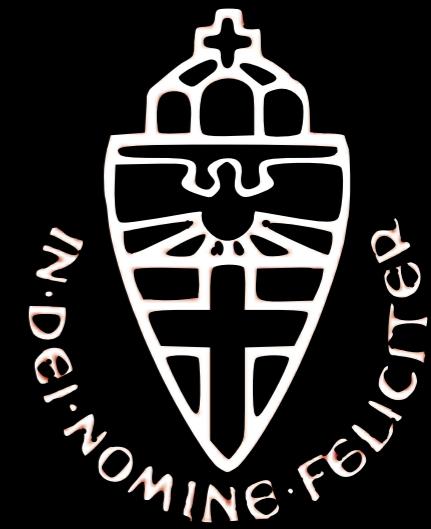
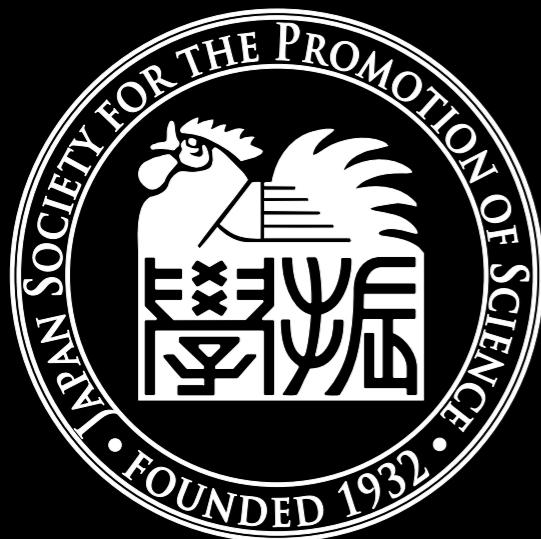


# Ultrafast optical manipulation of magnetic materials

D. Bossini

*Technical University Dortmund, Germany*



# Ultrafast

# Spintronics

- ultrafast demagnetisation
- all-optical switching
- inverse-Faraday effect
- magnetic phase-transitions

- spin pumping
- spin-Hall effect
- spin Seebeck effect
- magneto-resistance

Optics

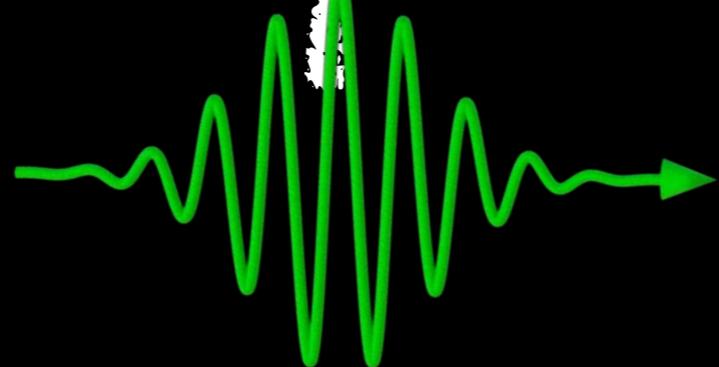
Transport

# Antiferromagnets

# Ultrafast

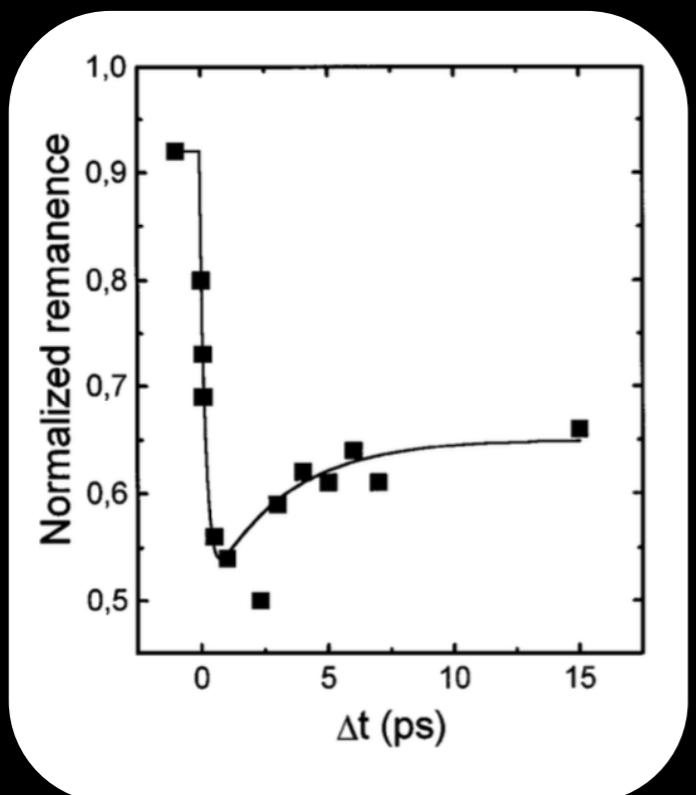
# Spintronics

Femtosecond time-scale  
Limited dissipations



## Antiferromagnets

# Metallic magnets



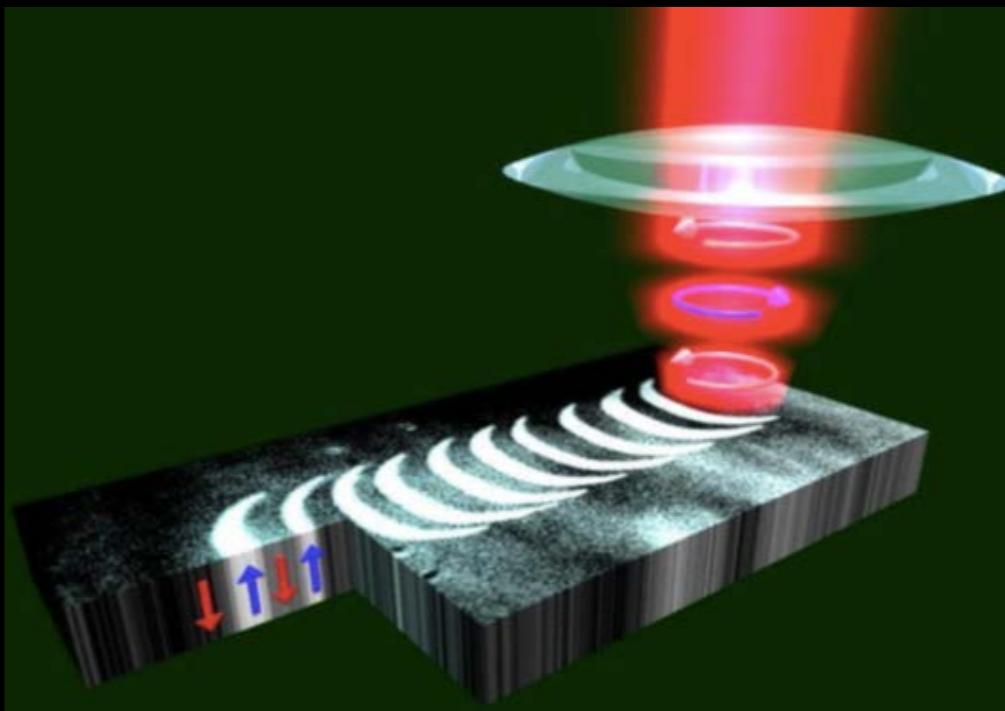
E. Beaurepaire et al. PRL **76**, 4250 (1996)



E. Beaurepaire  
1959-2018

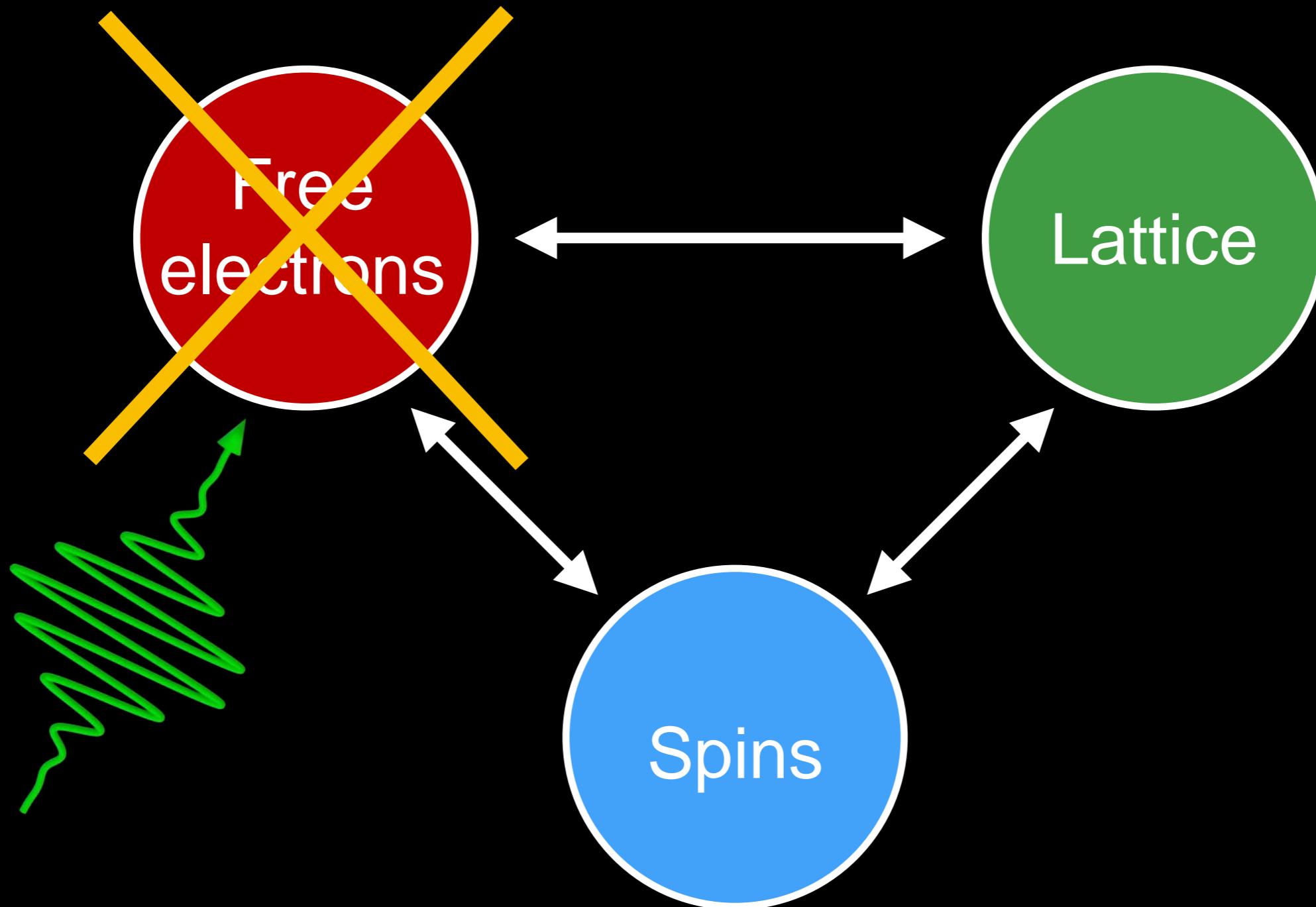


J. Y. Bigot  
1956-2018

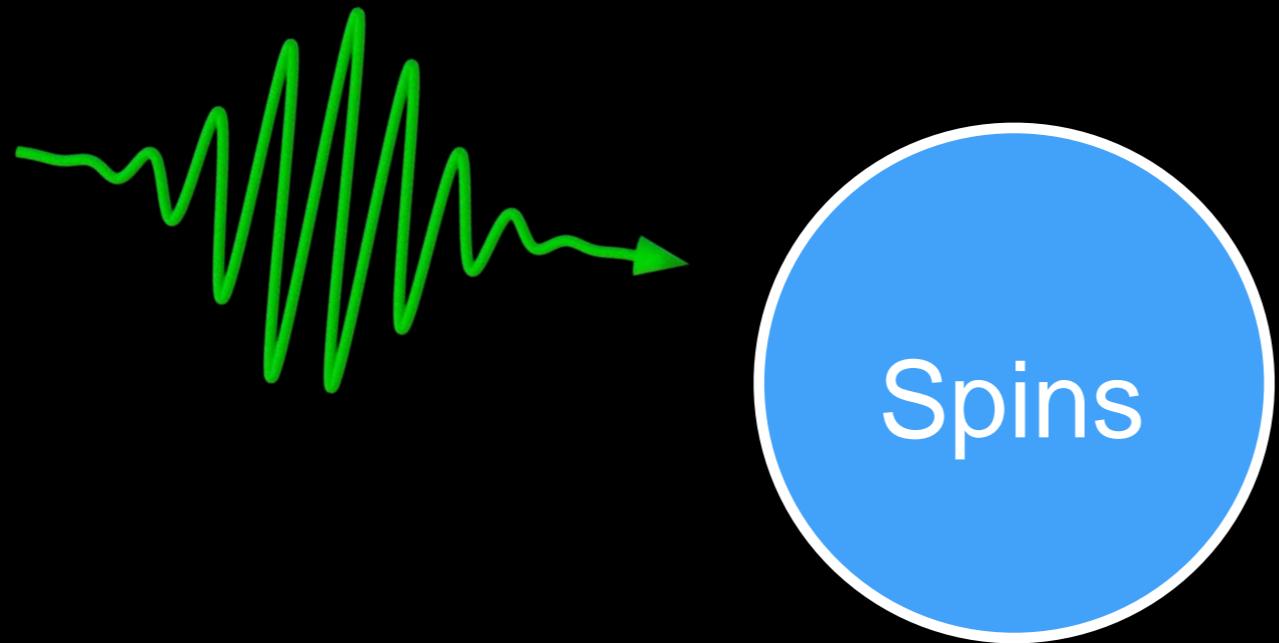


D. Bossini et al. ACS Photonics **3**, 1385 (2016)

# Metallic magnets



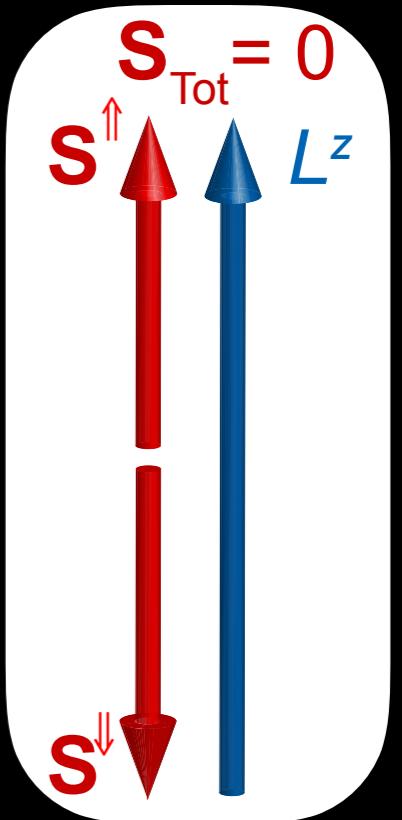
# Dielectric magnets



Spin dynamics ?

# Dielectric antiferromagnet

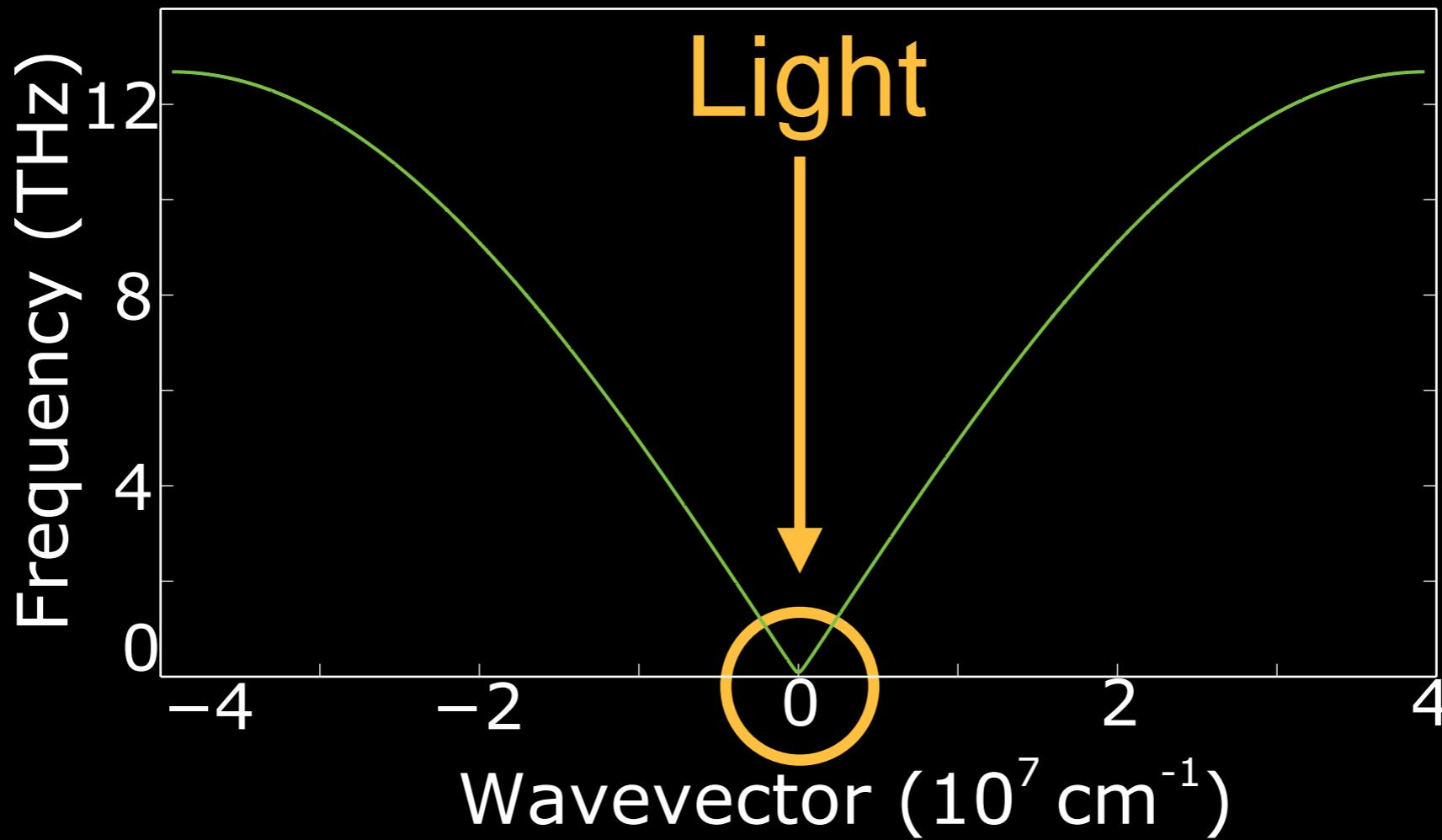
- No free electrons
- Majority of ordered materials
- No stray field
- Intrinsic faster spin dynamics



$$L \equiv S^{\uparrow\uparrow} - S^{\downarrow\downarrow}$$

$$\hat{H} = J \sum_{\langle i,j \rangle} \hat{S}_i^{\uparrow} \cdot \hat{S}_j^{\downarrow} + g\mu_B H_A \left( \sum_i \hat{S}_i^{z\uparrow} - \sum_j \hat{S}_j^{z\downarrow} \right)$$

