Towards the ultimate limits of spintronics: ultrafast optical control of magnetism

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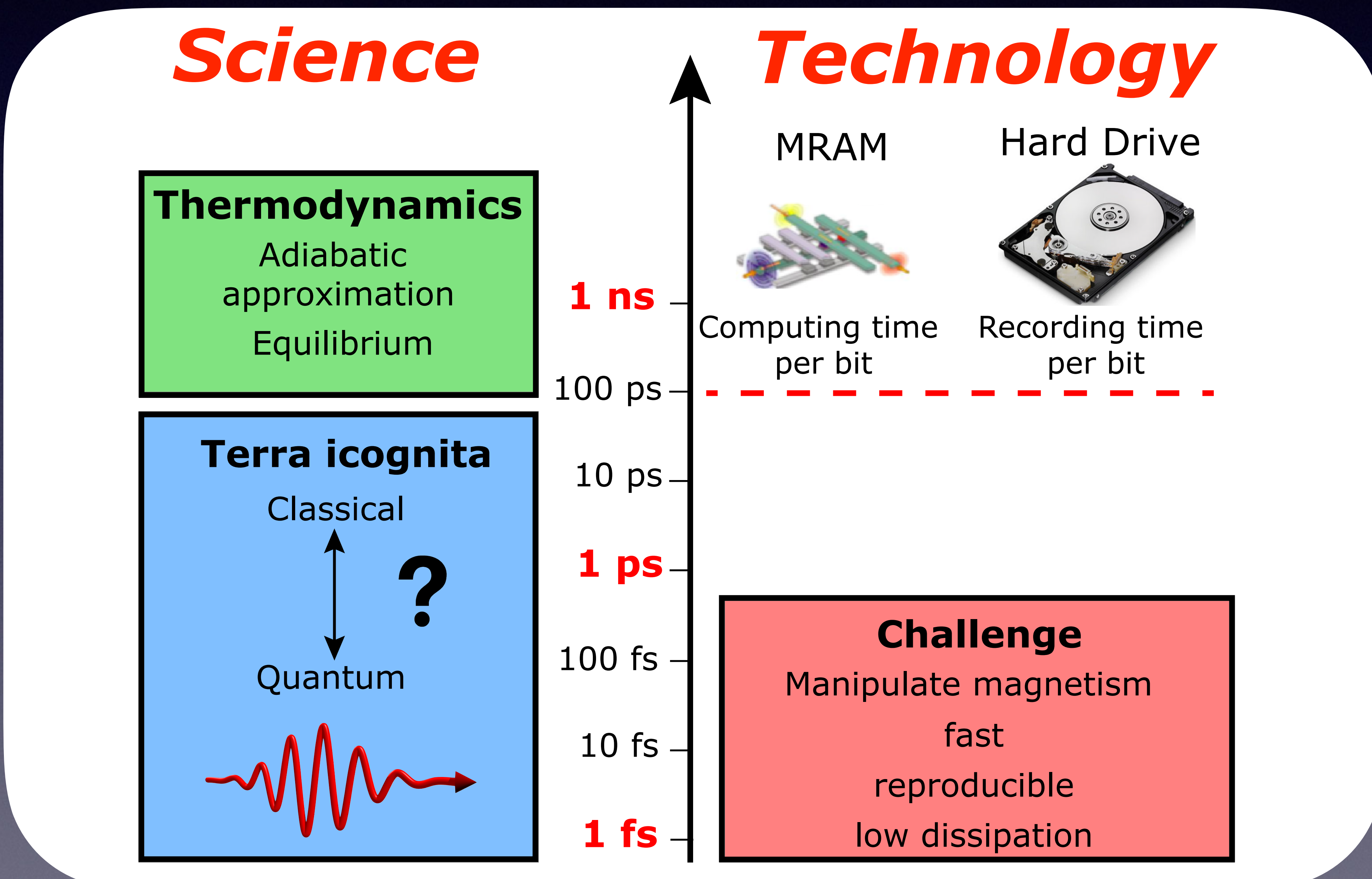
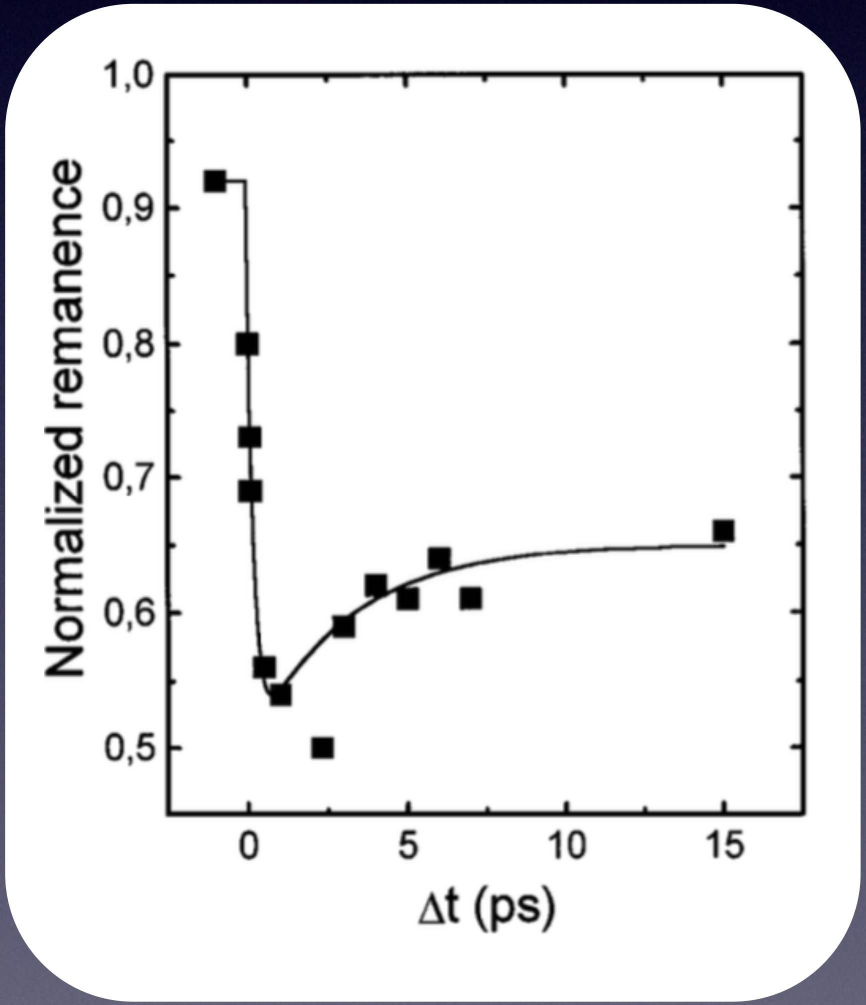
Scientific goal

