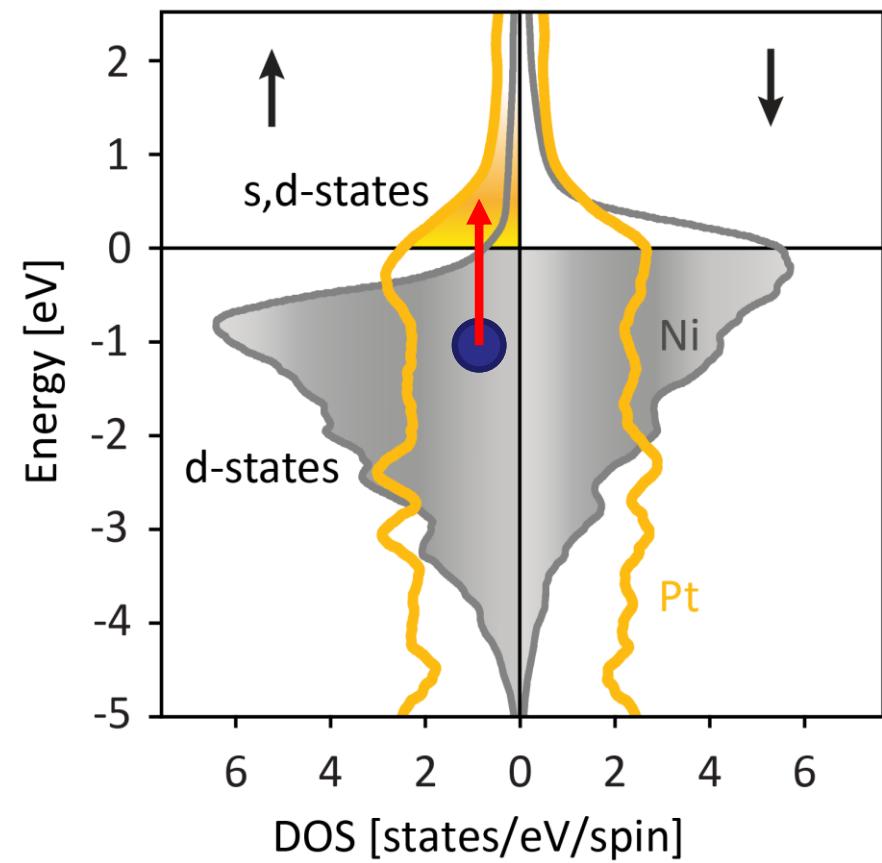


Optically Induced Spin Transfer (OIISTR):  
coherent spin motion

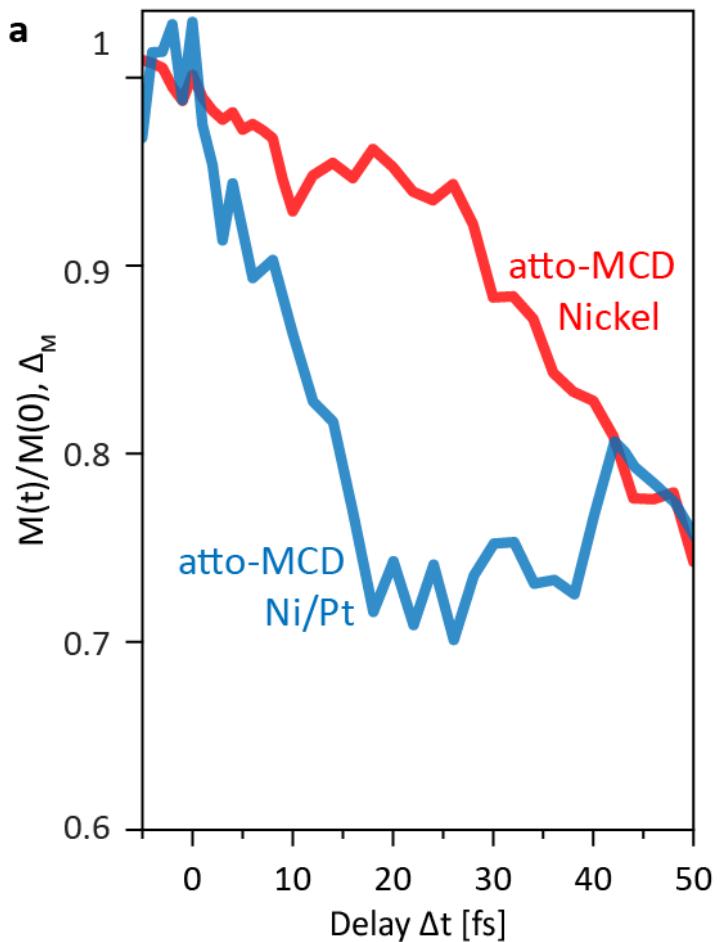


# Lightwave Spintronics – Attosecond