# Collective spin dynamics



Ferromagnet

$$\omega \propto H_A$$

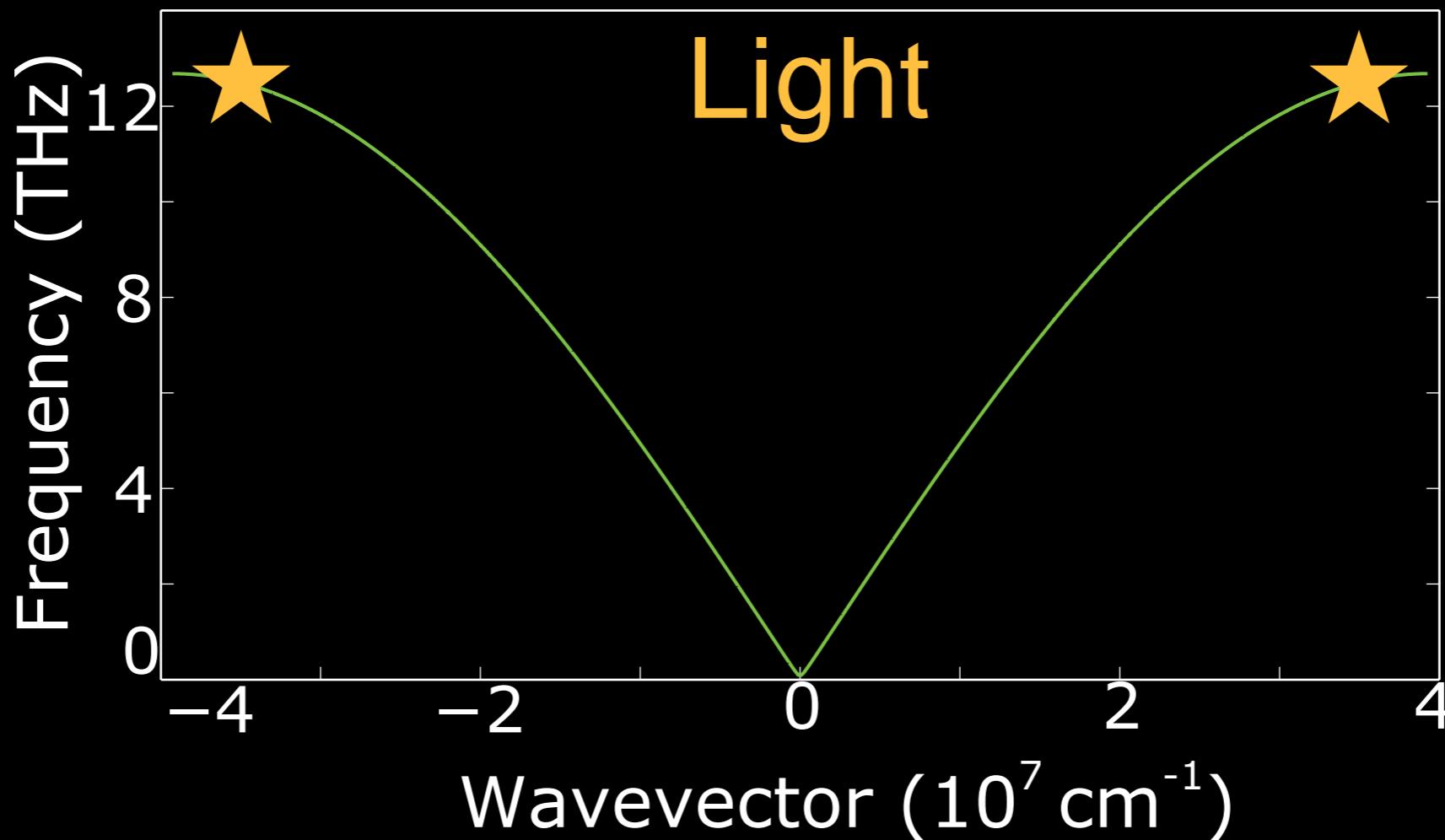
1-10 GHz

Antiferromagnet

$$\omega \propto \sqrt{H_A H_{ex}}$$

0.1-1 THz

# Faster dynamics in AFs



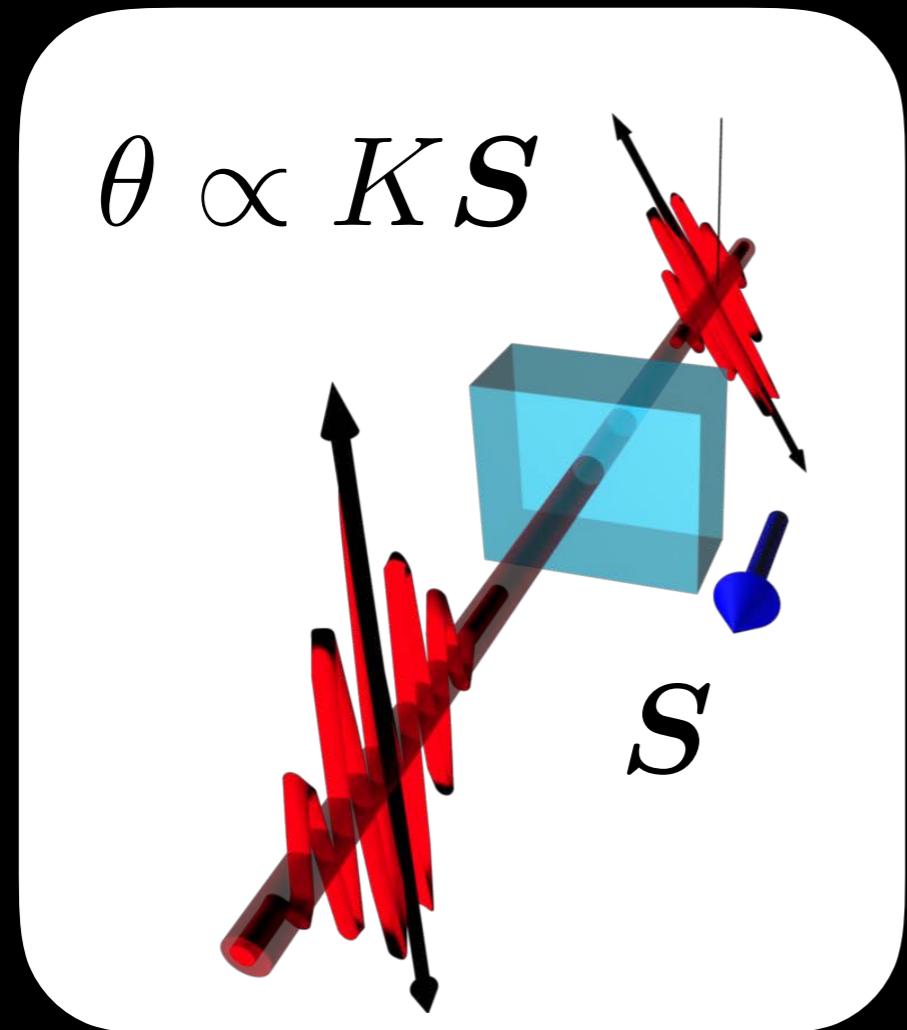
Antiferromagnet

10-100 THz

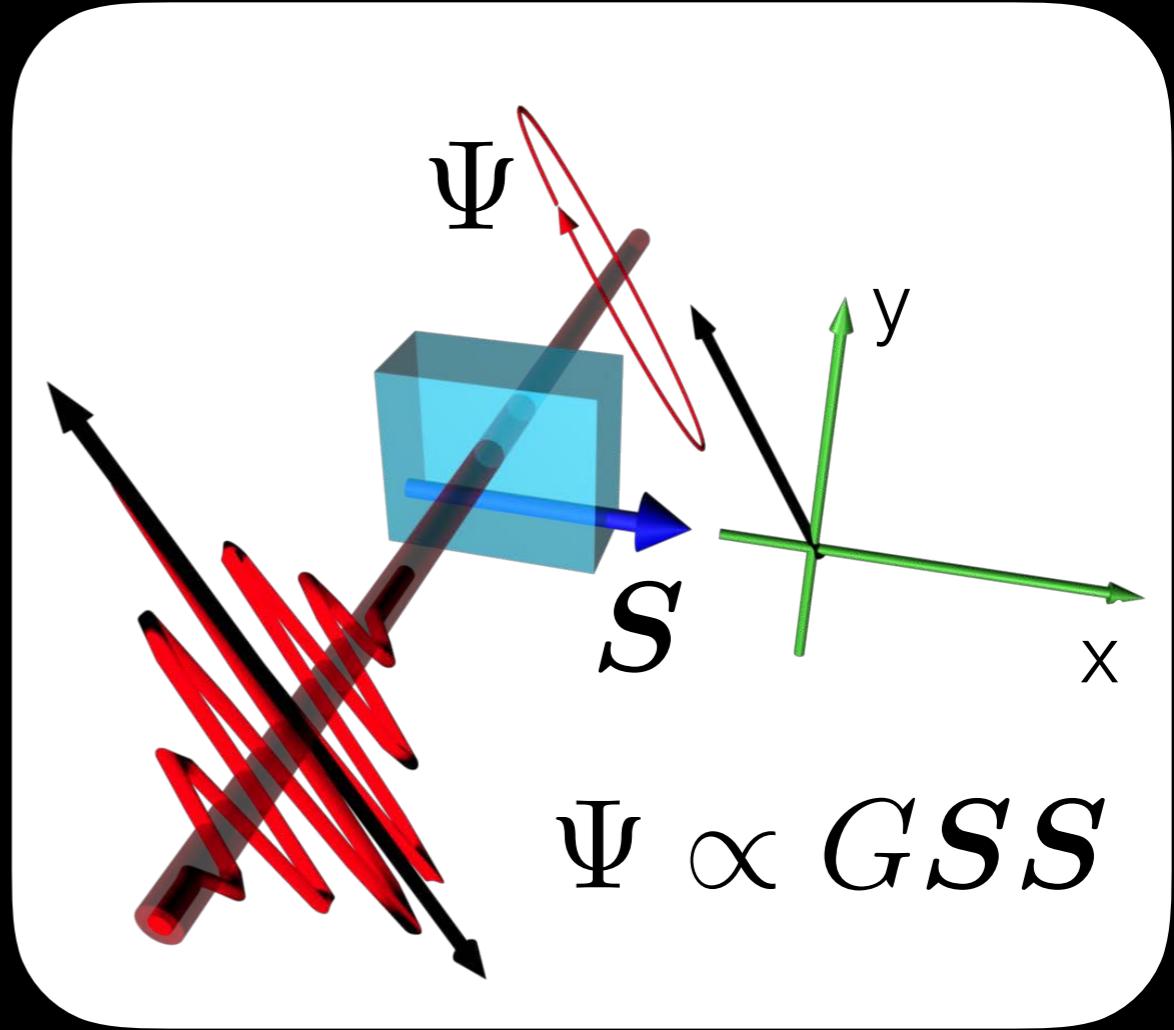
Ferromagnet

1-10 GHz

# Magneto-optical effects



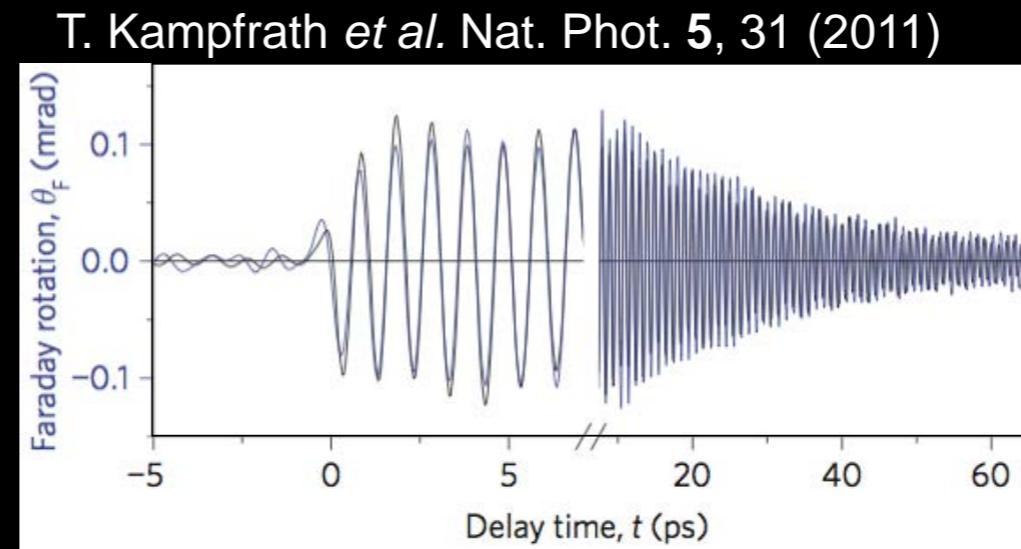
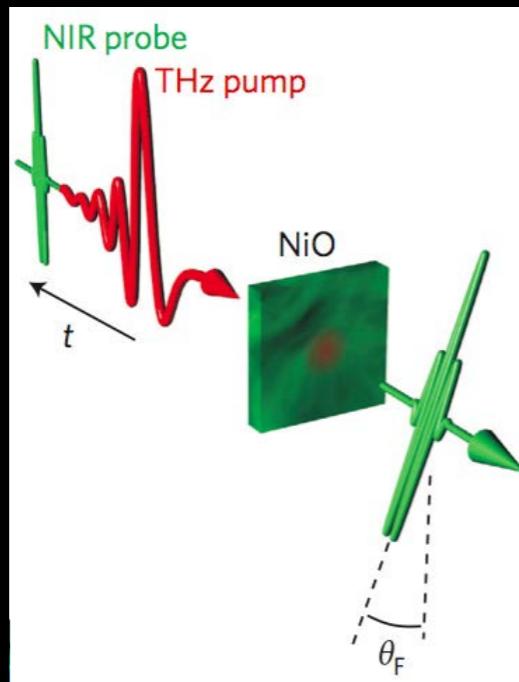
Faraday rotation



Magnetic linear  
birefringence

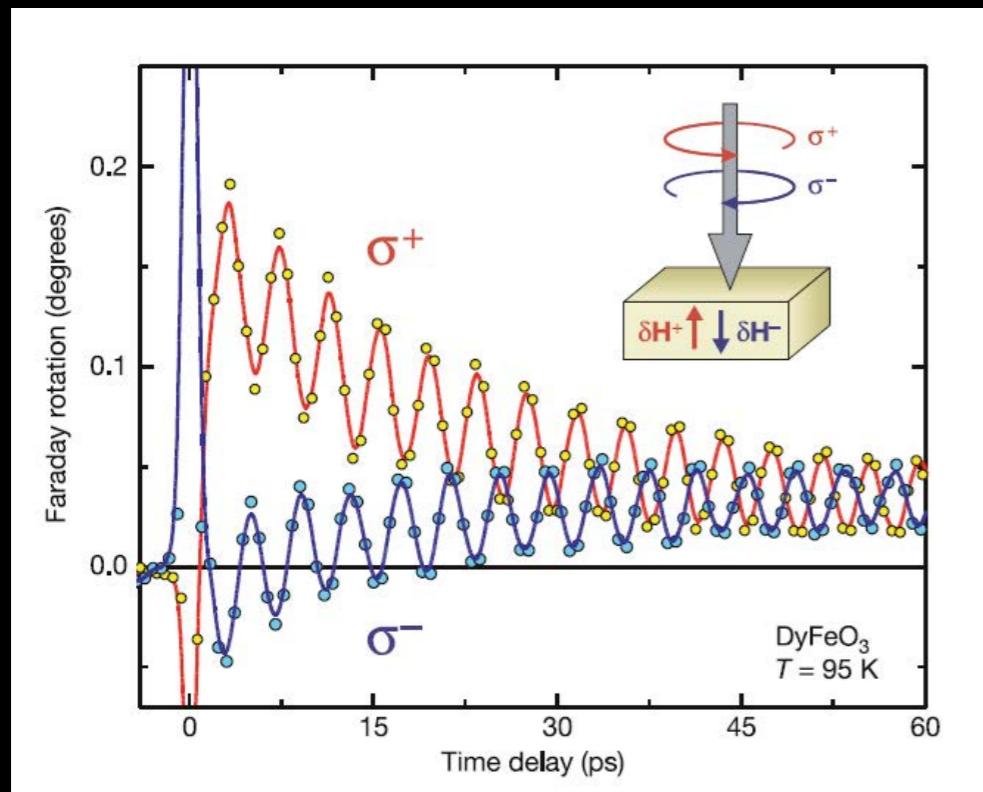
# Ultrafast spin excitation

- ✓ Refieldsgiotndrstineudnasspins
- ✓ Spinsfrapiprcessantovconditio~~N~~ $\Delta$  = 1
- ✓ Stealotfield? Abutio~~H~~?pulsed magnetic field in resonance condition (magnetic dipole transitions)



# Opto-magnetism

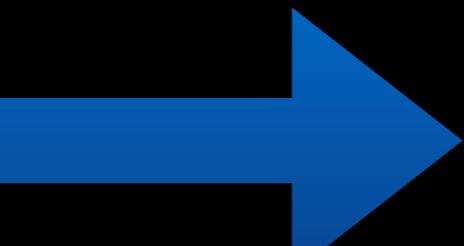
A. Kimel *et al.* Nature 435, 655 (2005)



- ✓ Excitation of coherent spin waves
- ✓ Magneto-optical probe:  
Faraday rotation
- ✓ Pump wavelength: 800 nm