Ultrafast manipulation of the magnetic order

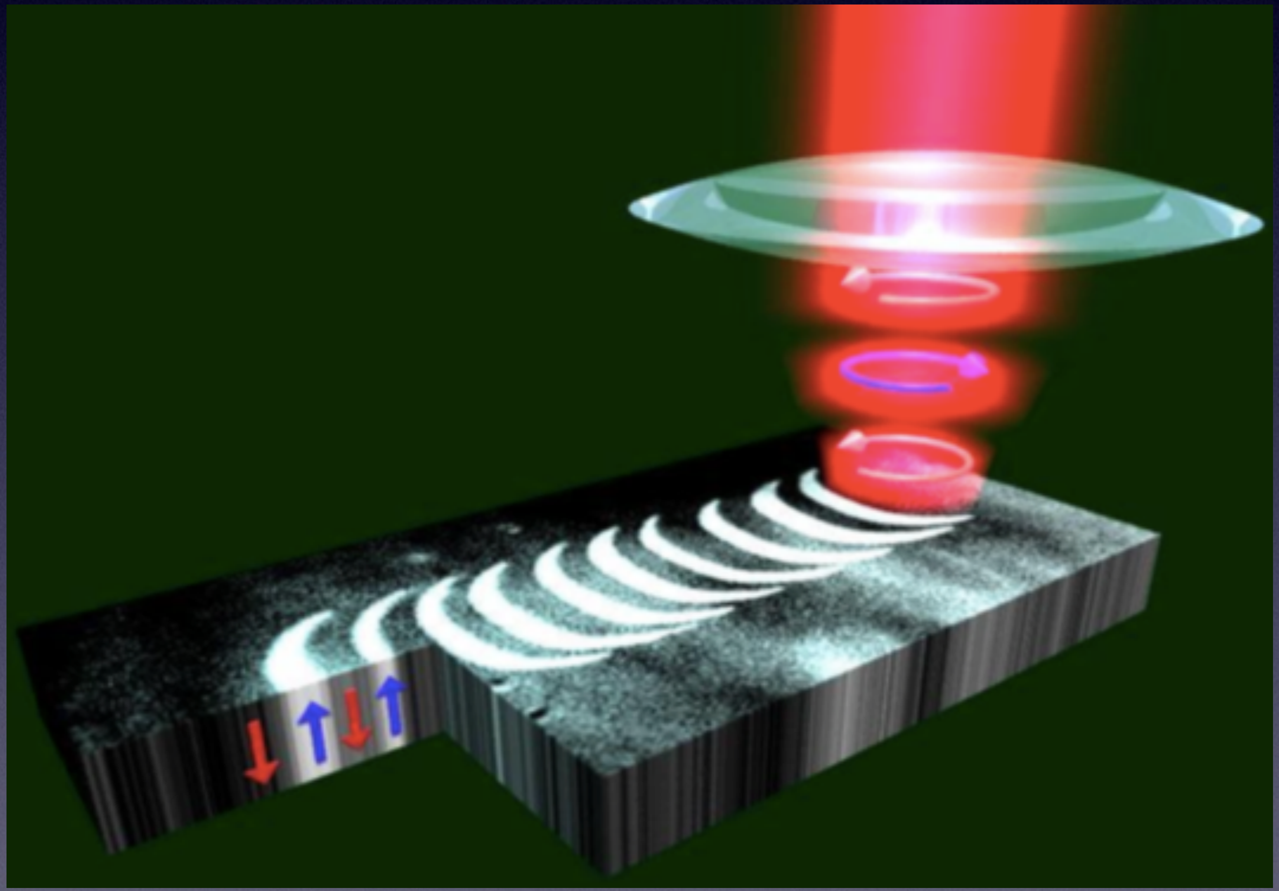


Scientific goal