dynamics

Optically Induced Spin Transfer (OIISTR):  
predicted ab-initio by time resolved DFT



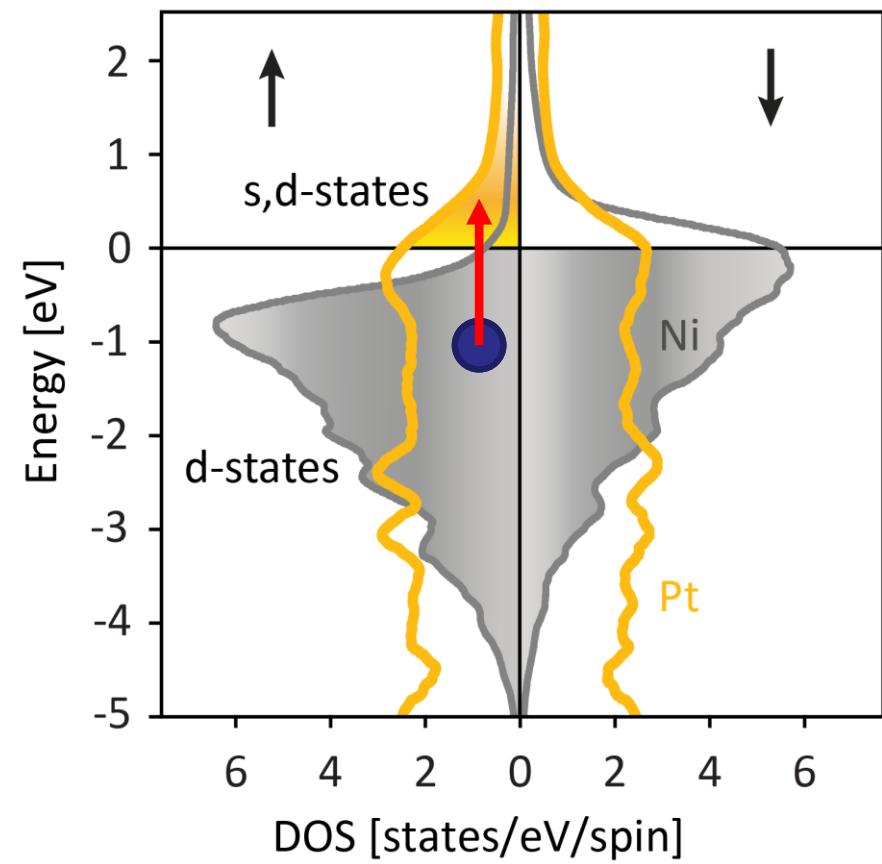
Ab-initio theory by trDFT:  
Sangeeta Sharma MBI Berlin