Pump beam  $\approx 10^{14} \text{ Hz}$

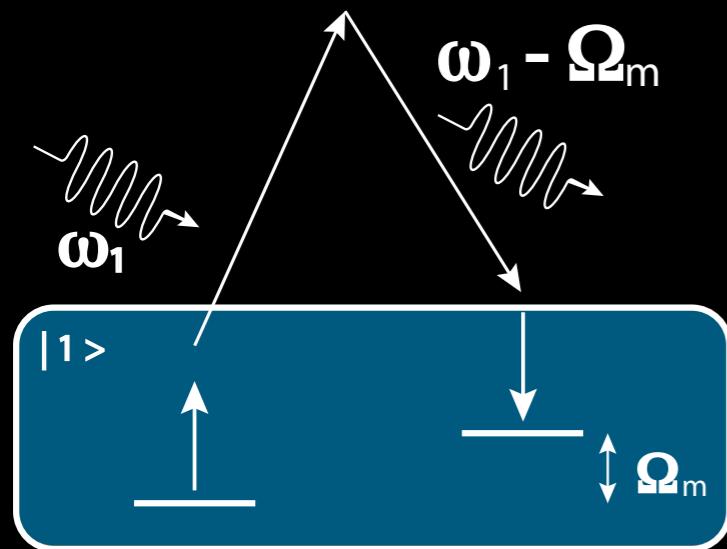
Spin wave  $\approx 10^{11} \text{ Hz}$

 ~~Resonant magnetic dipole coupling?~~

# Raman Scattering

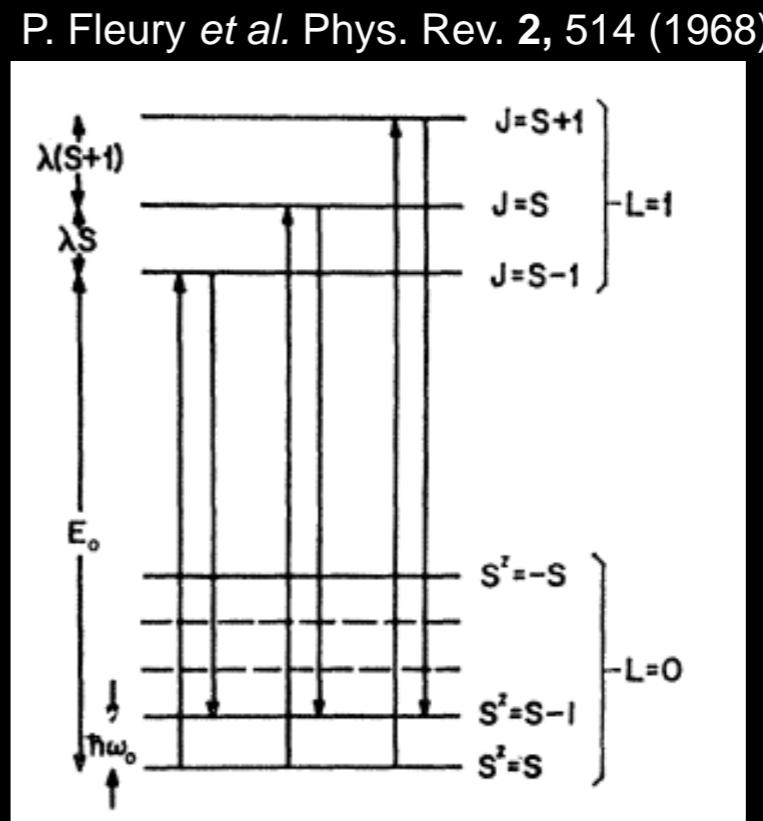
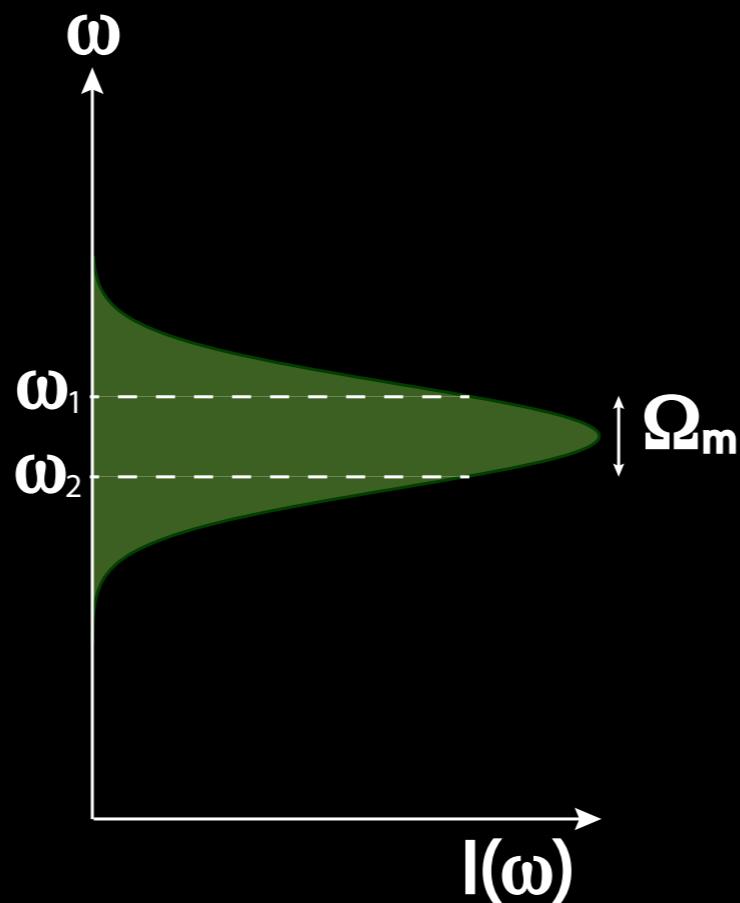
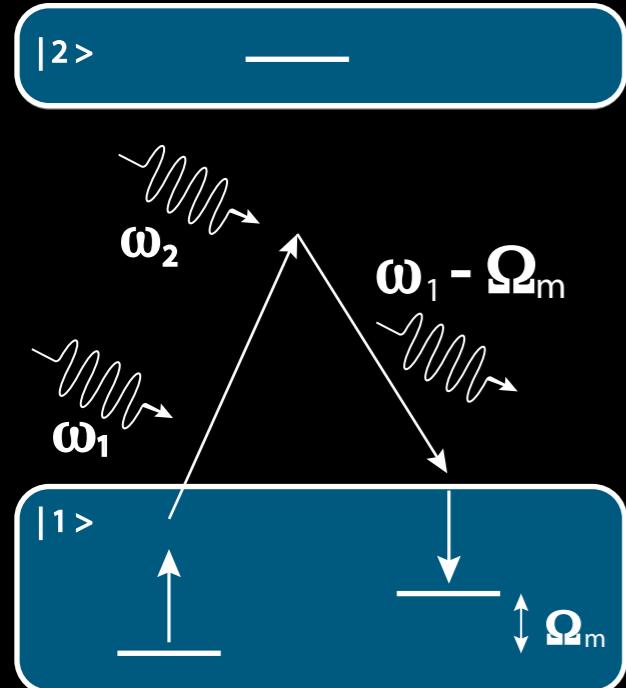
|2>

—



- ✓ Inelastic light scattering
- ✓ Spontaneous process  
low efficiency ( $10^{-7}$ - $10^{-9}$ )

# Impulsive Stimulated Raman Scattering (ISRS)



Raman  
on

Spin-flip driven by L-S coupling in  
the excited state

# Light-spin interaction

- Light scattering: non-dissipative approximation
- Electric dipole approximation

$$\hat{\mathcal{H}} = \sum_{\lambda, \nu} \epsilon^{\lambda \nu}(\hat{S}) E^\lambda E^\nu$$

Expansion dielectric tensor

$$\epsilon^{\lambda \nu} = \sum_i \sum_\gamma K^{\lambda \nu \gamma} \langle \hat{S}_i^\gamma \rangle + \sum_i \sum_{\gamma \delta} G^{\lambda \nu \gamma \delta} \langle \hat{S}_i^\gamma \rangle \langle \hat{S}_i^\delta \rangle + \sum_{i,j} \sum_{\gamma \delta} \rho^{\lambda \nu \gamma \delta} \langle \hat{S}_i^{\gamma \uparrow} \hat{S}_j^{\delta \downarrow} \rangle$$

M. Cottam and D. Lockwood, *Light Scattering in Magnetic Solids* (Wiley-Interscience, 1986)

$$\epsilon^{\lambda\nu} = \sum_i \sum_{\gamma} K^{\lambda\nu\gamma} \langle \hat{S}_i^{\gamma} \rangle + \sum_i \sum_{\gamma\delta} G^{\lambda\nu\gamma\delta} \langle \hat{S}_i^{\gamma} \rangle \langle \hat{S}_i^{\delta} \rangle + \sum_{i,j} \sum_{\gamma\delta} \rho^{\lambda\nu\gamma\delta} \langle \hat{S}_i^{\gamma\uparrow\uparrow} \hat{S}_j^{\delta\downarrow\downarrow} \rangle$$

## Raman scattering on magnons

Opto-magnetism

Magneto-optics

Non-dissipative light-matter  
interaction

# General concept

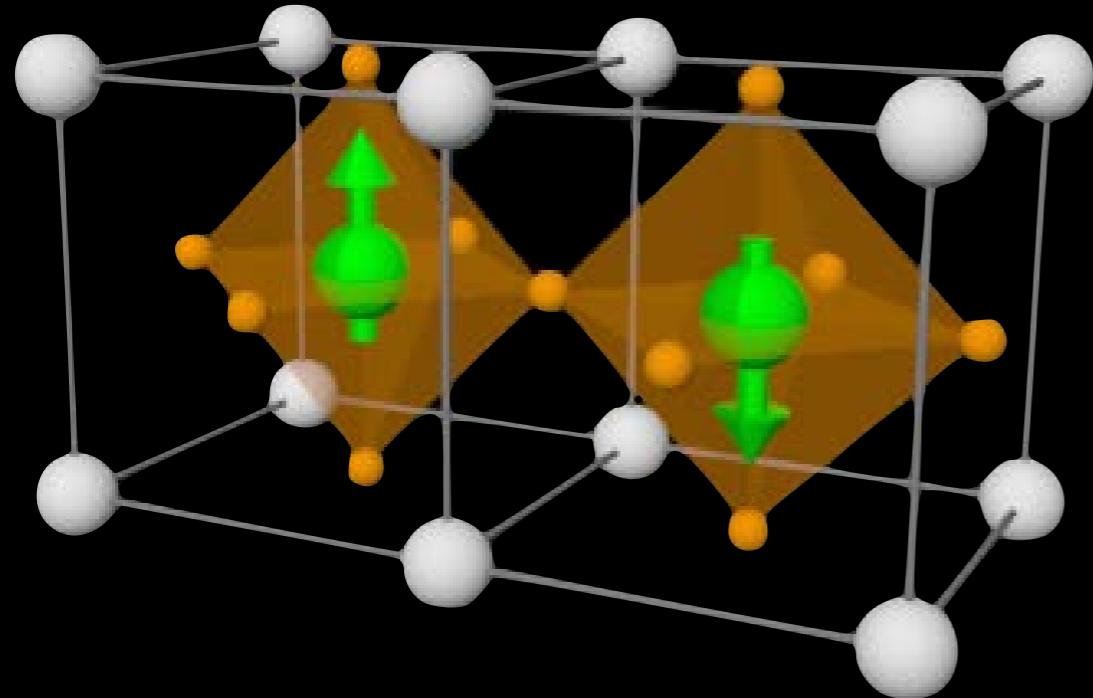


✓ Is magnetization necessary ?

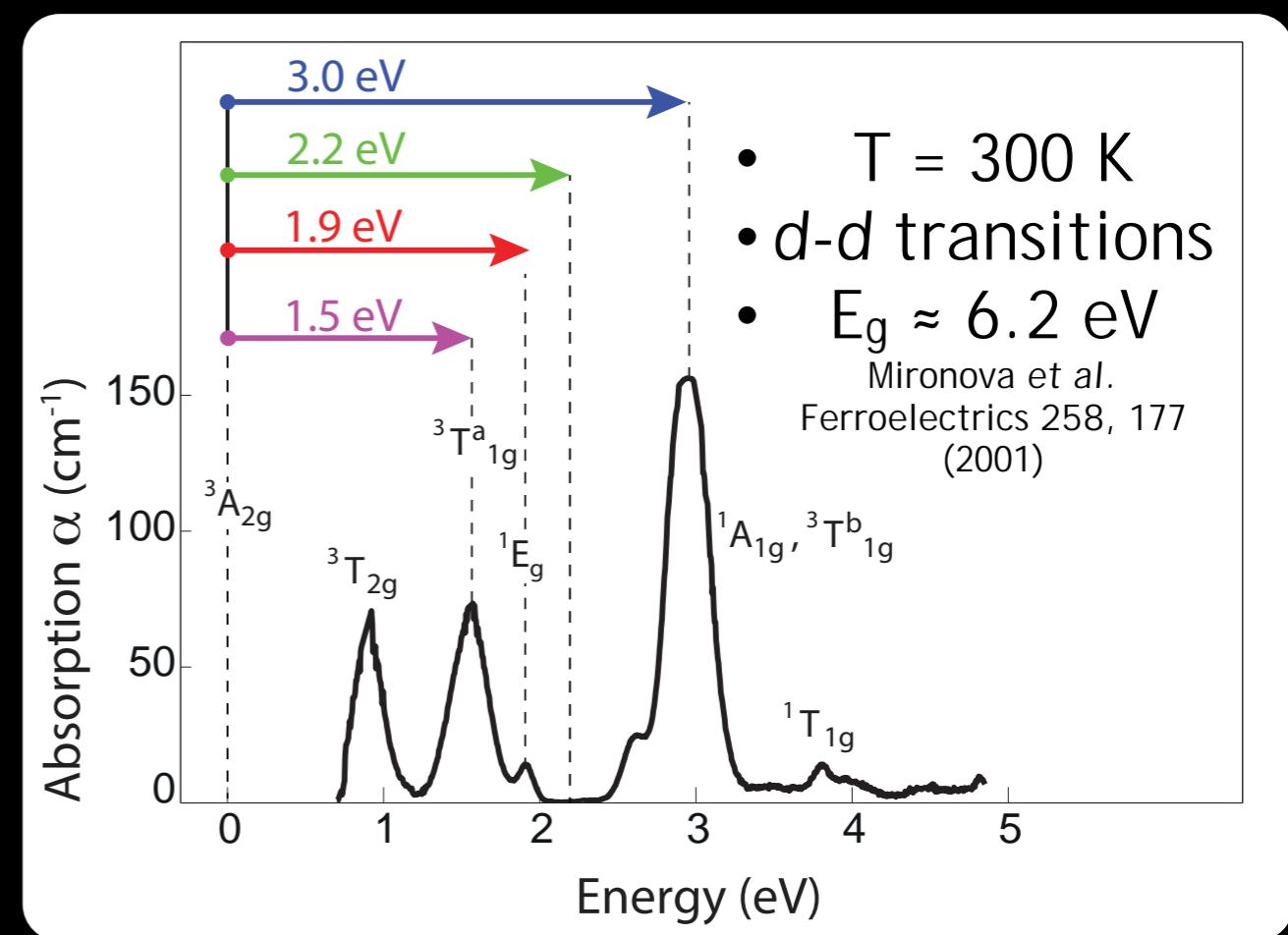
- ✓ Unavoidable direct laser-induced thermal load
- ✓ Giant spin-orbit coupling in the ground state required
- ✓ Limited to low-frequencies

# Opto-magnetism without absorption

Sample:  $\text{KNiF}_3$



Cubic Heisenberg AF  
( $T_N = 246 \text{ K}$ )



- $T = 300 \text{ K}$
- $d$ - $d$  transitions
- $E_g \approx 6.2 \text{ eV}$

Mironova et al.  
Ferroelectrics 258, 177  
(2001)

# Magnetic linear birefringence

**Dynamics:**  $\Delta M \propto \gamma L \times \frac{\partial L}{\partial t}$

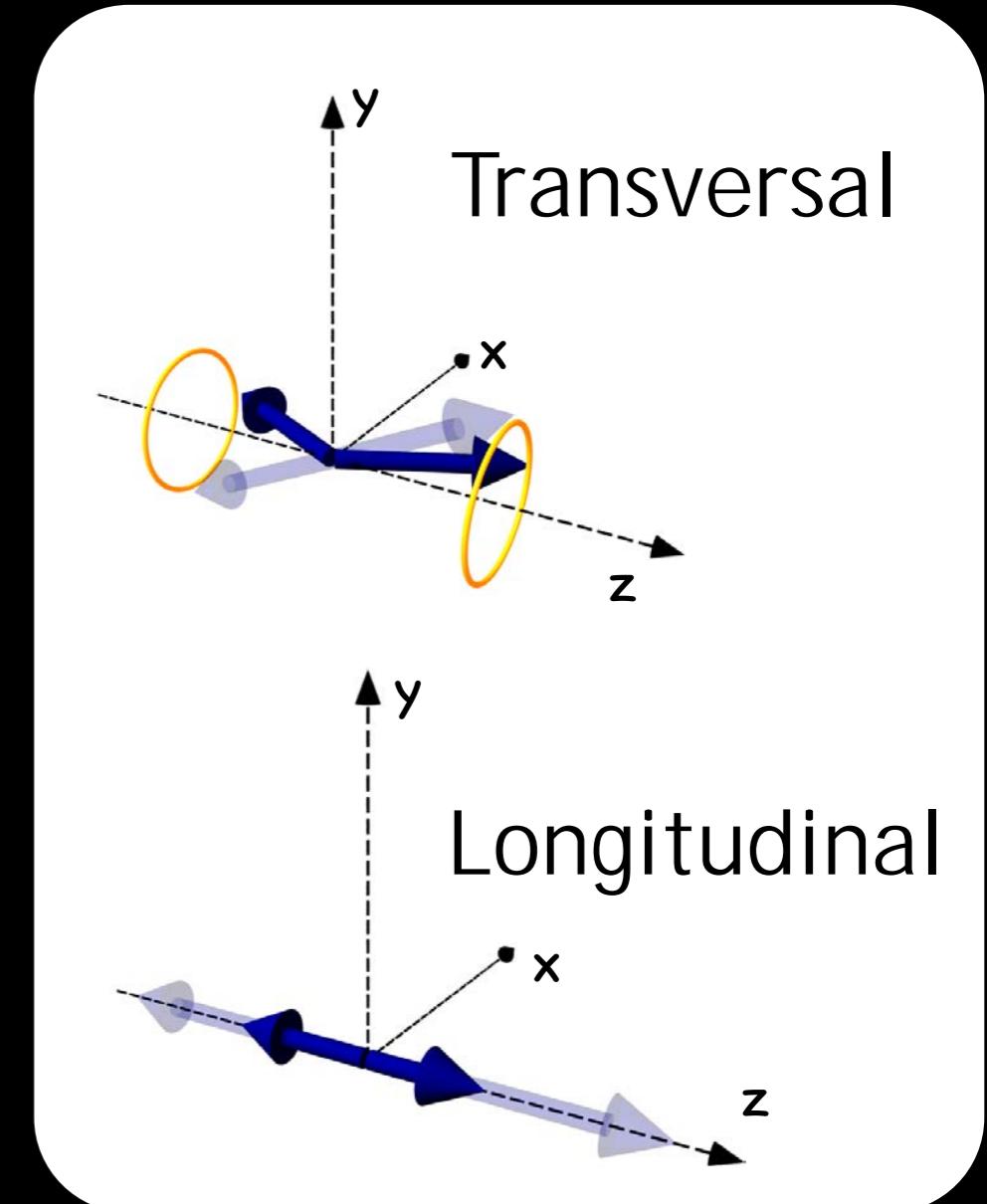
A.F Andreev et al Sov. Phys. Usp. **23**, 21 (1980)