Femto + Nano + Spin



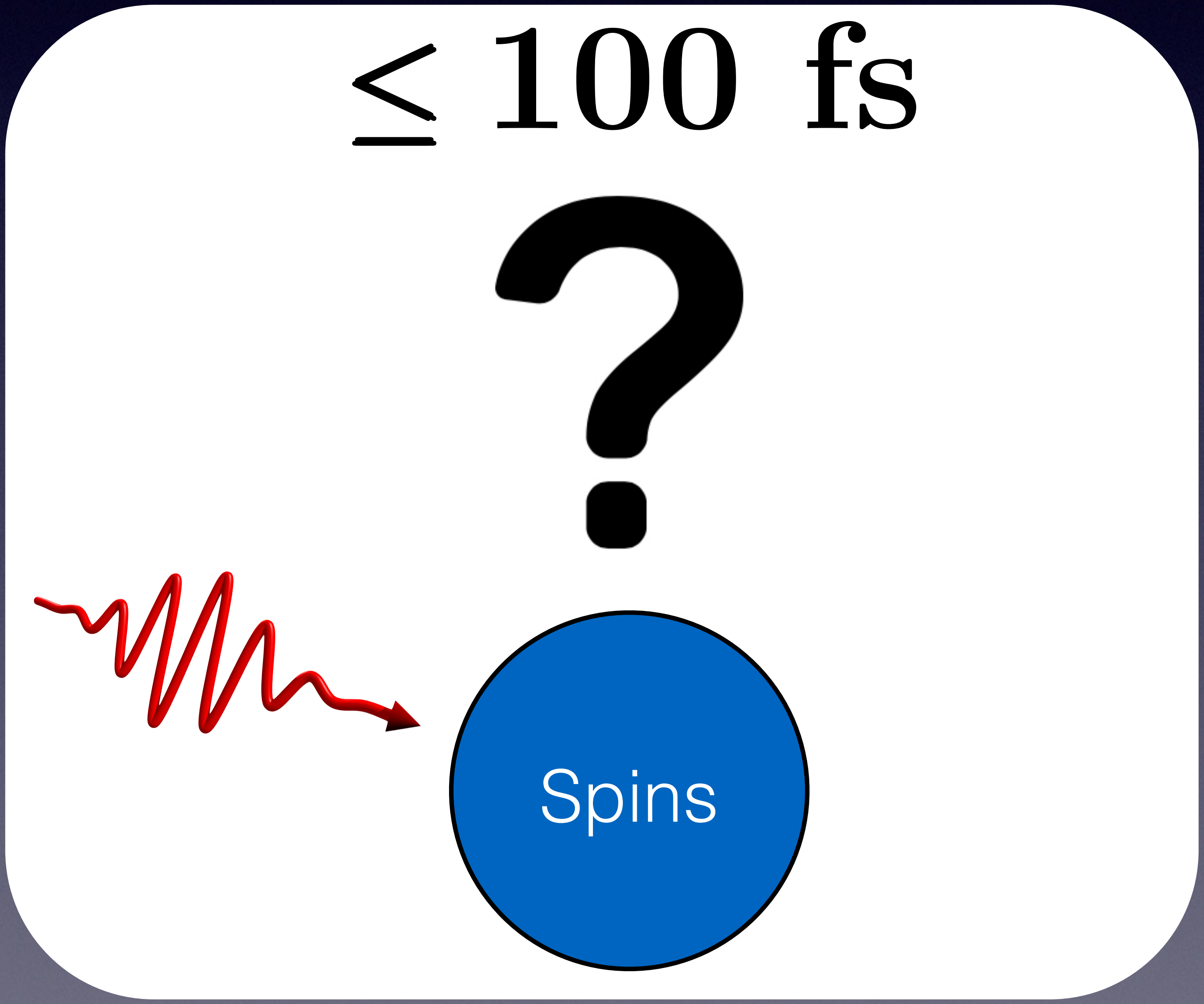
E. Beaurepaire et al. PRL **76**, 4250 (1996)