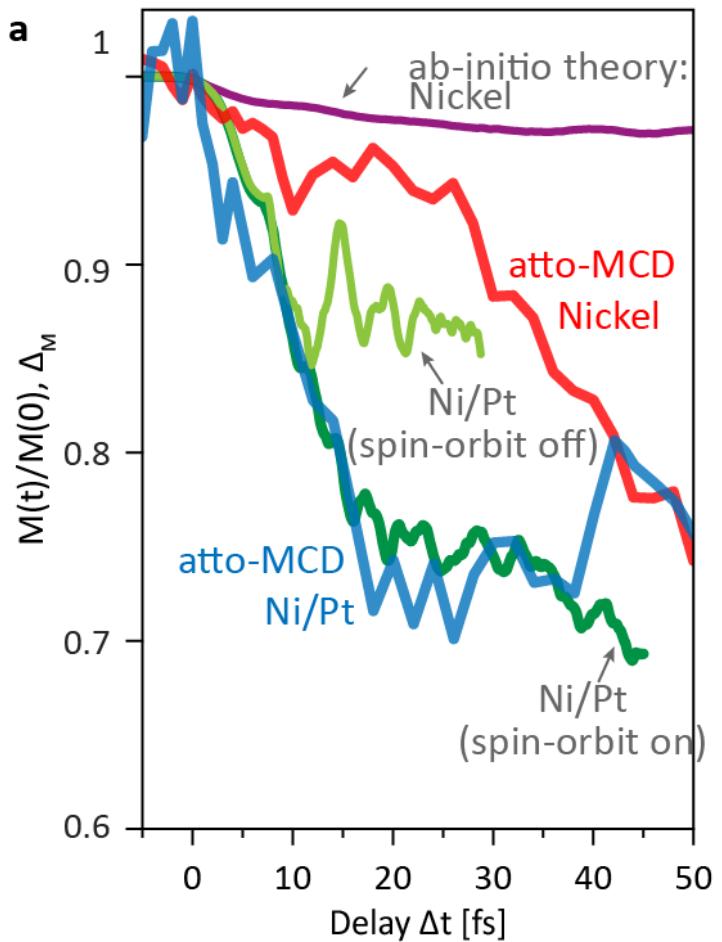
# Lightwave Spintronics – Attosecond dynamics



Optically Induced Spin Transfer (OISTR):  
predicted ab-initio by time resolved DFT



Ab-initio theory by trDFT:  
Sangeeta Sharma MBI Berlin



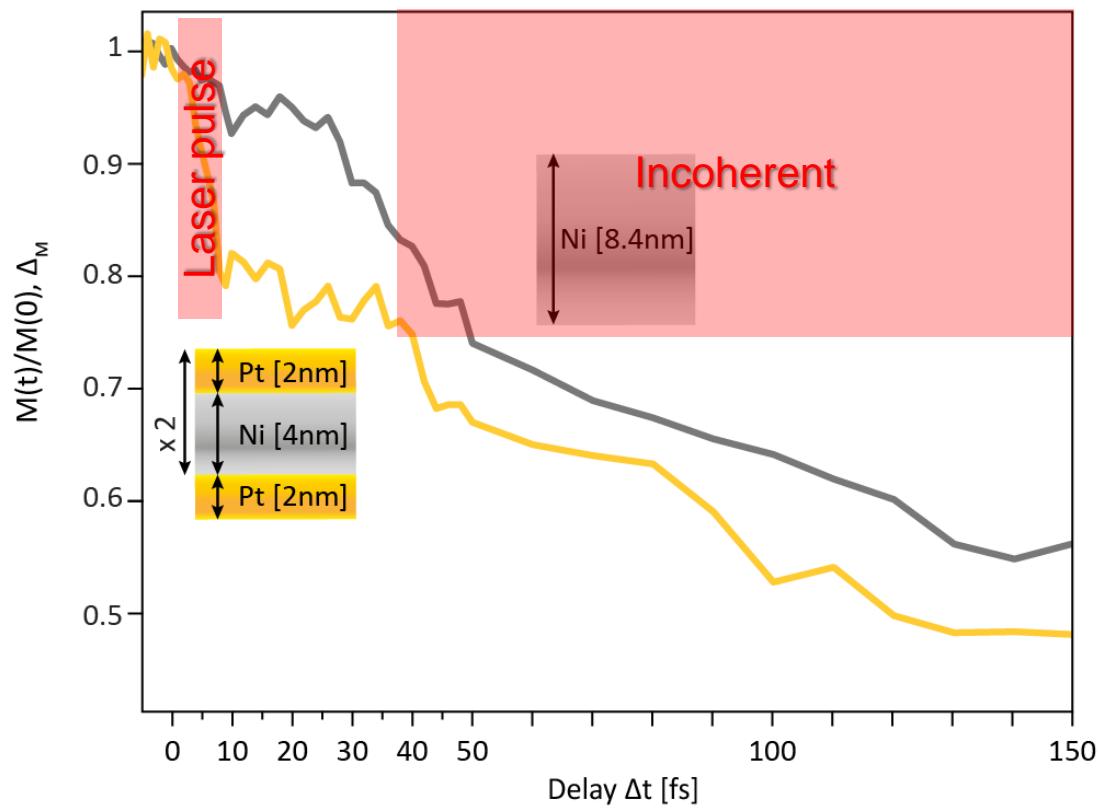
Fluence NIR =  $2 \times 10^{12}$  W cm $^{-2}$