Quadratic MO effect:  $\propto L_z L_y + L_z^2$

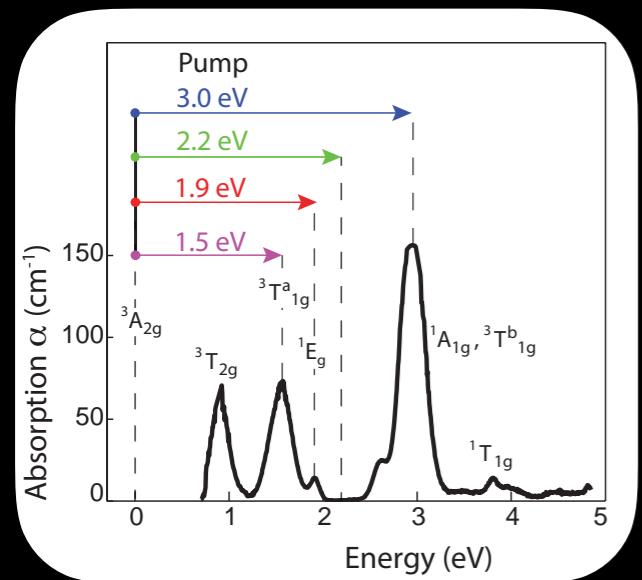
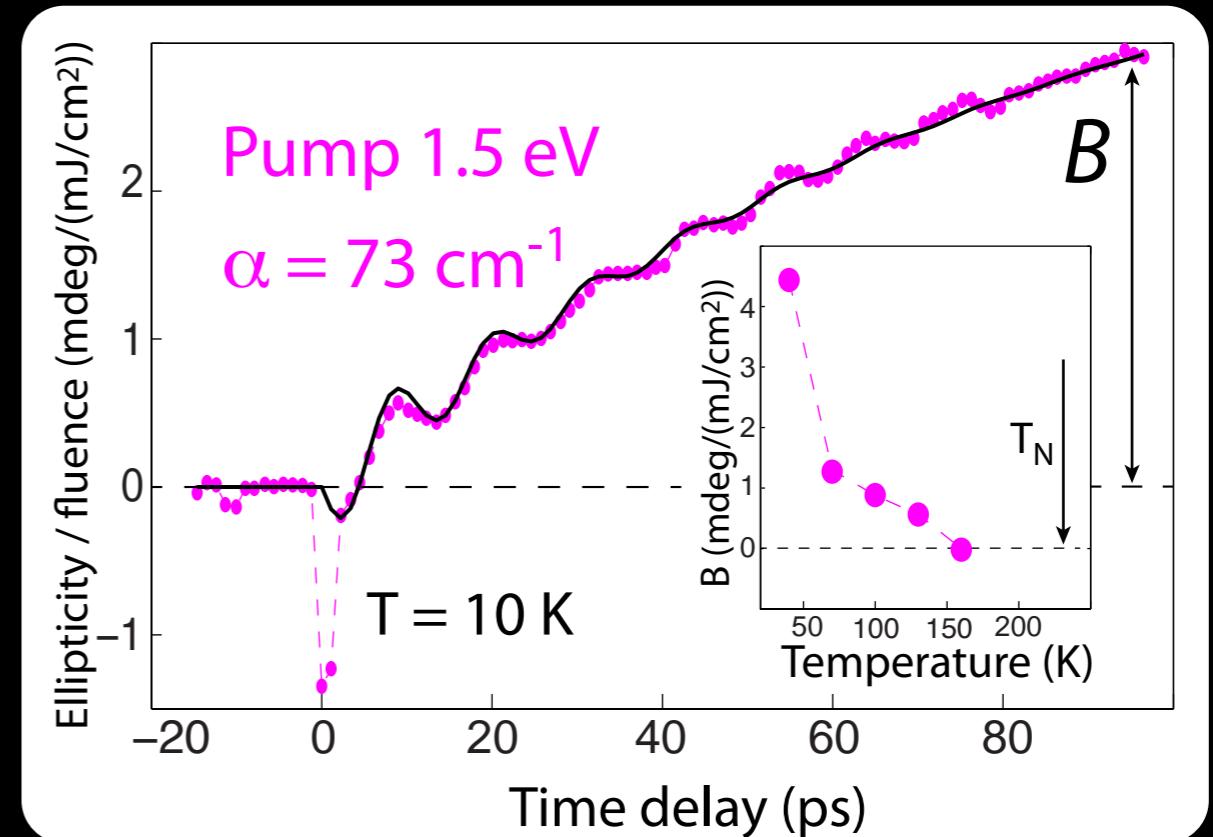
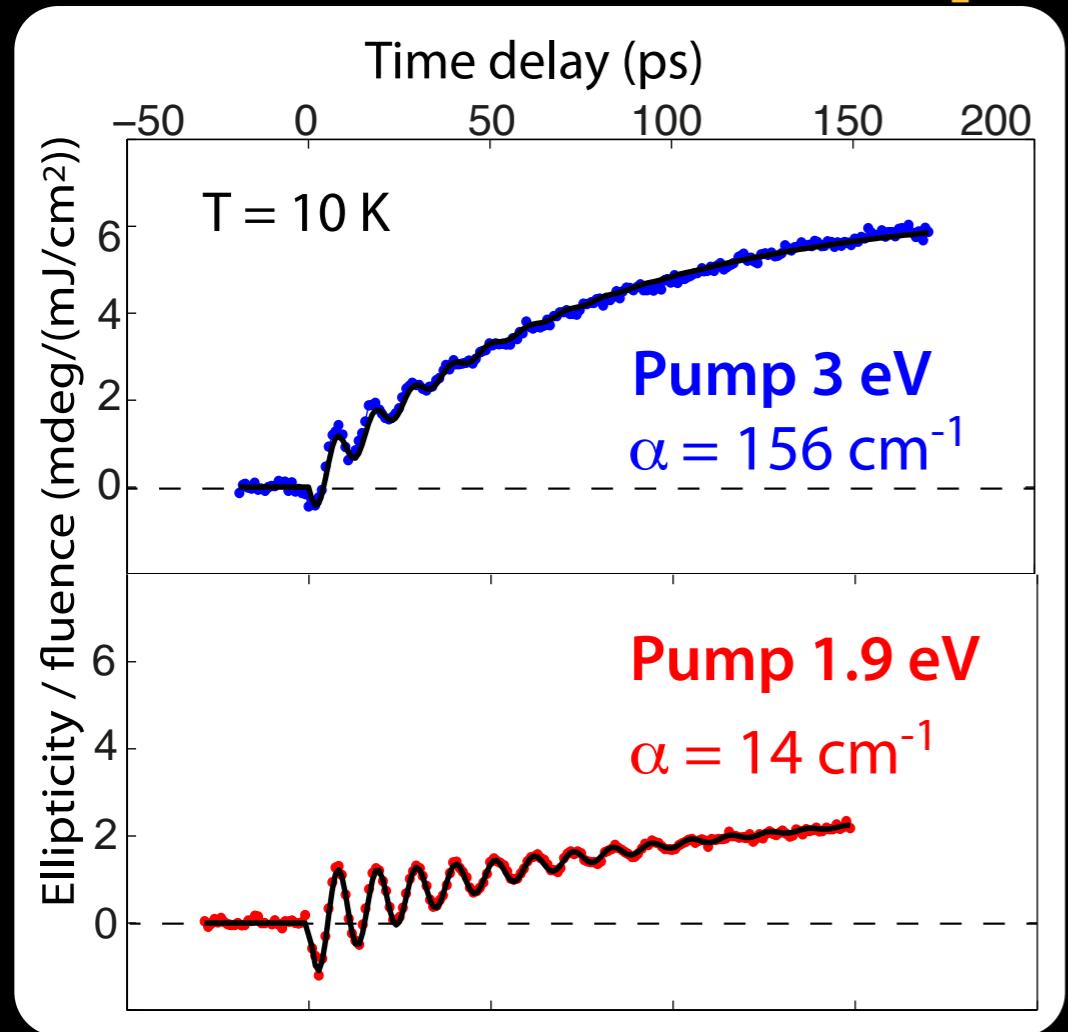
J. Ferre and G. Gehring, Rep. Prog. Phys. **47**, 513 (1984)

$$\Delta \text{MLB} \propto L_z \Delta L_y + L_z \Delta L_z$$

Simultaneous measurement of  
**transversal and**  
**longitudinal** spin dynamics



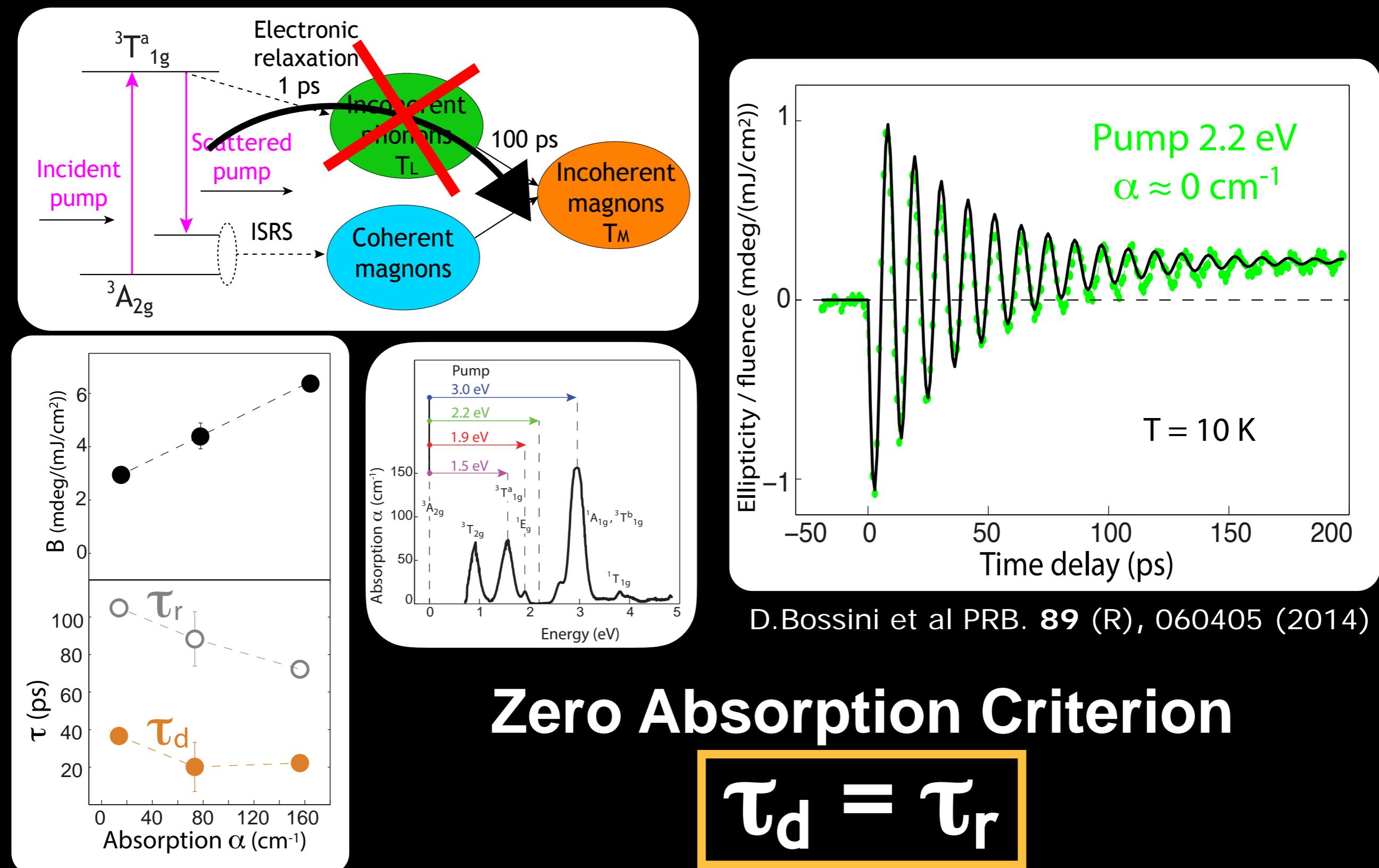
# Dissipative regime

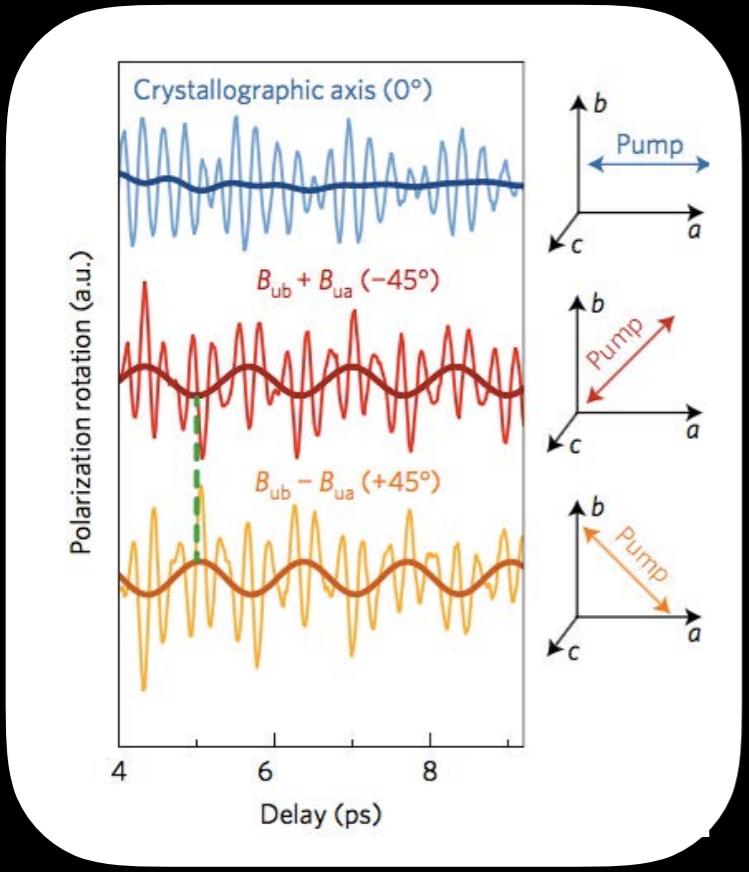


$$f(t) = \theta(t)[A \sin(2\pi\nu t)e^{(-t/\tau_d)} + B(1 - e^{(-t/\tau_r)})]$$

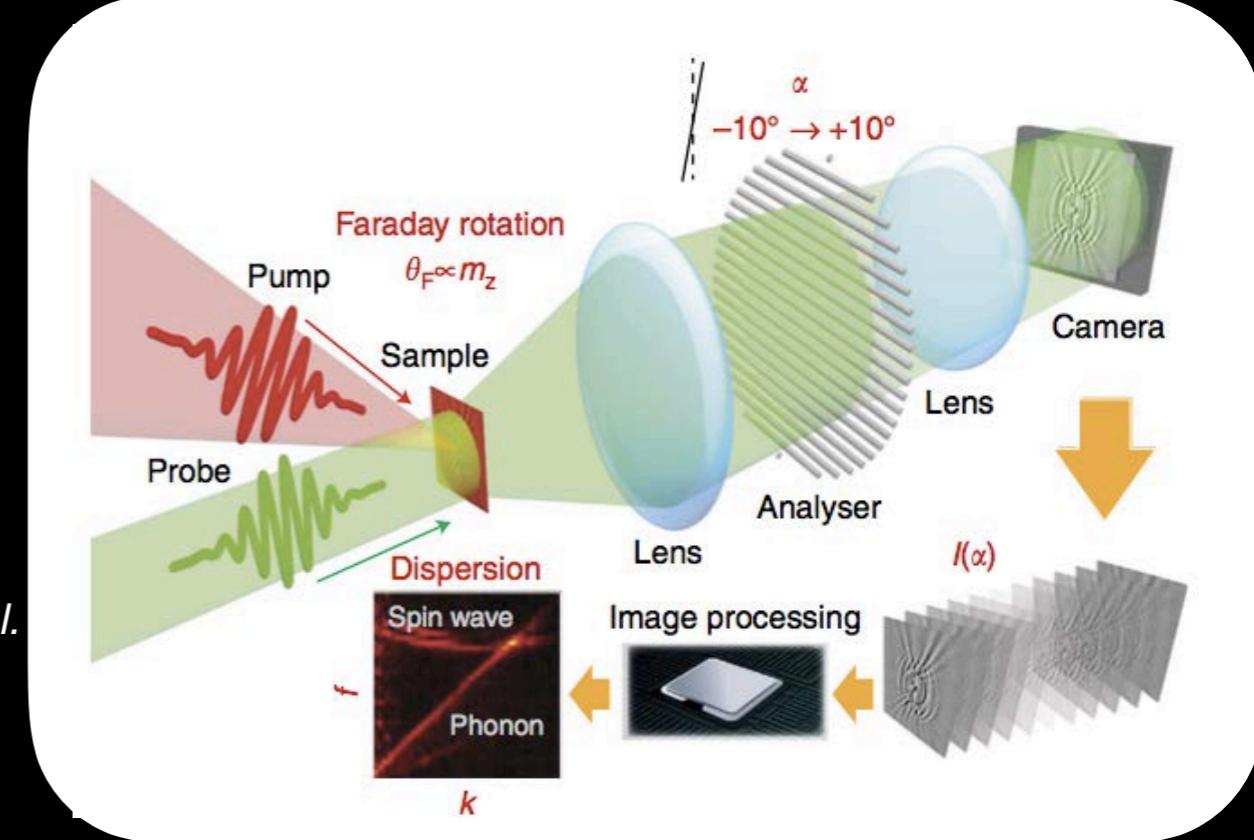
Transversal      Longitudinal  
**Two time-scales**