C. Stanciu et al. Phys. Rev. Lett. **99**, 047601 (2007)
I. Radu et al. Nature **472**, 205 (2011)
J.H. Mentink et al. Phys. Rev. Lett. **108**, 057202 (2012)
T. Ostler et al. Nat. Comm. **3**, 666 (2012)

Scientific goal

Femto + Nano + Spin

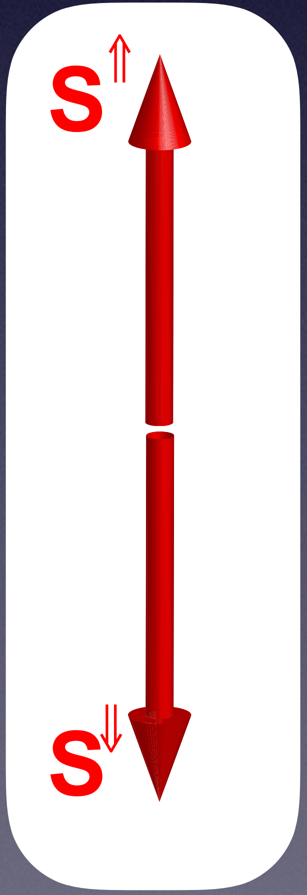


Dielectric antiferromagnet

- ✓ No free electrons
- ✓ Majority of magnetically ordered materials
- ✓ No stray field, technological potential
- ✓ Intrinsically faster spin dynamics

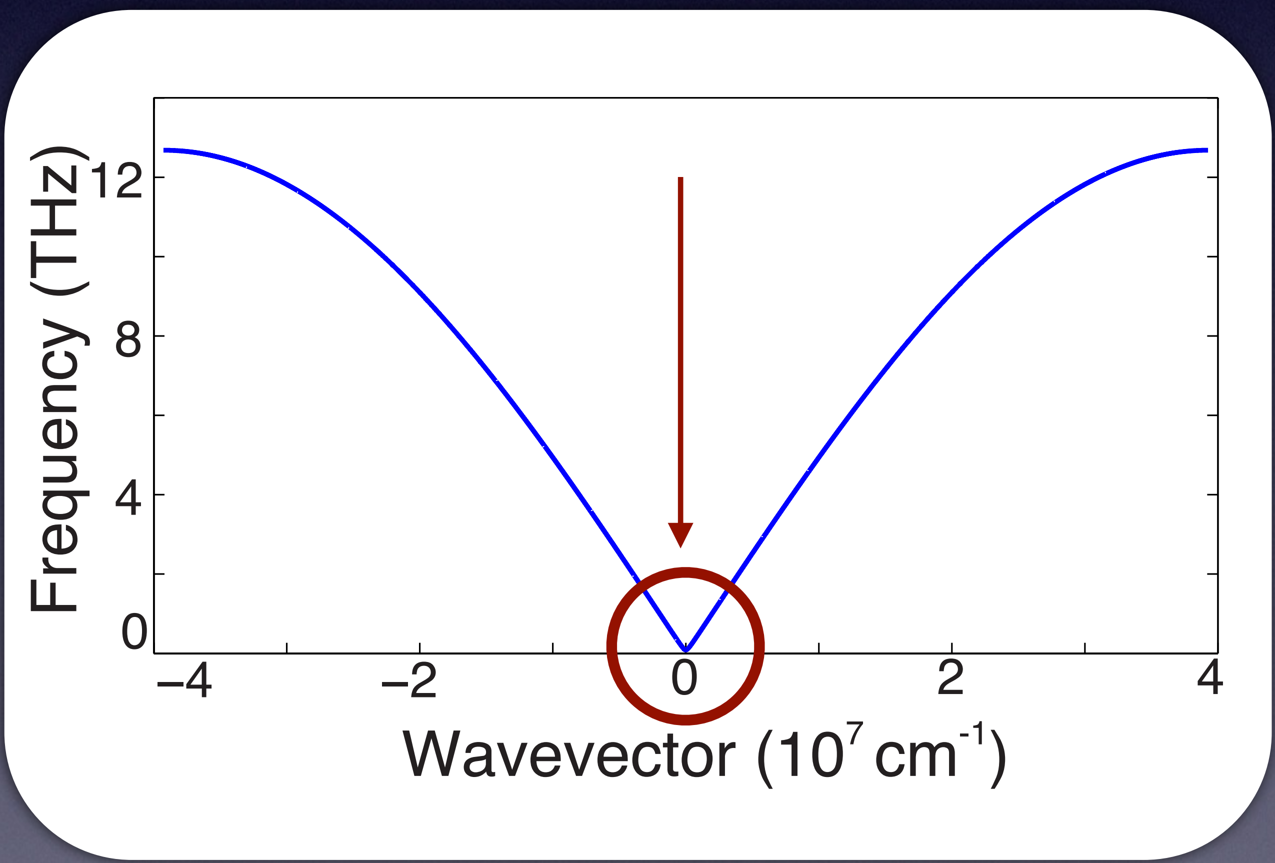
$$\hat{H} = J \sum_{\langle i,j \rangle} \hat{\boldsymbol{S}}_i \cdot \hat{\boldsymbol{S}}_j$$