F. Siegrist *et al.*, Light-wave dynamic control of magnetism,  
Nature 571, 240–244 (2019)

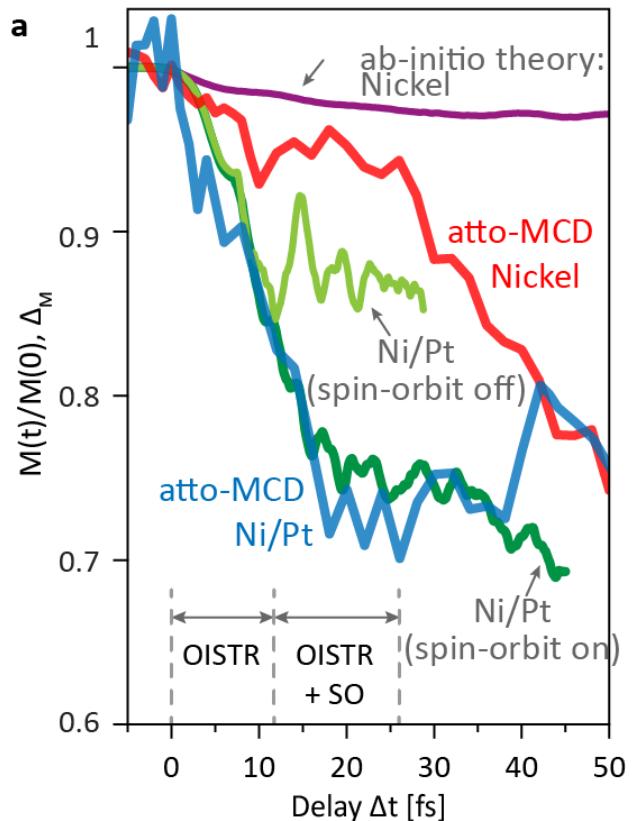


## Optically Induced Spin Transfer (OISTR): predicted