# Non-dissipative regime

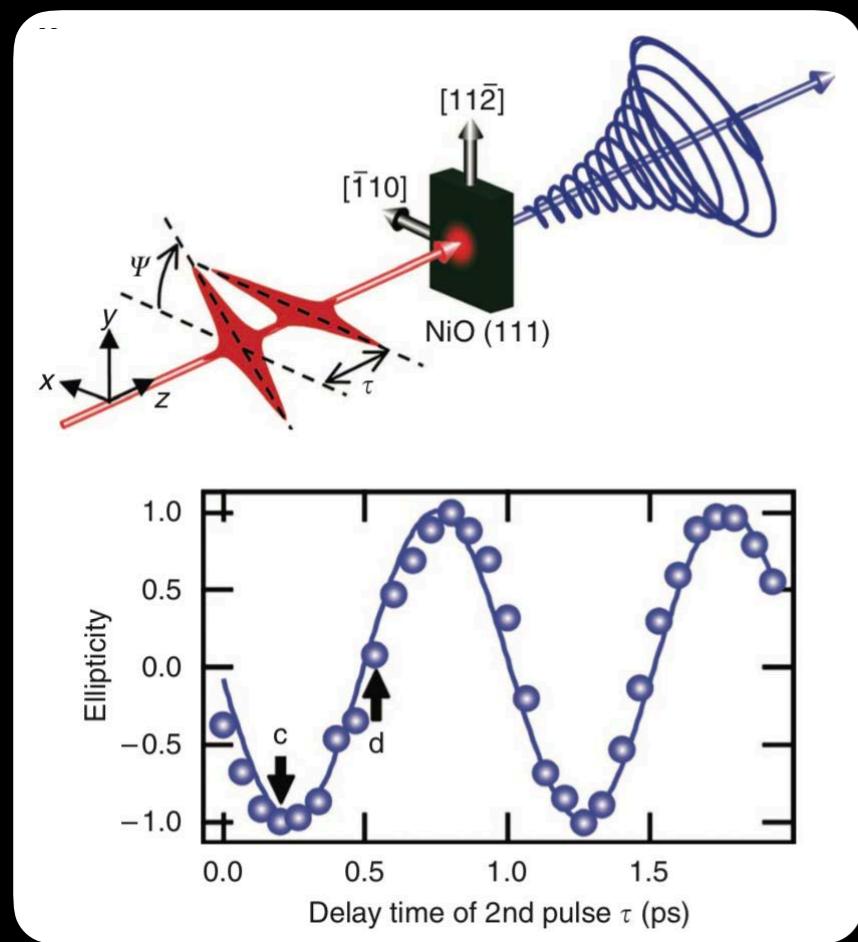




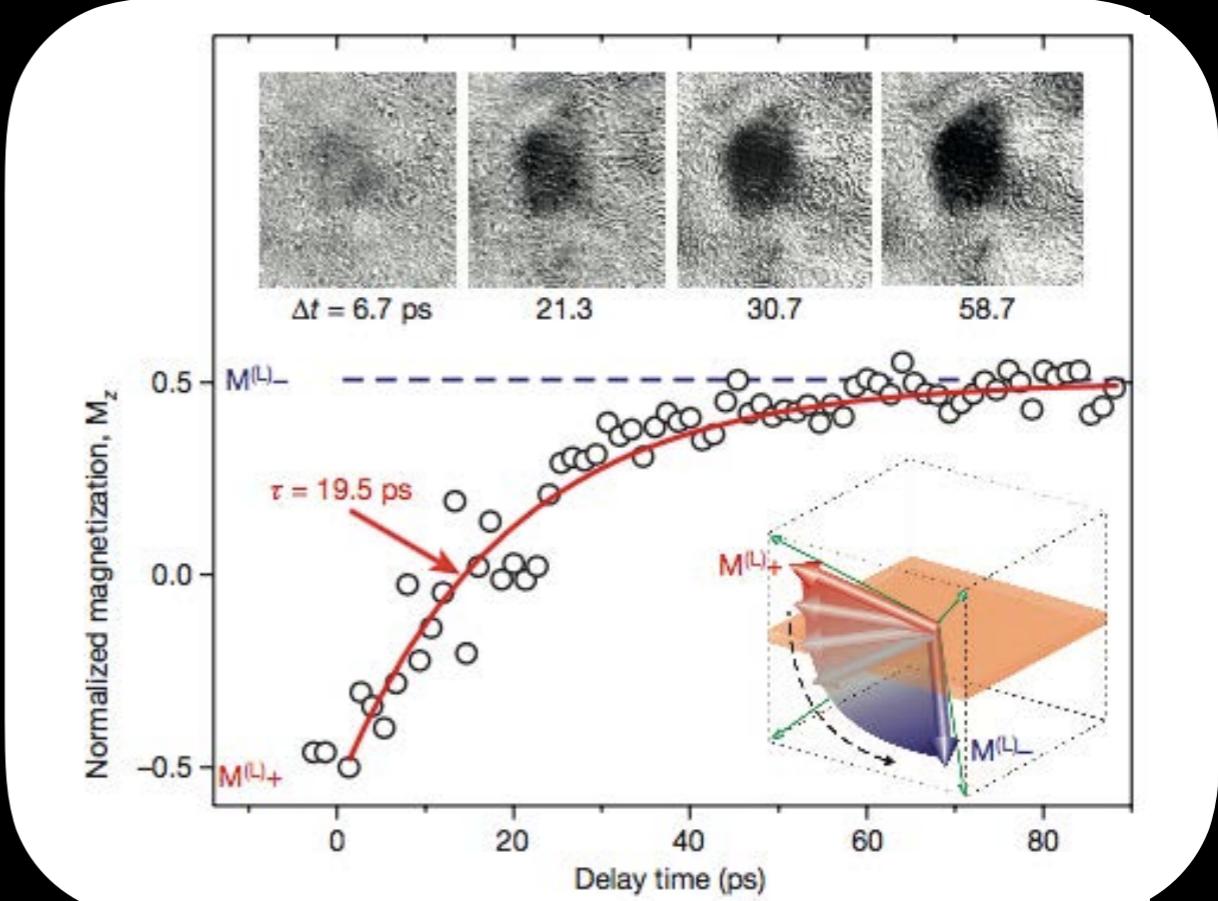
T. Nova, DB et al.  
Nat. Phys. **13**, 132 (2016)



Y. Hashimoto, DB et al.  
Nat. Comm. **8**, 15859  
(2017)



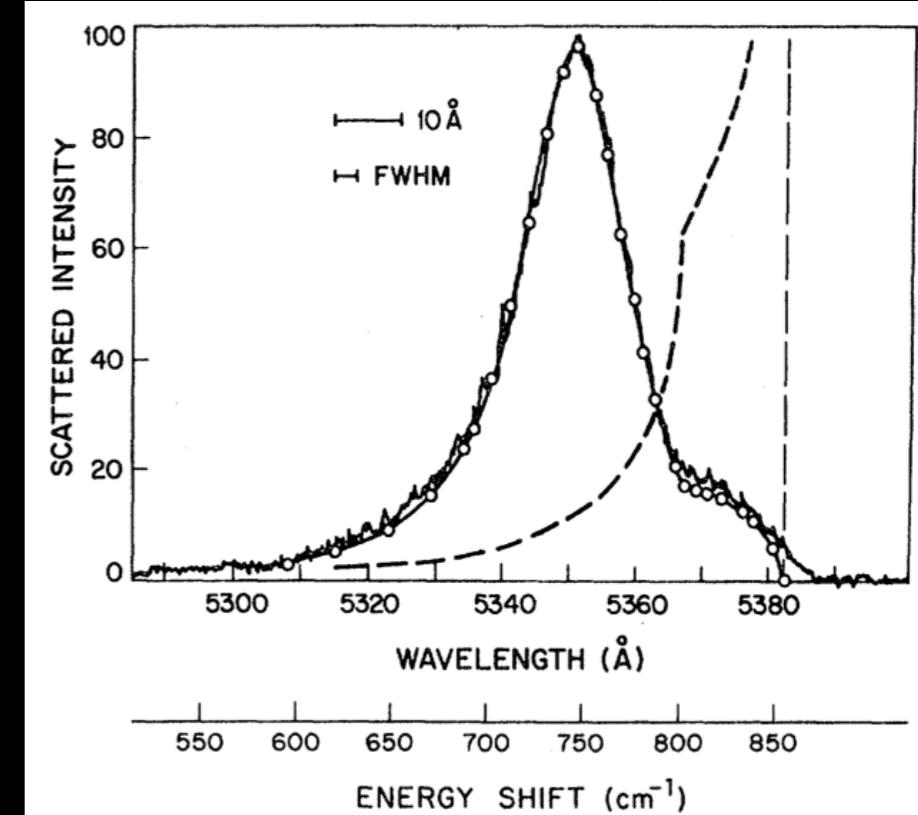
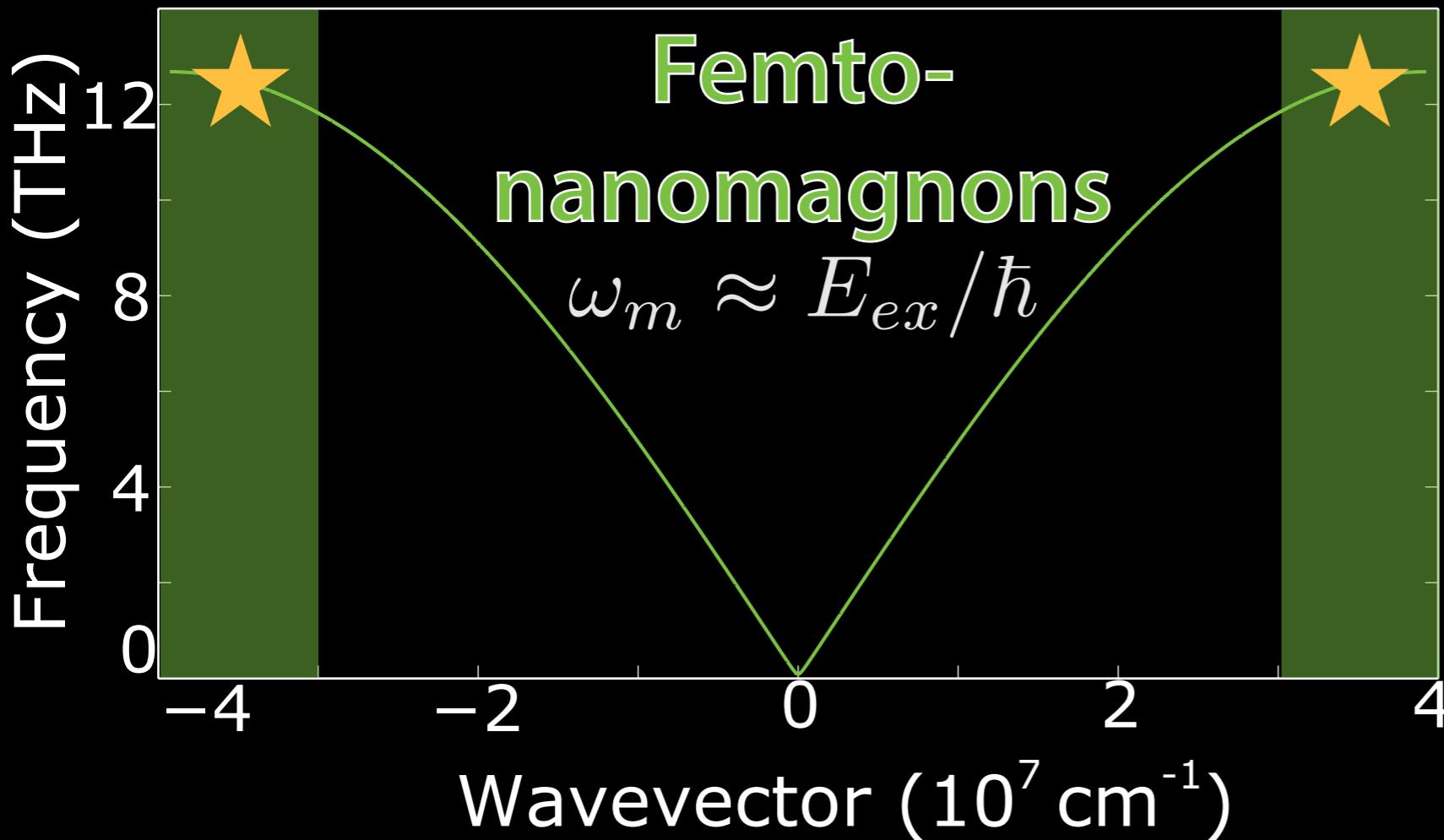
N. Kanda et al. Nat. Comm. **2**, 362 (2011)



A. Stupakiewicz et al. Nature **542**, 71 (2017)

# Shortest spin timescale

S. Chinn et al. PRB 3, 1709



- Light-induced magnon pairs: ***two-magnon mode***

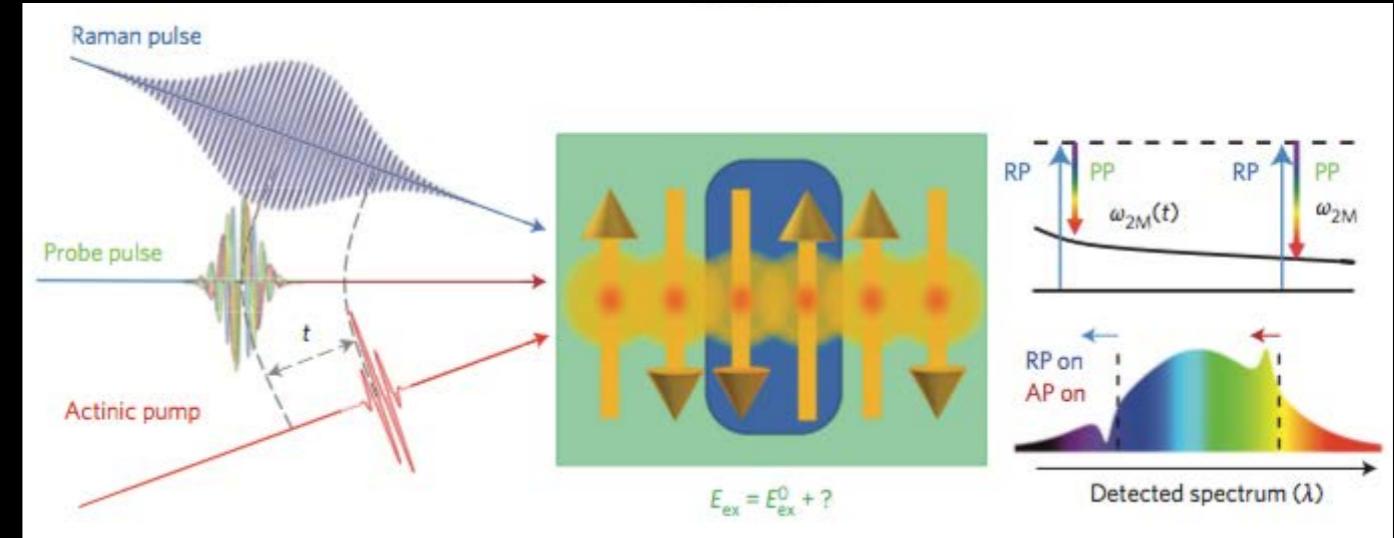
•  $\mathbf{k}_{\text{light}} = \mathbf{k}_{\text{High wavevector region}} + \mathbf{k}_{\text{DOS}} + \omega_2$