Collinear magnetic sublattices



Dispersion in AF

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



$$\omega_{\boldsymbol{q}} = \sqrt{(\omega_E + \omega_A)^2 - (\omega_E \gamma_{\boldsymbol{q}})^2}$$

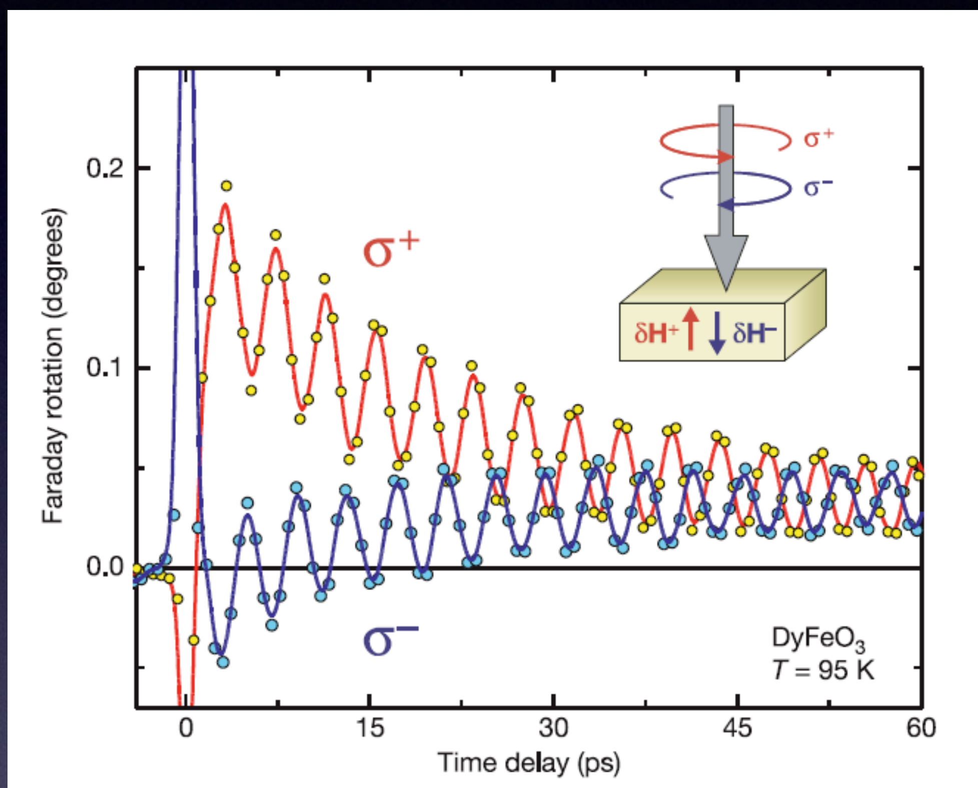
$$\omega_E \equiv JSz$$

$$\omega_A \equiv g\mu_B H_A$$

$$\gamma_{\boldsymbol{q}} = \frac{1}{z} \sum_{\boldsymbol{\delta}} \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{\delta}}$$