ab-initio by time resolved DFT

### Experiment



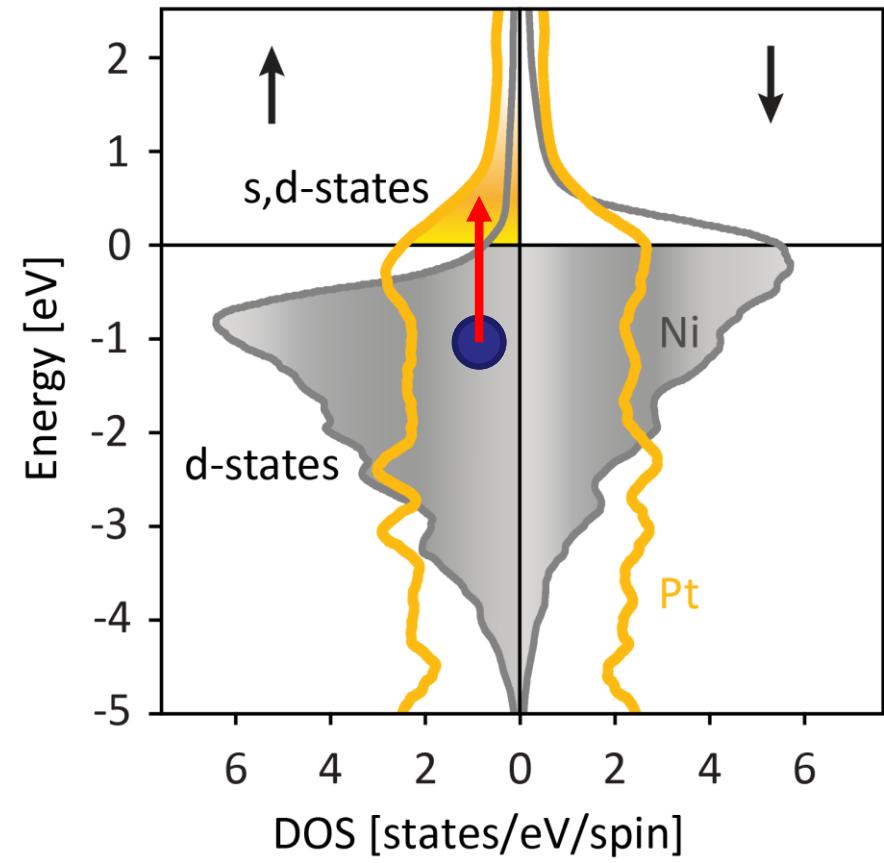
### Theory



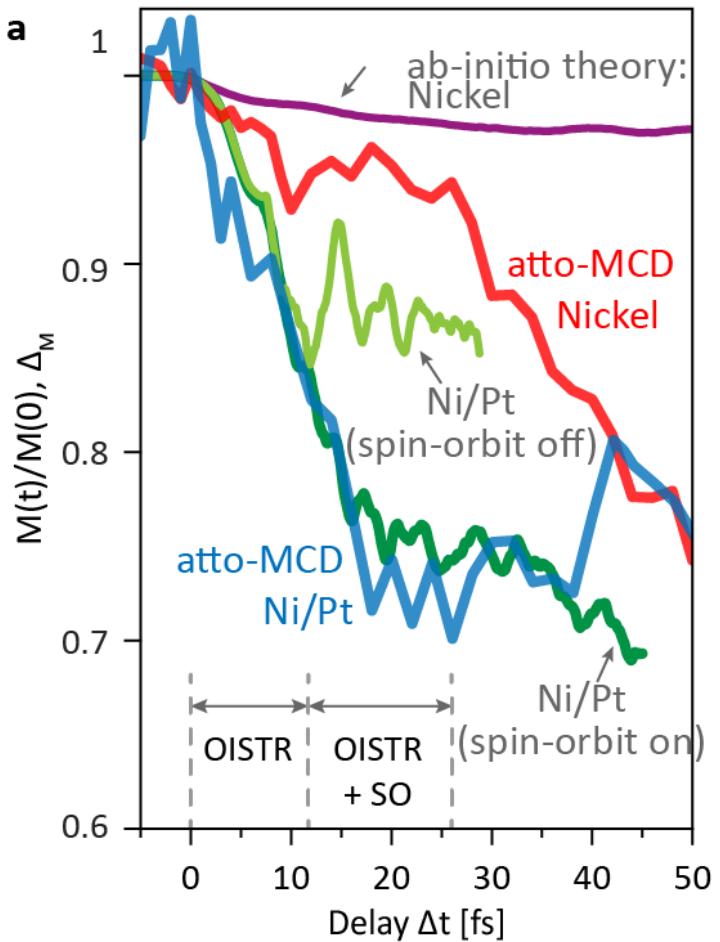
# Lightwave Spintronics – Attosecond dynamics