Spin and momentum conservation

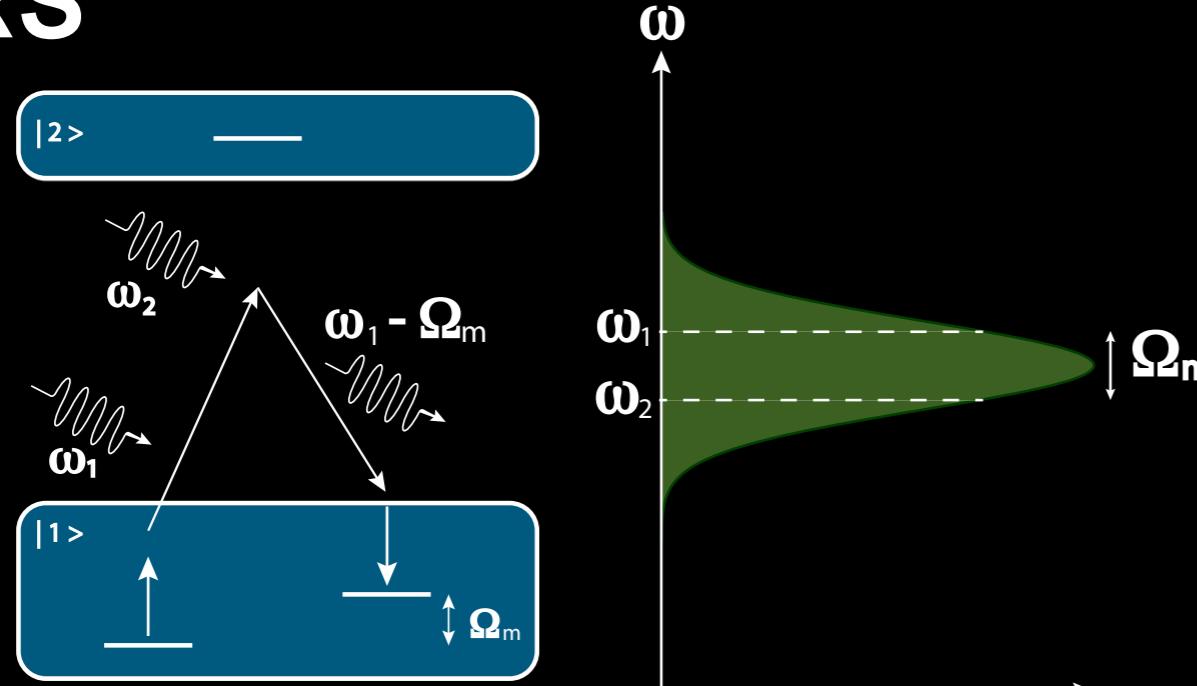
# Experimental approaches

G. Batignagni, DB, et al Nat. Phot. 9, 506 (2015)

## Time-resolved stimulated Raman spectroscopy



## ISRS



2M period in KNiF<sub>3</sub>: 45 fs

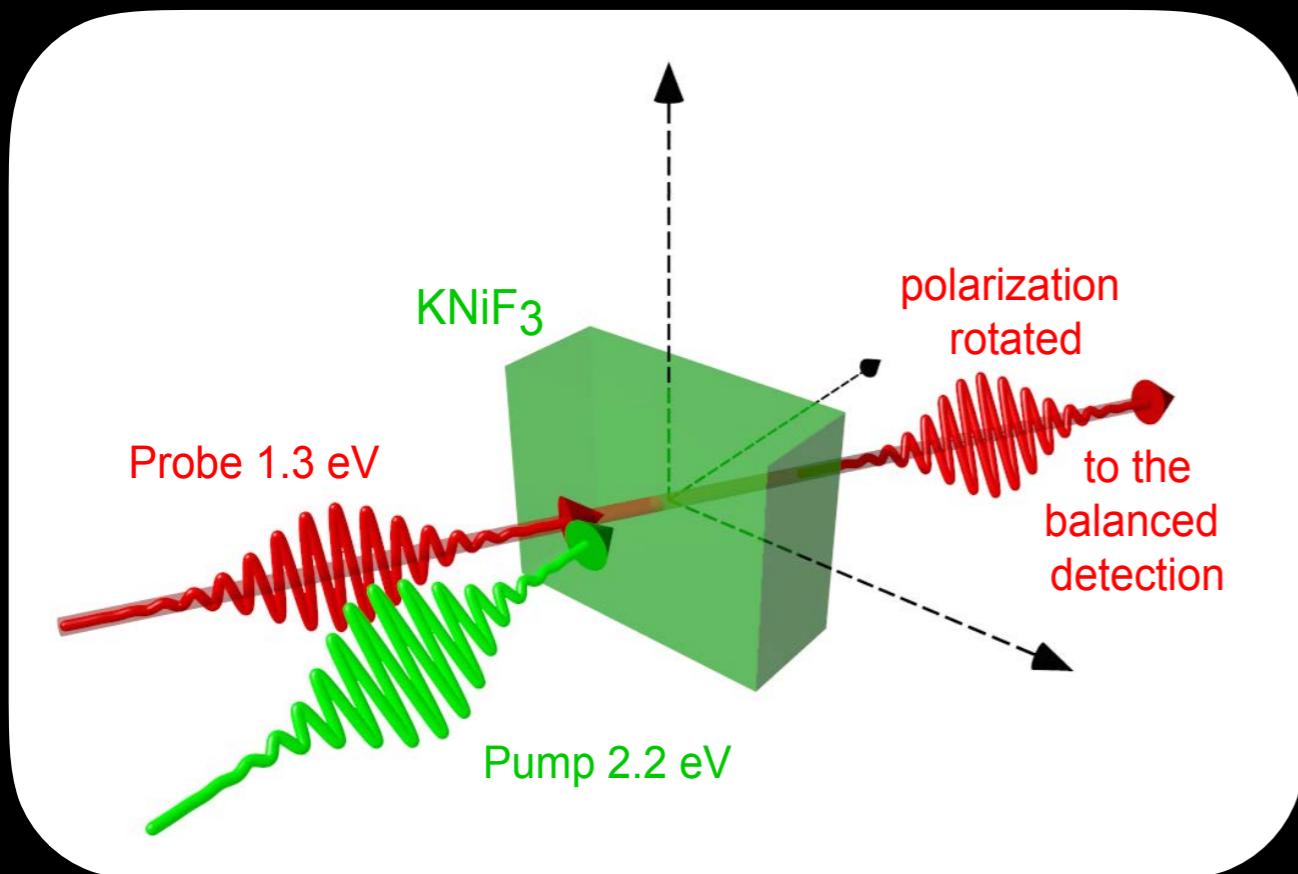
Time domain

Pulses  
shorter than  
period

10 fs

laser pulses

# Experimental scheme



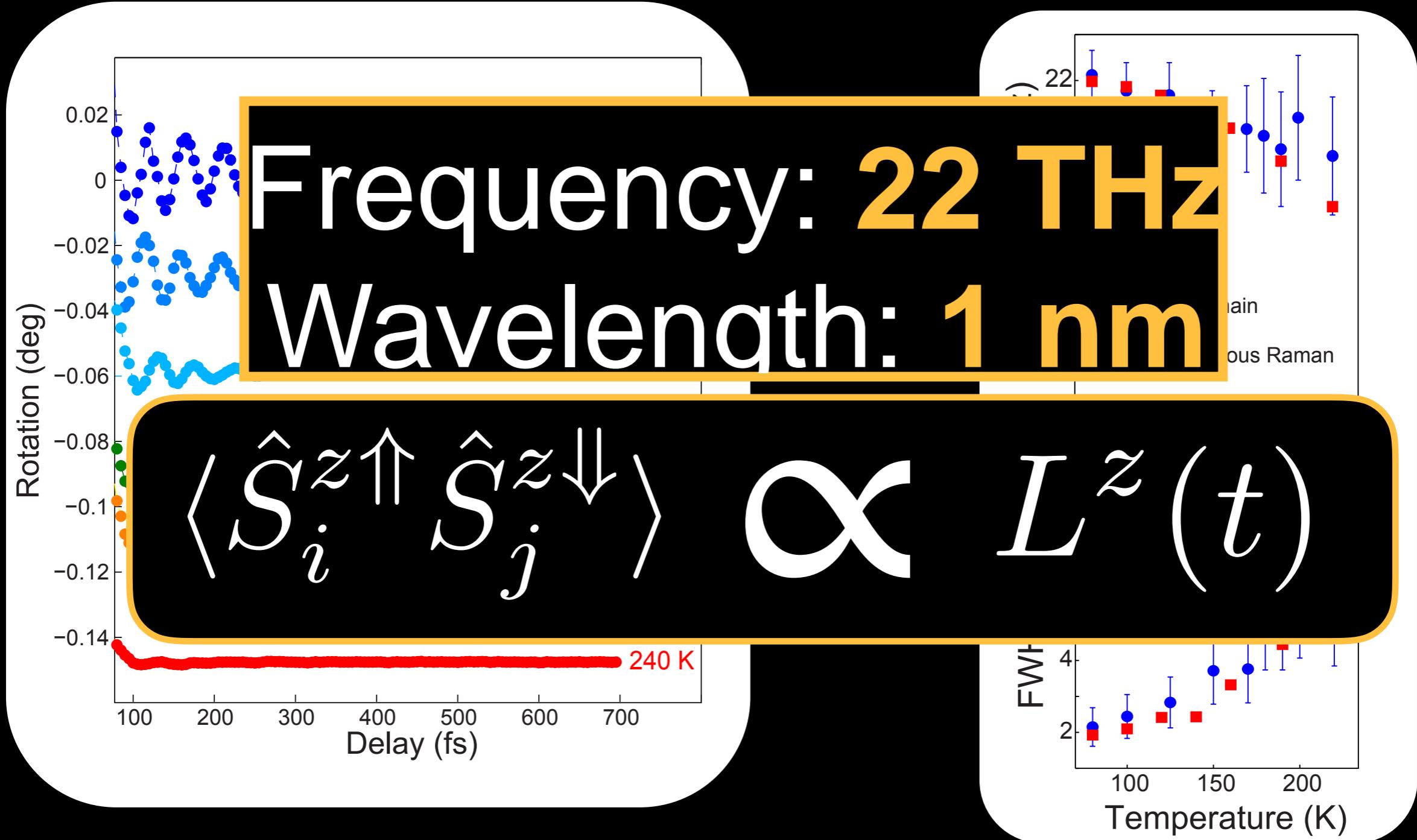
**Low-dissipations:**  
Photon energies in  
transparency regions

All-optical detection via  
**second-order**  
magneto-optics

$$\epsilon_s^{\lambda\nu} = \sum_{ij} \sum_{\gamma\delta} \rho^{\lambda\nu\gamma\delta} \langle \hat{S}_i^{\gamma\uparrow} \hat{S}_j^{\delta\downarrow} \rangle$$

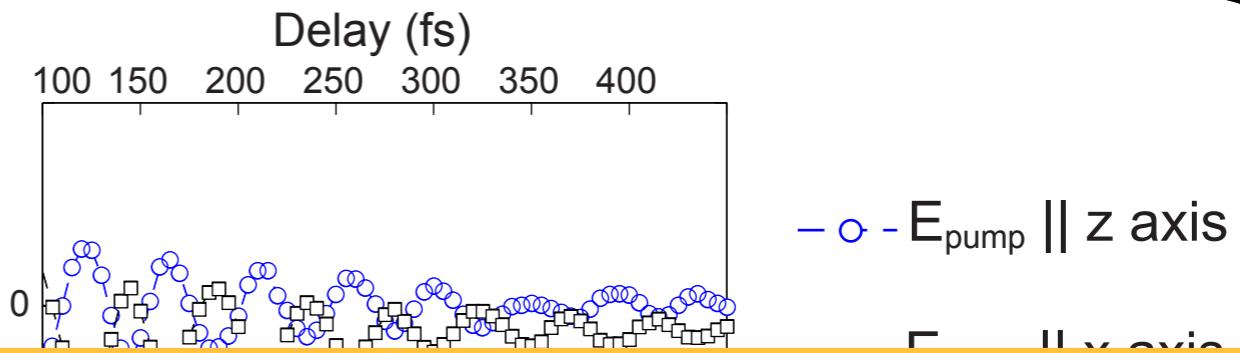
J. Ferrè *et al.* Rep. Prg. Phys 47, 513 (1984)

# Results

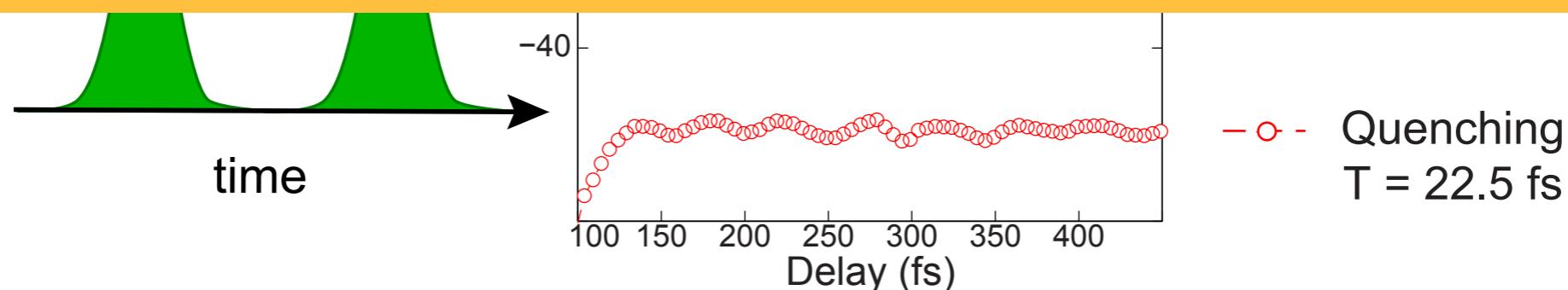


# Manipulate the dynamics

One pump  
pulse

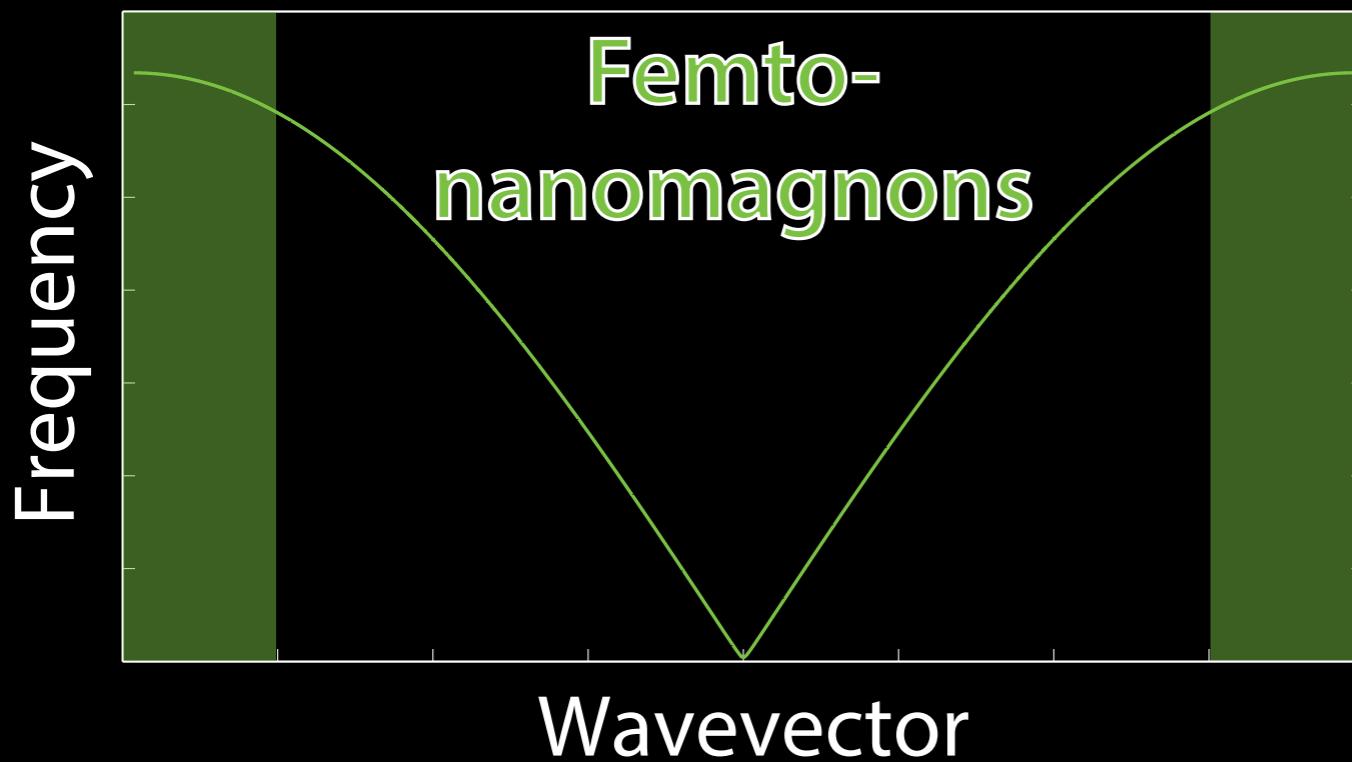


Coherent femtosecond  
manipulation of



D. Bossini et al. Nat. Comm. 7, 10645 (2016)

# Magnon pumping



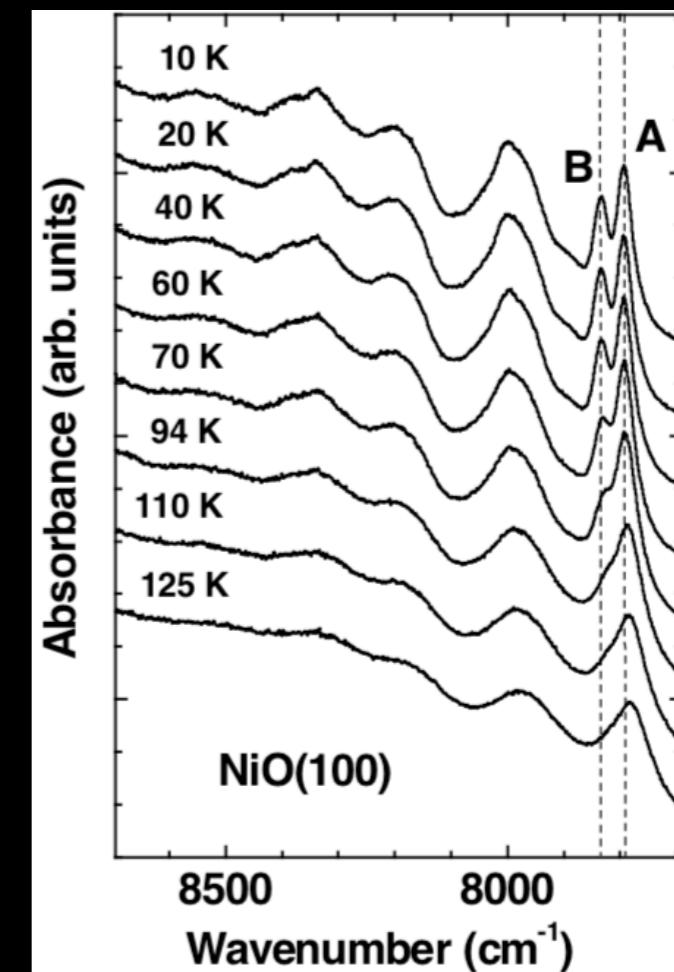
**Exciton-Magnon**

Tanabe et al. *Excitons in Magnetic Insulators* (1982)  
Moriya et al. PRL 15, 1023 (1965)

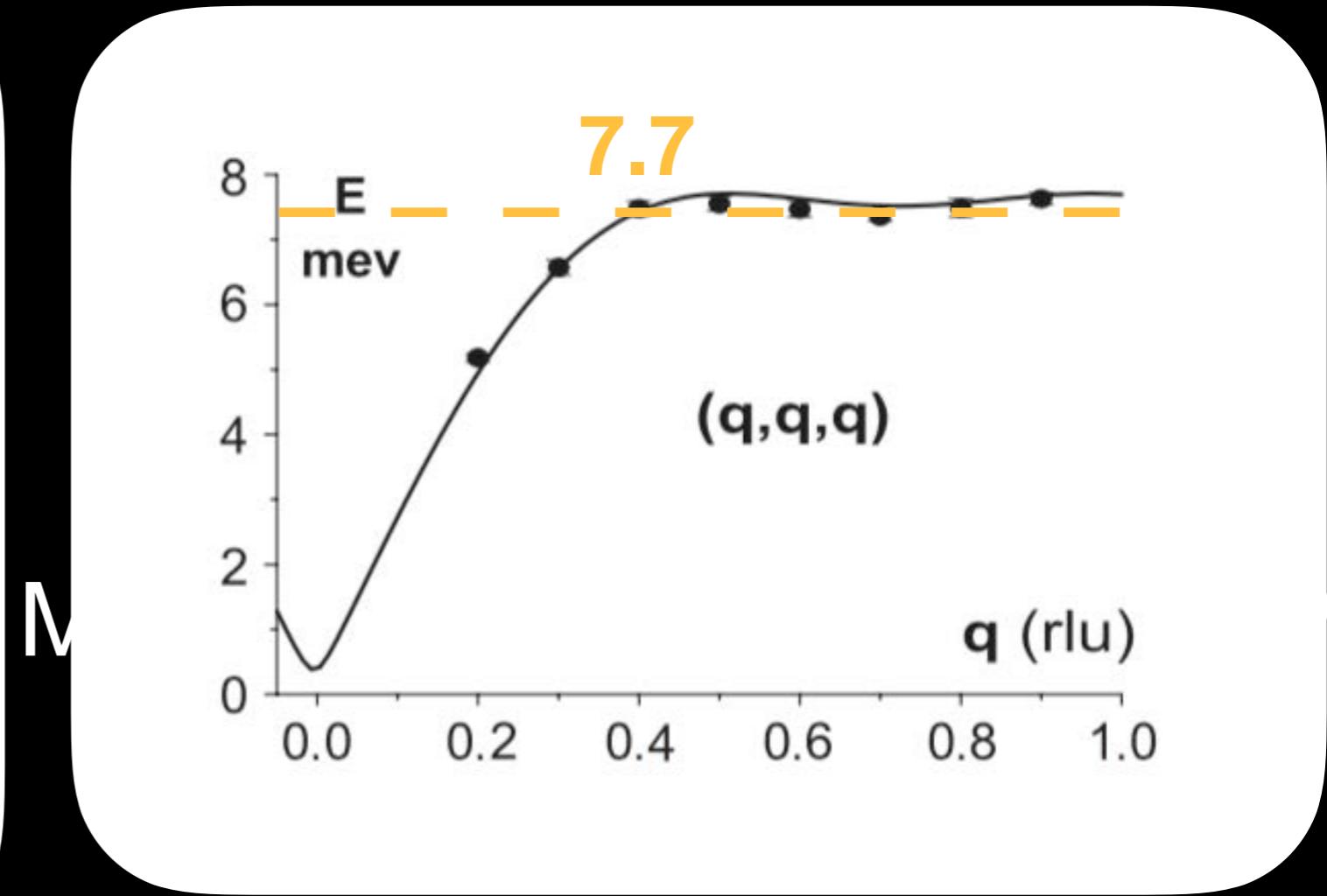
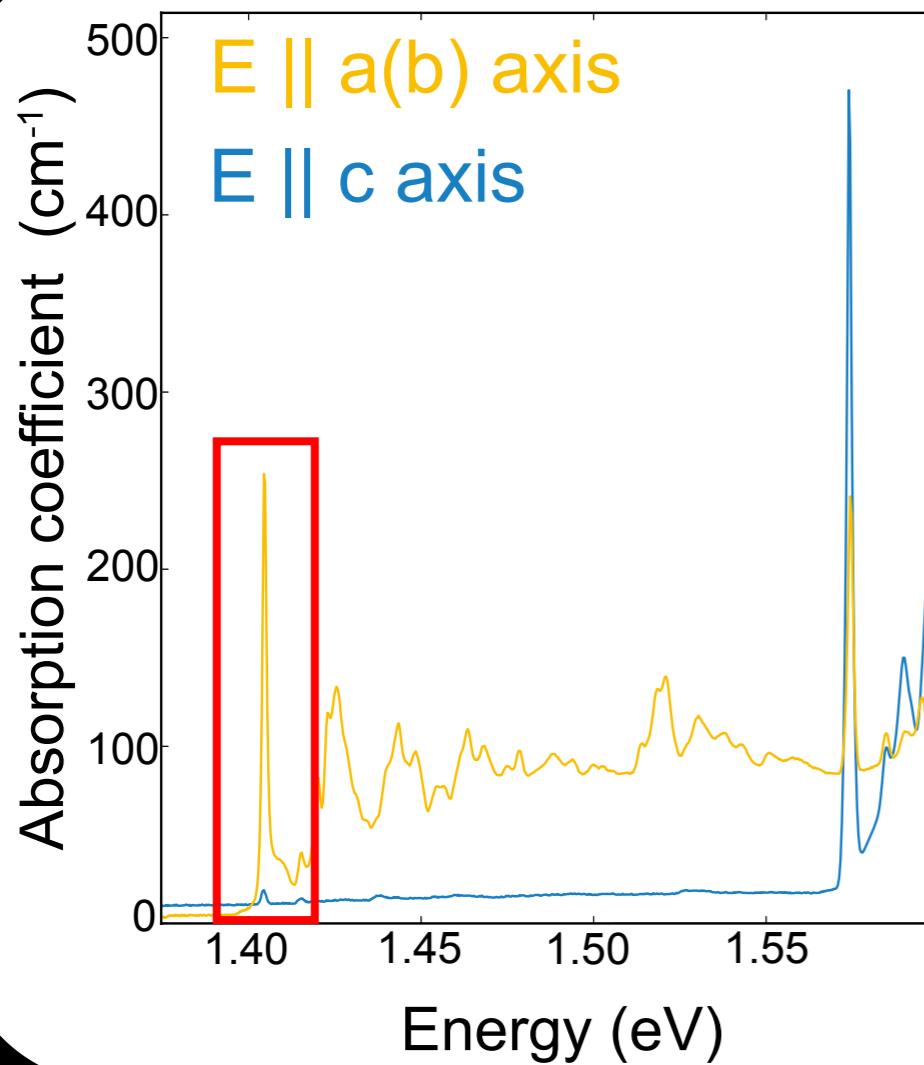
charges + magnons

$$\Delta S = 0$$

Two-magnon mode:  
resonant pumping?