Opto-magnetism

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



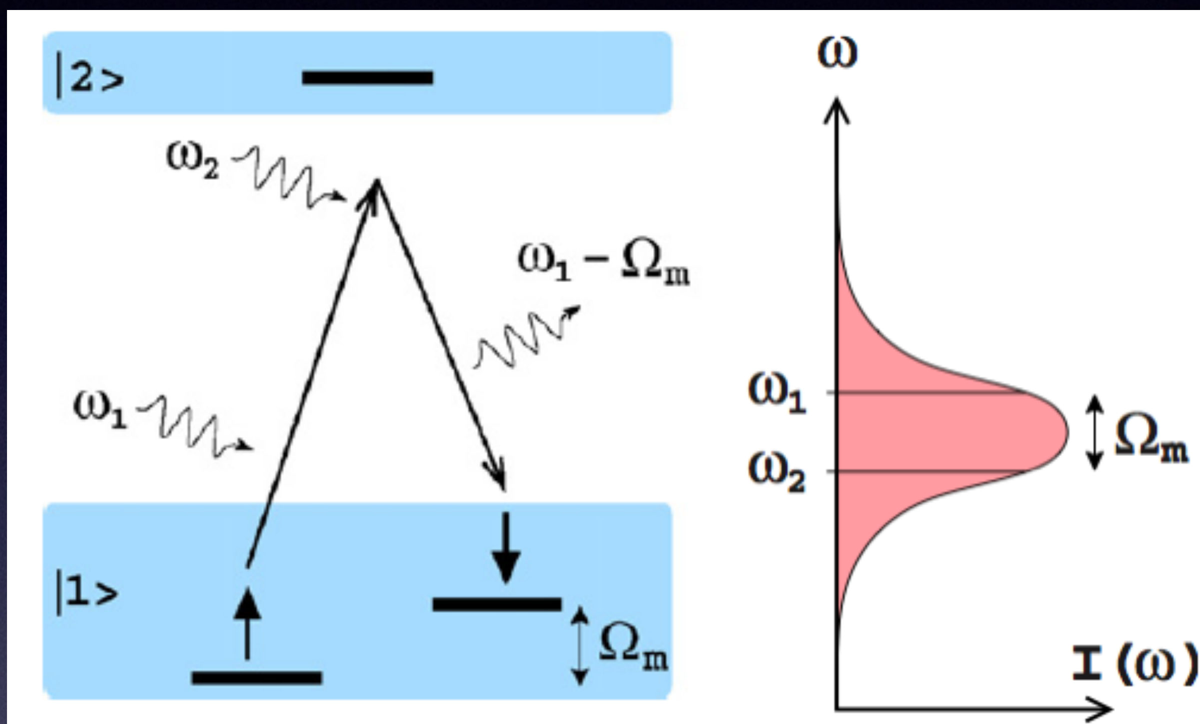
- ✓ Excitation of coherent spin waves
- ✓ Magneto-optical probe: Faraday rotation
- ✓ Pump wavelength: 800 nm

Pump beam ≈ 10^{14} Hz

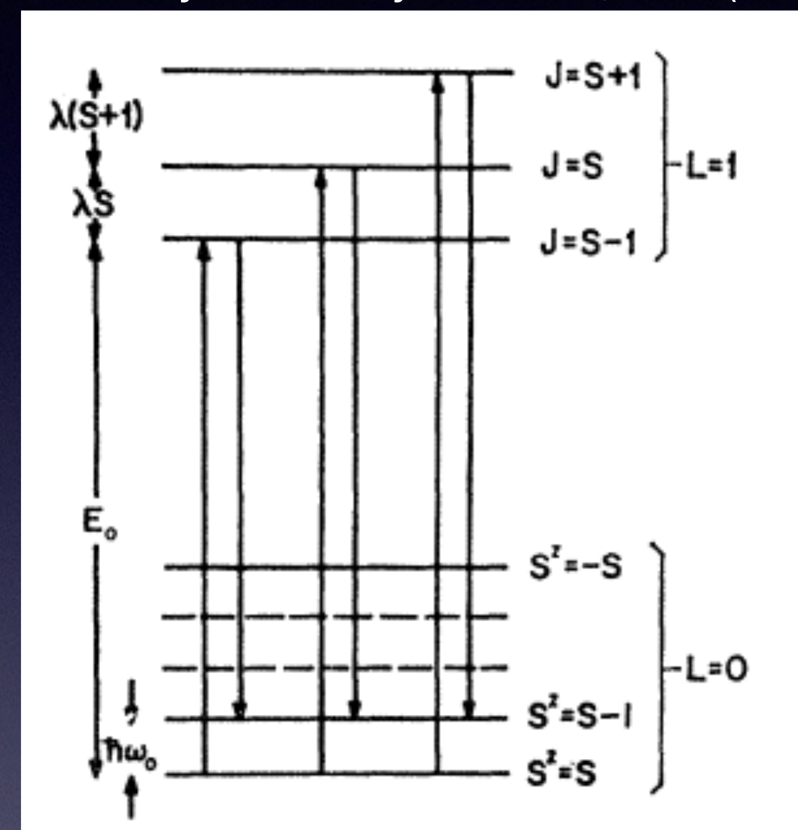
Spin wave ≈ 10^{11} Hz

Resonant magnetic dipole coupling ?

Impulsive Stimulated Raman Scattering (ISRS)