Optically Induced Spin Transfer (OISTR):  
predicted ab-initio by time resolved DFT



Ab-initio theory by trDFT:  
Sangeeta Sharma MBI Berlin



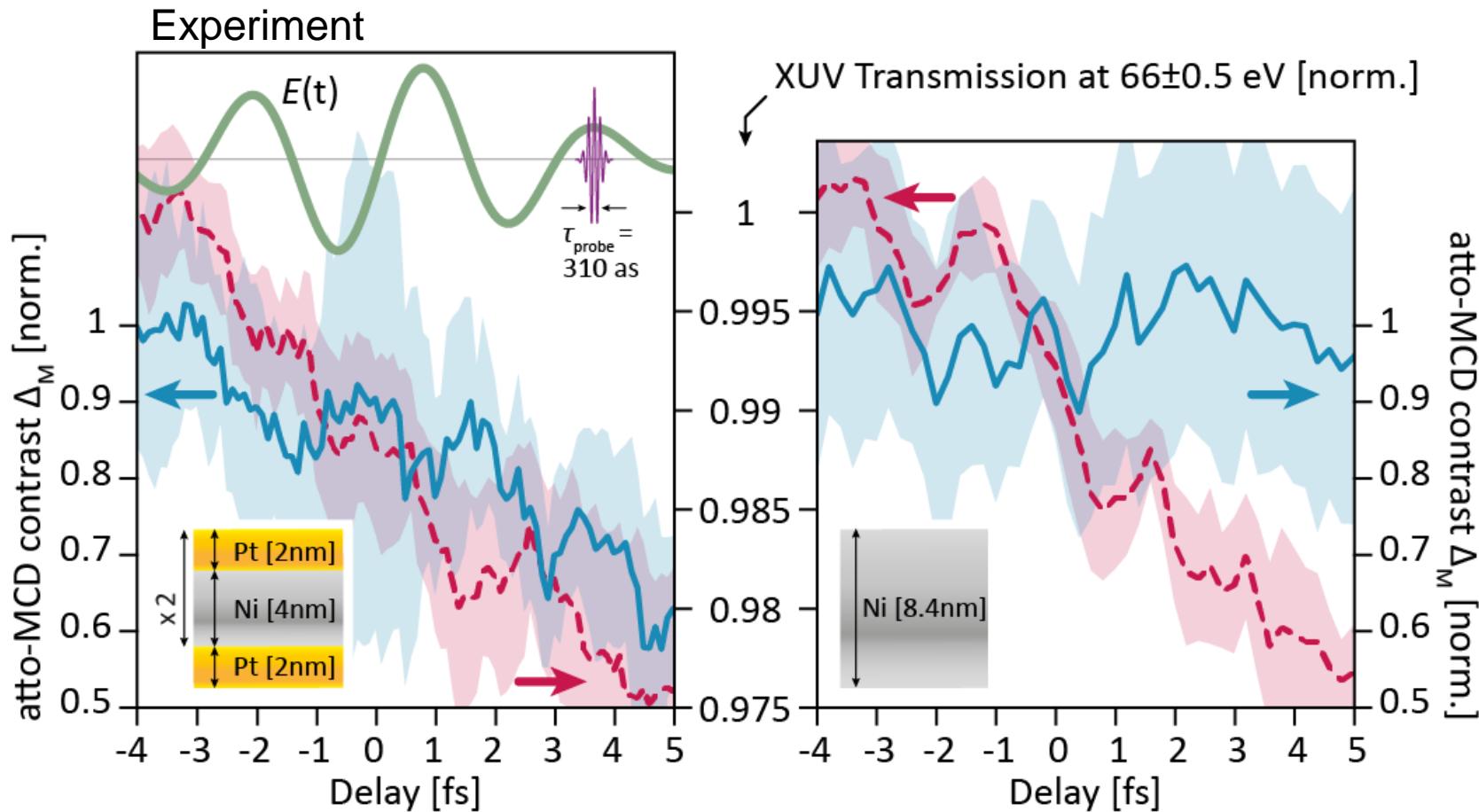
Fluence NIR =  $2 \times 10^{12}$  W cm<sup>-2</sup>