# Resonant magnon pumping

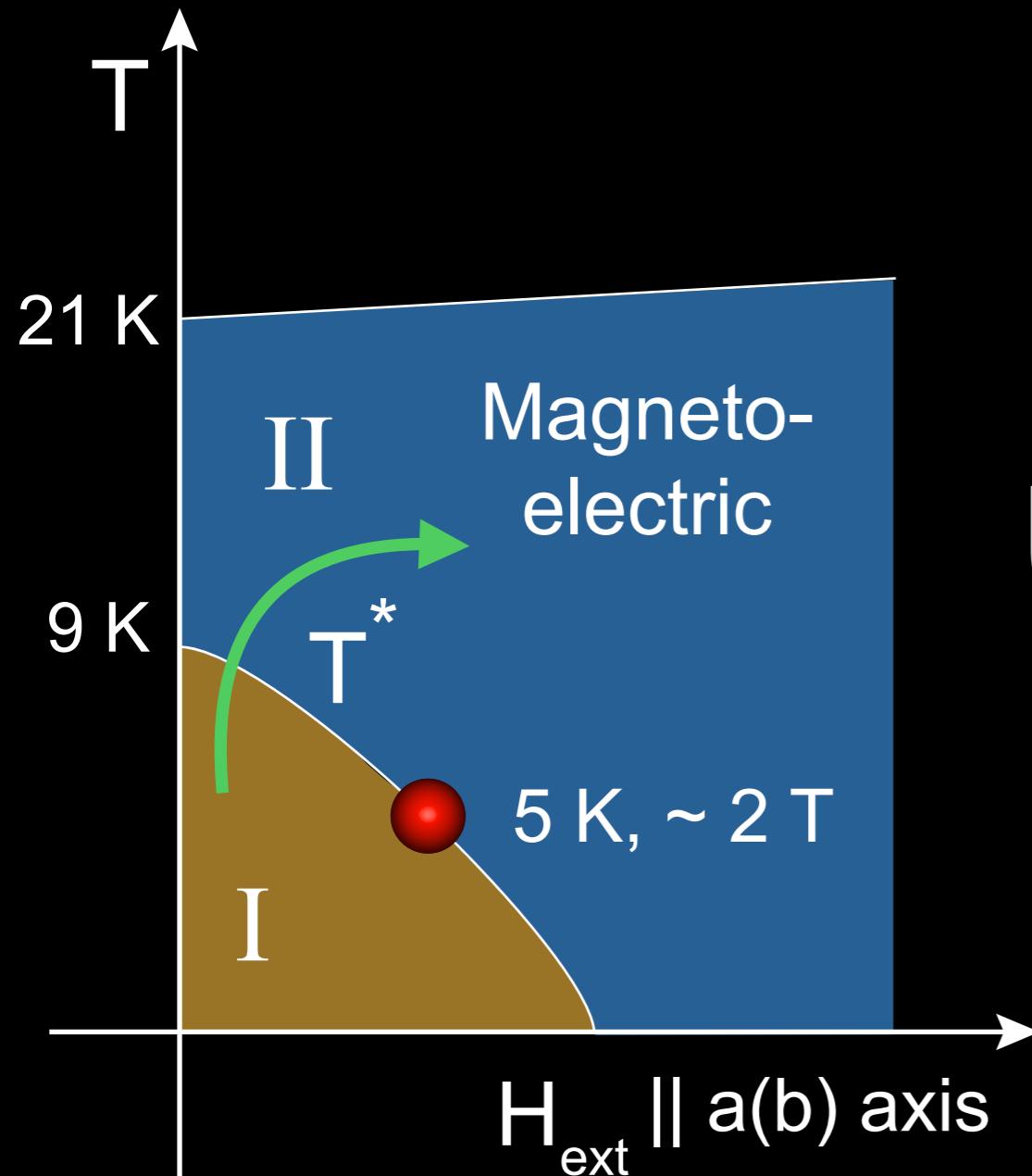


Martynov et al. J. MMM 299, 75 (2006)

Band gap ~ 3.7 eV

**Exciton-Magnon  
Femto-nanomagnons**

# Ultrafast ME activation



$\text{CuB}_2\text{O}_4$

Fiebig et al. J. App. Phys. 93, 6960 (2003)

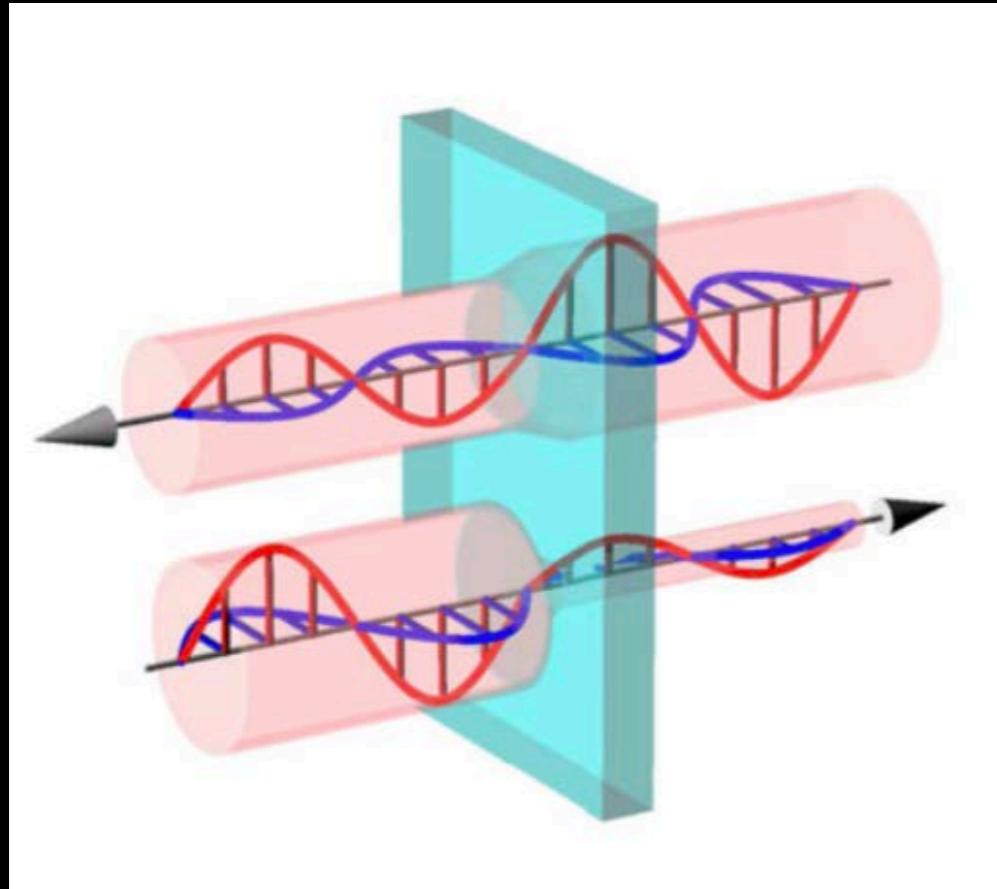
Phase-transition

Ultrafast ME activation

E-field control of spins

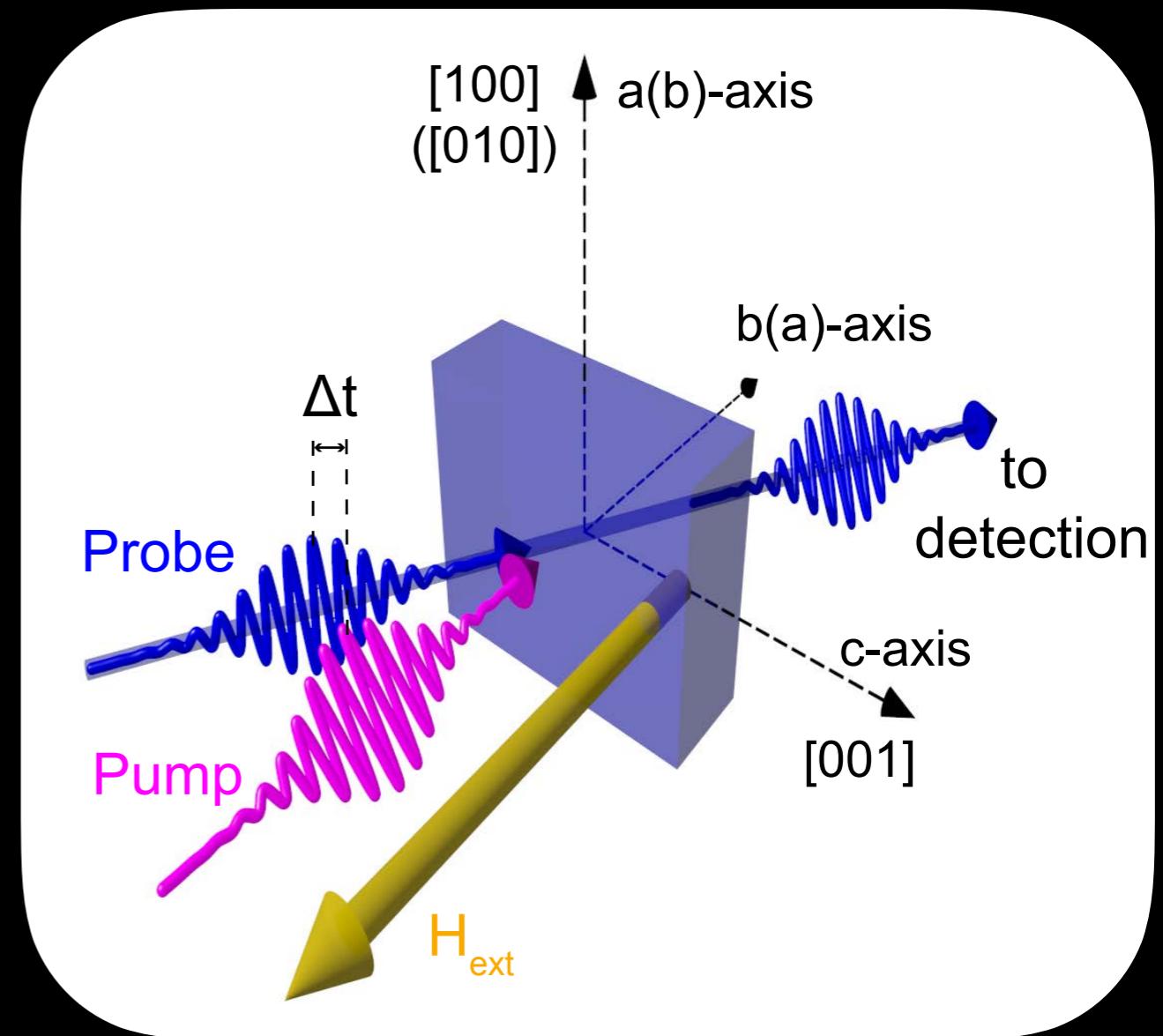
Femtosecond  
Low-dissipations

# Experimental scheme



S. Toyoda et al., Phys. Rev. Lett 115, 267207

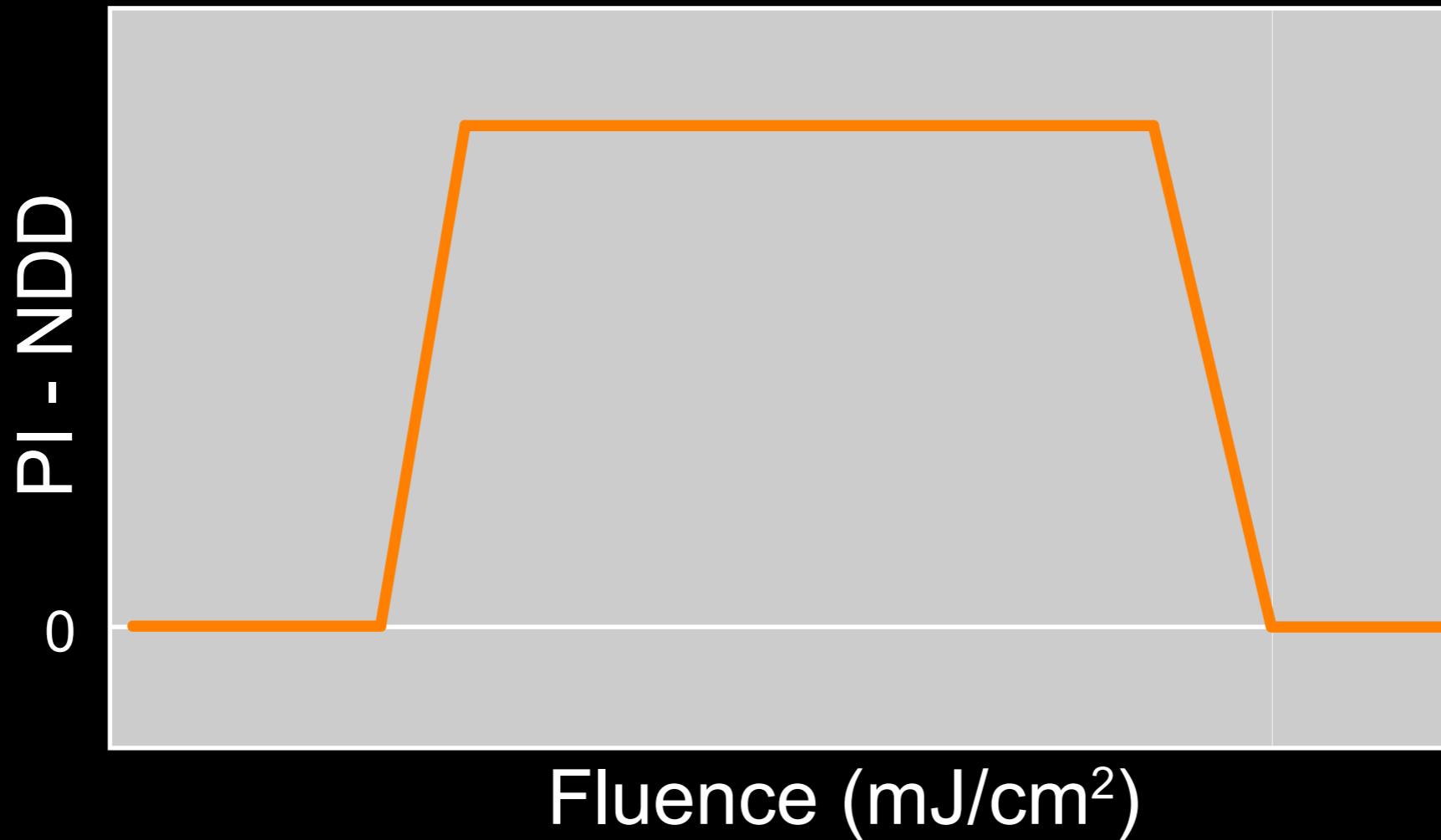
**Non-reciprocal  
directional  
dichroism (NDD)**