P. Fleury *et al.* Phys. Rev. **2,** 514 (1968)



Raman on magnons

- ✓ Stimulated Raman emission
- ✓ Coherent process

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

Light-spin interaction

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

$\hat{S}_i^{x,y}$ spin deviations

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

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

$\Delta S = 1$

Faraday effect

$\langle \hat{S}_i^z \rangle \langle \hat{S}_i^{x,y} \rangle$

LINEAR in the spin deviations

$\Delta S = 1$

$\langle \hat{S}_i^{x,y,z} \rangle \langle \hat{S}_j^{x,y,z} \rangle$

QUADRATIC in the spin deviations

$\Delta S = 0$

Magnetic Linear Birefringence

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

General concept

T. Satoh et al. Phys. Rev. Lett. **105**, 077402 (2010)

T. Satoh et al. Nat. Phot. **6**, 662 (2012)

T. Satoh et al. Nat. Phot. **9**, 25 (2014)

J. Li et al. Opt. Exp. **19**, 22550 (2011)

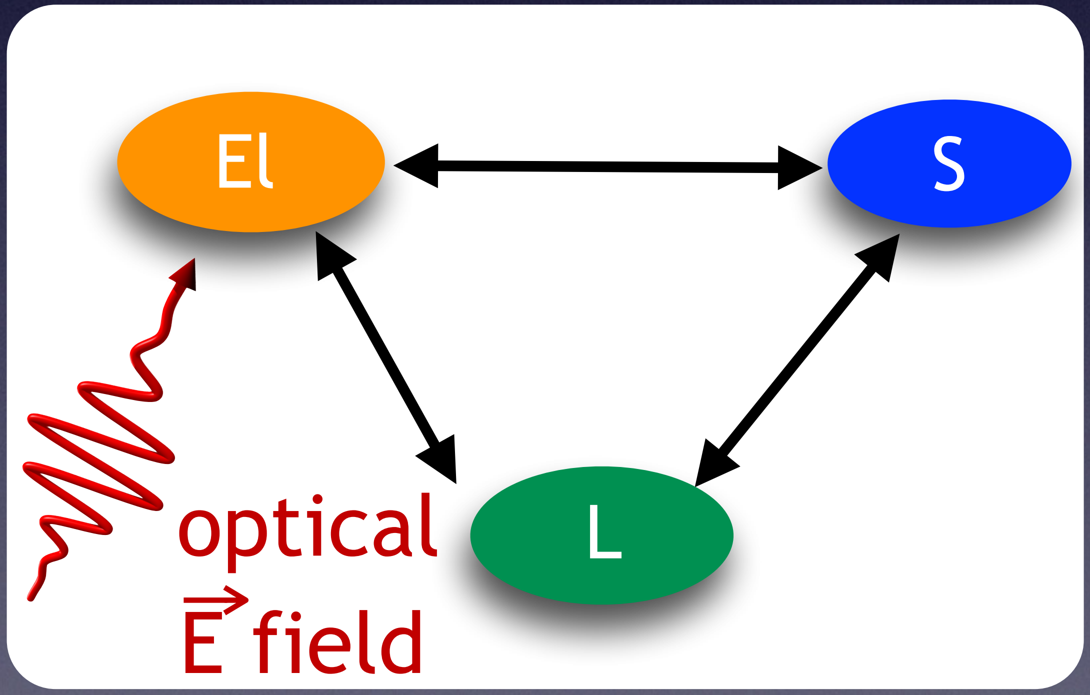
N. Kanda et al. Nat. Comm. **2**, 362 (2011)

A. Kirilyuk et al Rev. Mod .Phys. **82**, 2731, (2010)

S. Parchenko et al APL **108**, 032404, (2016)

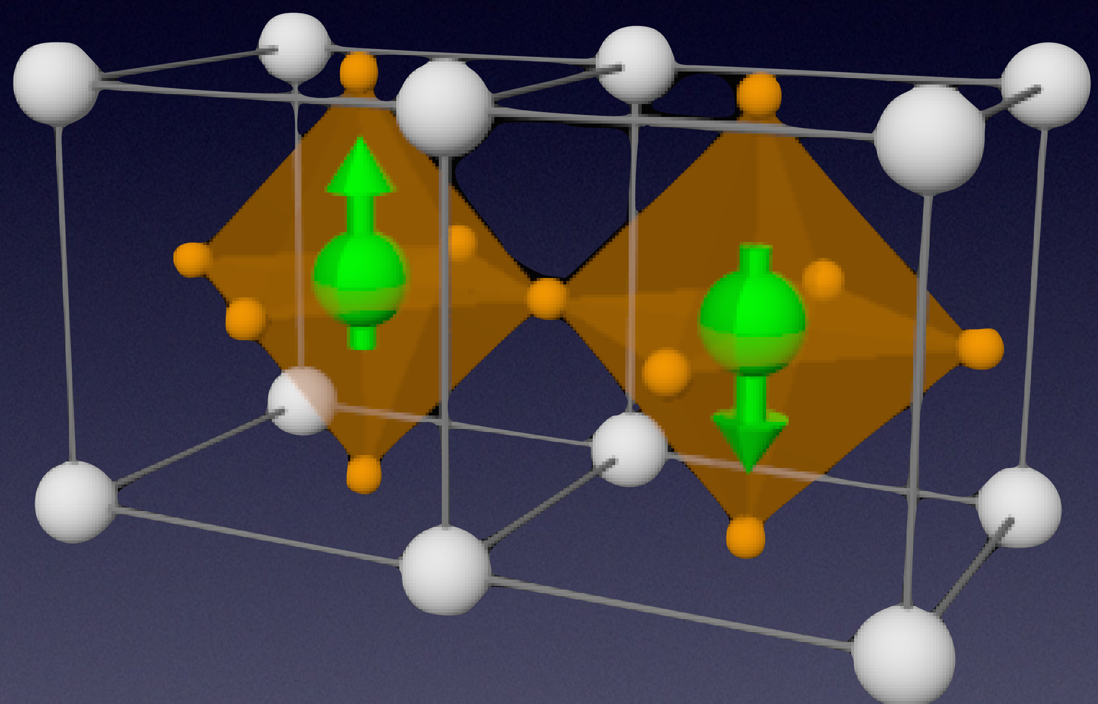
Different magnetic structures: ferrimagnet, canted AF, collinear AF