F. Siegrist *et al.*, Light-wave dynamic control of magnetism,  
Nature 571, 240–244 (2019)

# Lightwave Spintronics – Attosecond dynamics



Few fs step like decay with Pt interface (resolution 310 as)



Fluence NIR =  $4 \times 10^{12} \text{ W cm}^{-2}$

F. Siegrist *et al.*, Light-wave dynamic control of magnetism,  
Nature 571, 240–244 (2019)

# Collaborations



@spintronicsHGW



## Attosecond spin dynamics:

Florian Siegrist, Julia Gessner, Marcus Ossiander, Martin Schultze, Technical University Graz/ Max-Planck-Institute für Quantum Optics, Munich

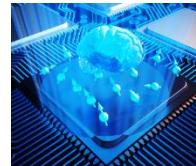
J. Dewhurst, Sangeeta Sharma, Max-Born-Institute, Berlin, Max-Planck-Institut for Microstructure Physics, Halle



Priority program  
Topologische  
Isolatoren



Priority program  
Skyrmionics



FET Open SpinAge



META ZIK PlasMark



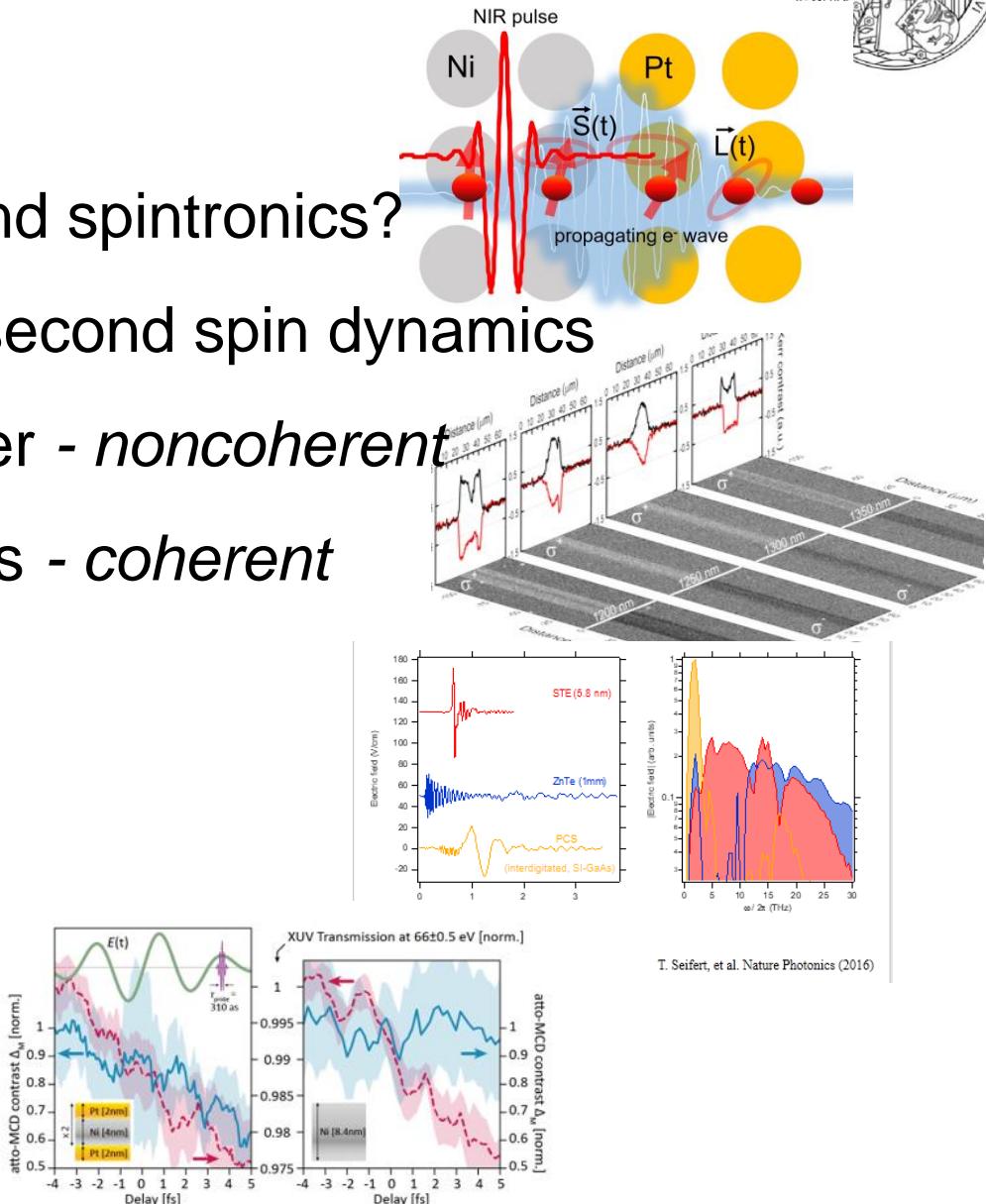
Bundesministerium  
für Bildung  
und Forschung