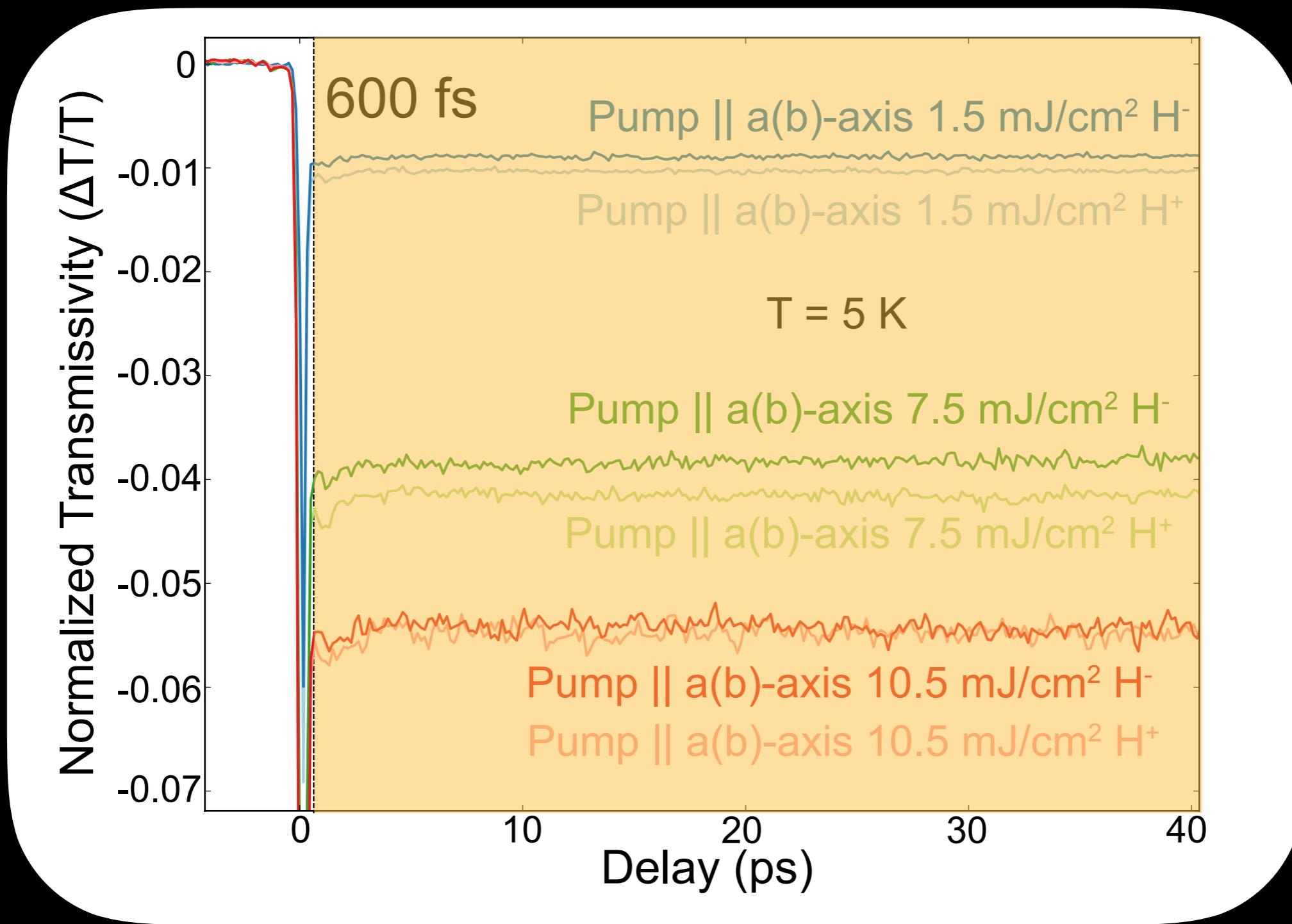
Detection:  $\Delta T/T$

# Expectation

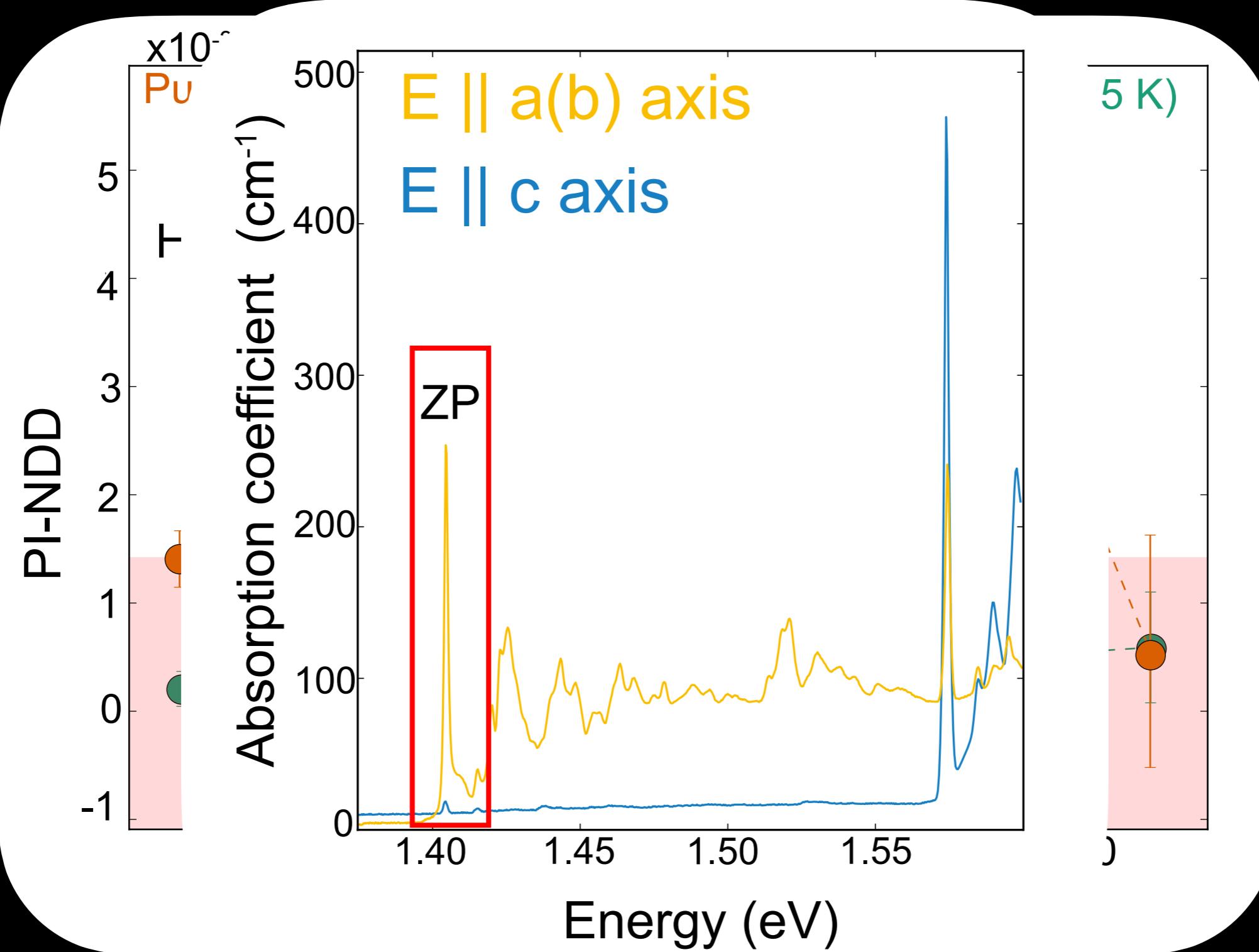
$$\text{PI-NDD} = \frac{\Delta}{\tau}(H^+) - \frac{\Delta}{\tau}(H^-)$$



# Pumping magnons

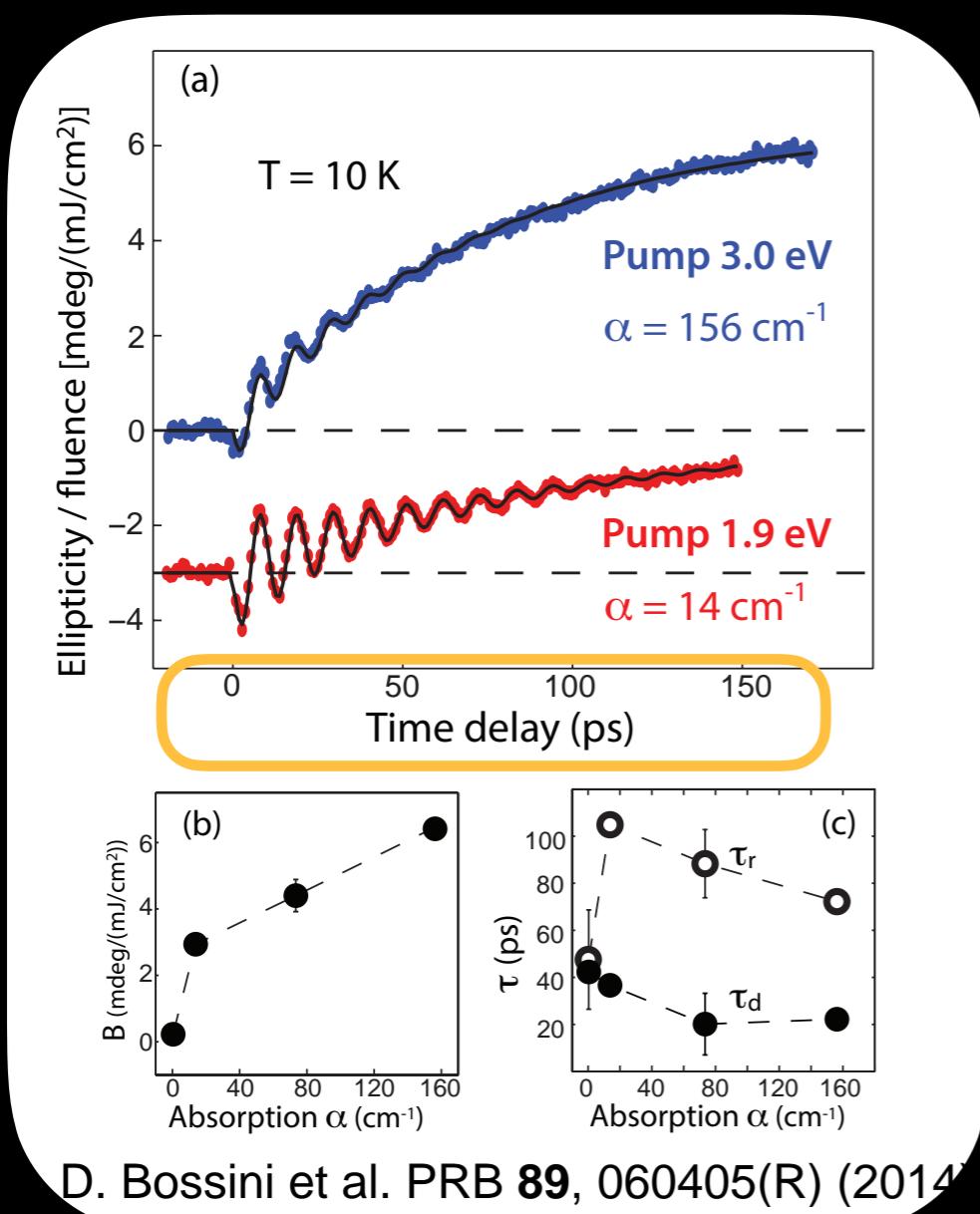


# PhotolInduced-NDD



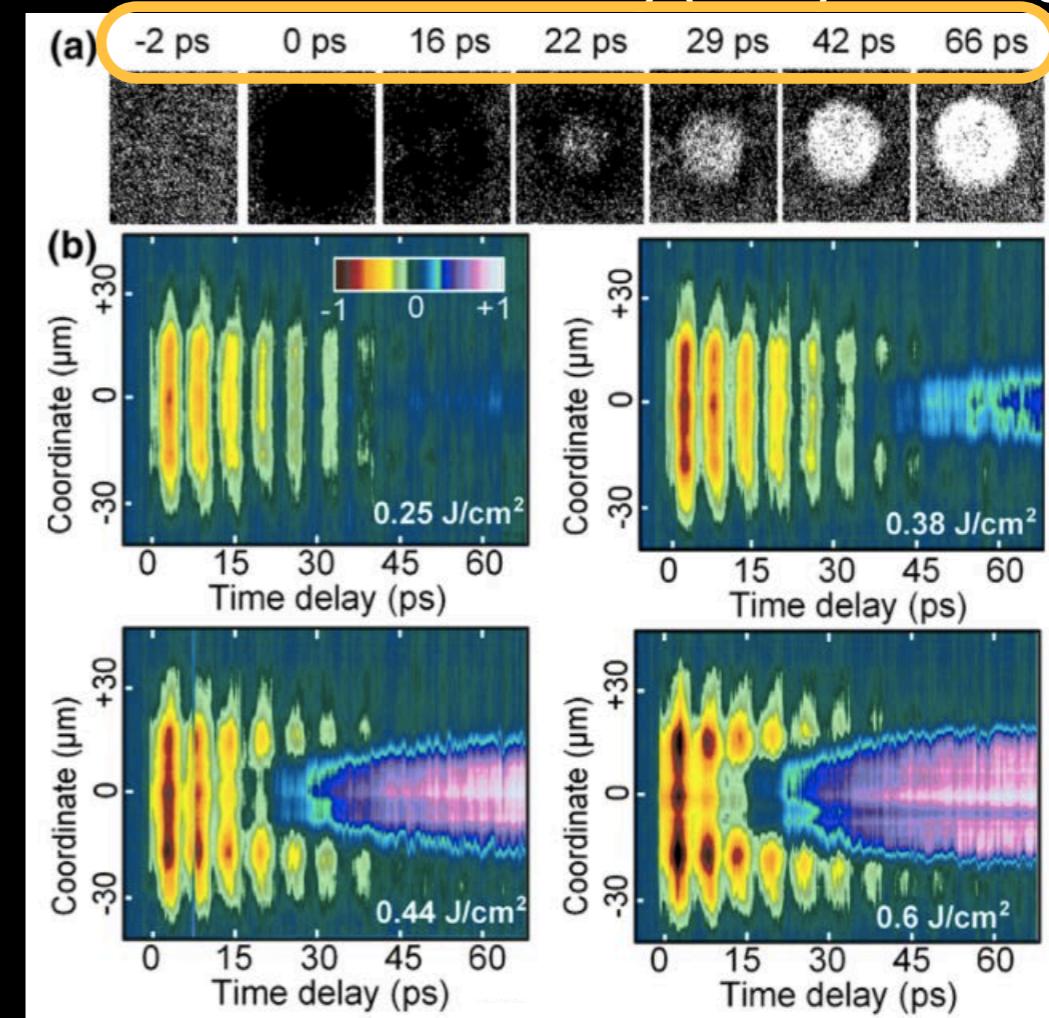
# Magnetoelectricity photoinduced in 600 fs

D. Bossini et al. Nature Phys. (2018)



D. Bossini et al. PRB **89**, 060405(R) (2014)

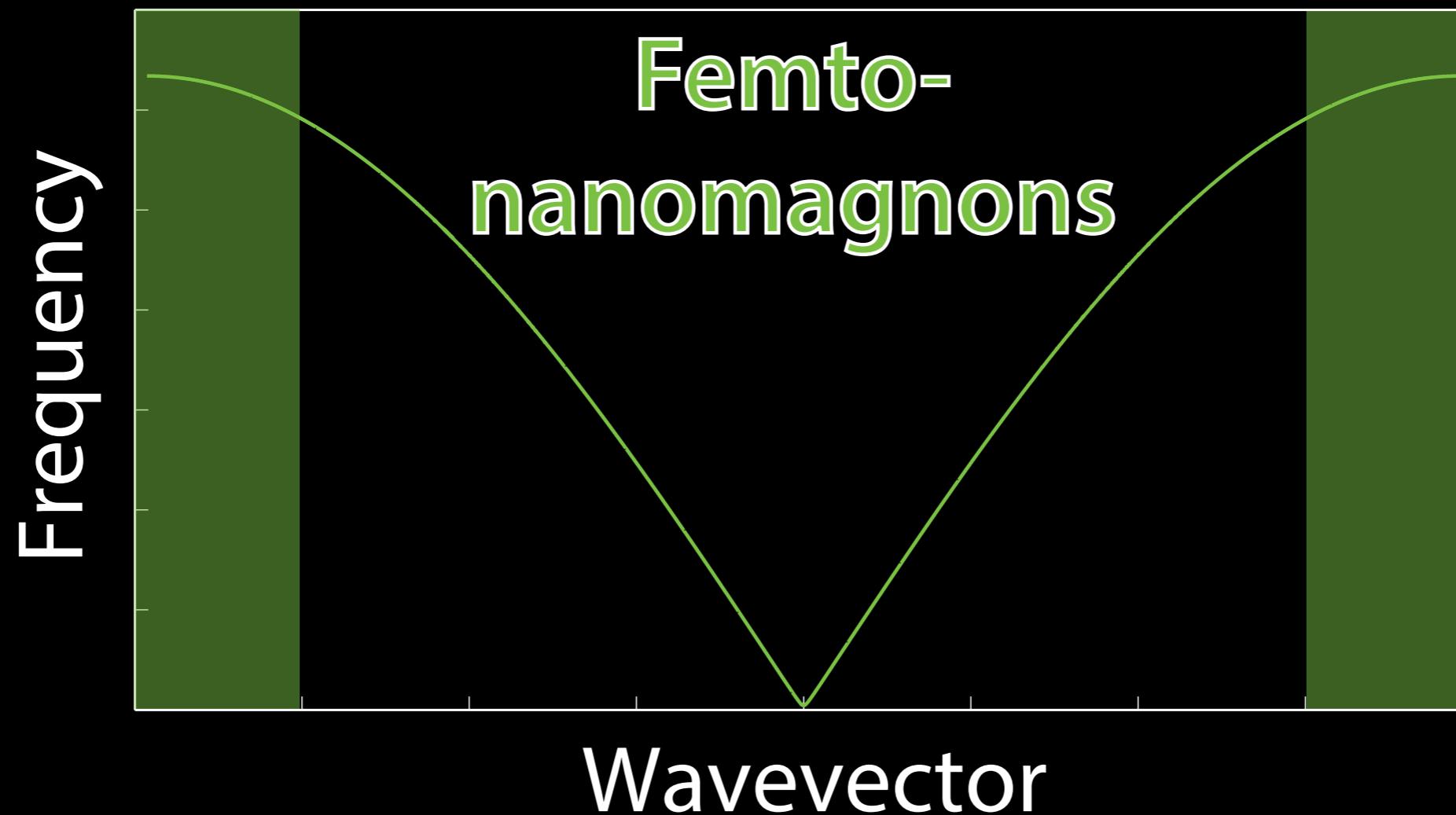
## Morin Point in Dy(Ho)-FeO<sub>3</sub>



D. Afanasiev et al. PRL **116**, 097401 (2016)

D. Afanasiev et al. J.Phys.Cond.Matt. **29**, 224003 (2017)

# Magnetic phase transition in 600 fs?



Magnon-magnon interaction: **670 fs**

V.G. Ivanov et al. PRB. **88**, 094301 (2013)

# Conclusions

- Excitation, control and detection of femtosecond coherent dynamics of order parameter
- Opto-magnetism without absorption throughout the Brillouin zone
- Femtosecond activation of the magneto-electricity

# Outlook

- Femtosecond impulsive reversal of L
- Ultrafast manipulation of the exchange
- Manipulation of the magneto-electricity

**Femtosecond Non-dissipative**

COST Action MAGNETOFON

Work Group 3: dielectrics and magneto-electrics

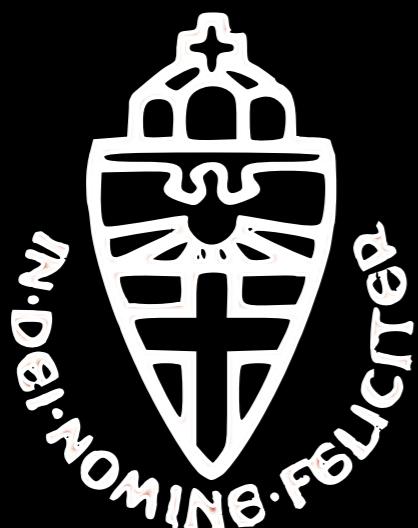
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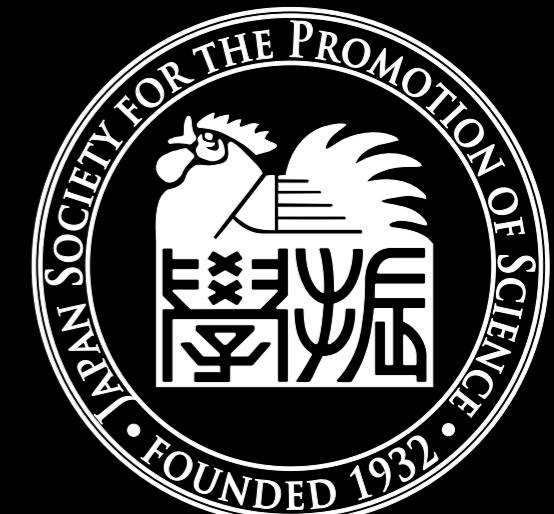
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T. Nova  
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di Milano**  
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S. Dal Conte



# Cinchetti group

## TU Dortmund