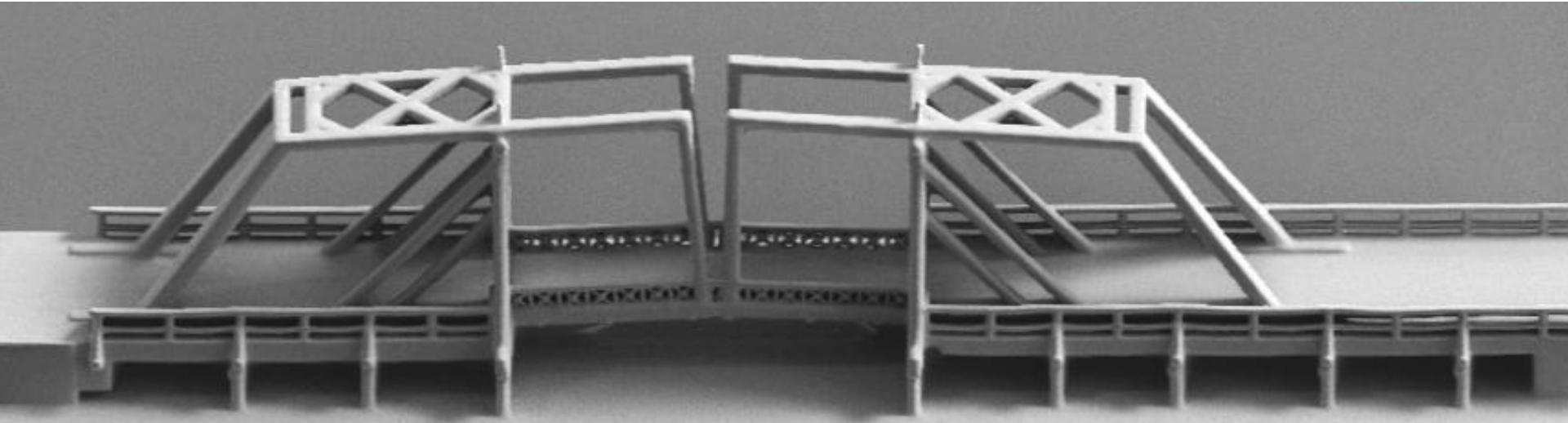
DAAD

# Outline



- A coherent attosecond spintronics?
- The nature of femtosecond spin dynamics
- THz spintronic emitter - *noncoherent*
- Lightwave electronics - *coherent*
- Summary





See NDR feature on our new labs: Nordmagazin or <http://www.physik.uni-greifswald.de/aktuelles.html>



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Greifswald

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