Absorption of the pump beam: **heating!**

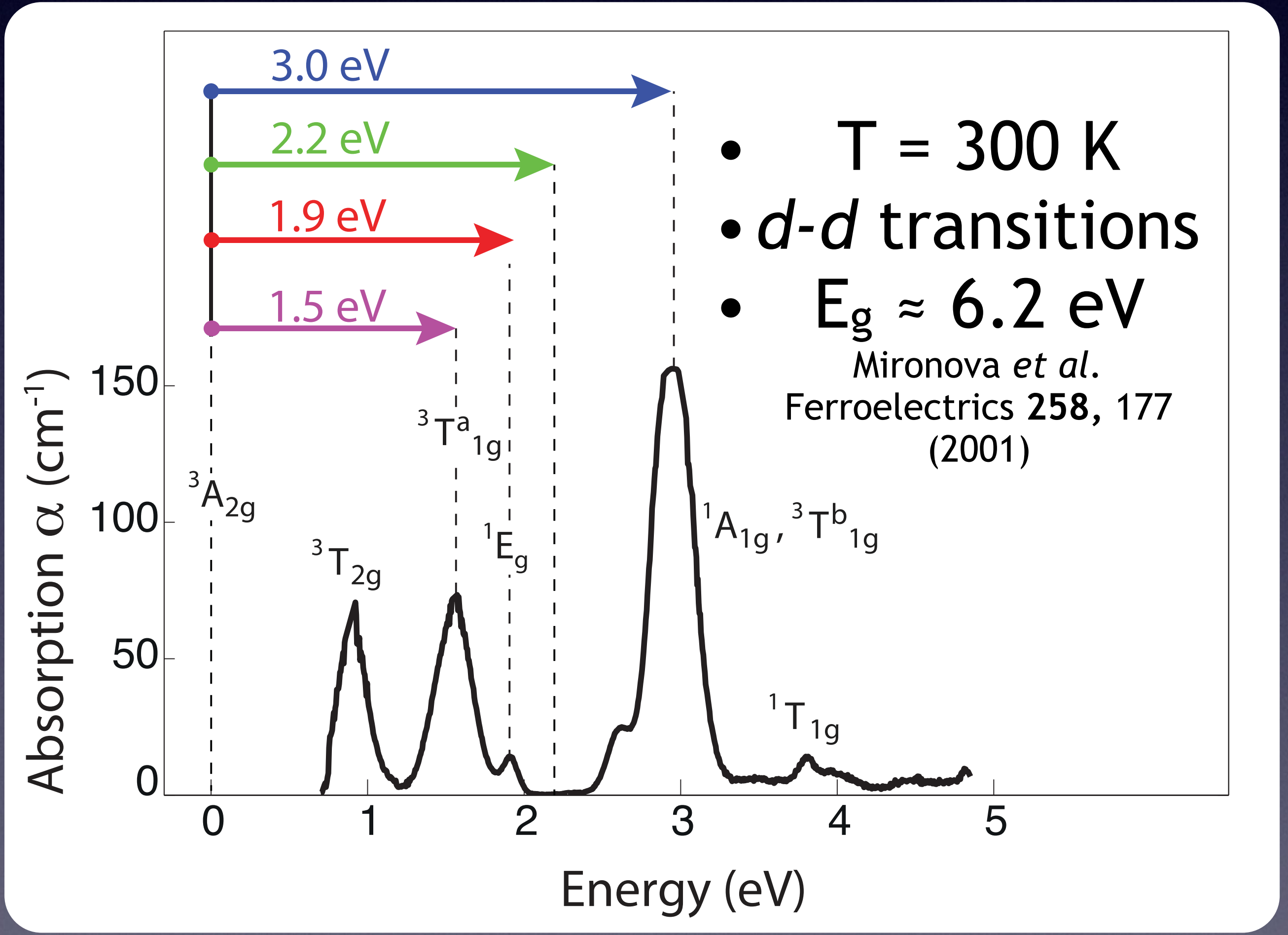


Opto-magnetism without absorption ?

Sample: $KNiF_3$

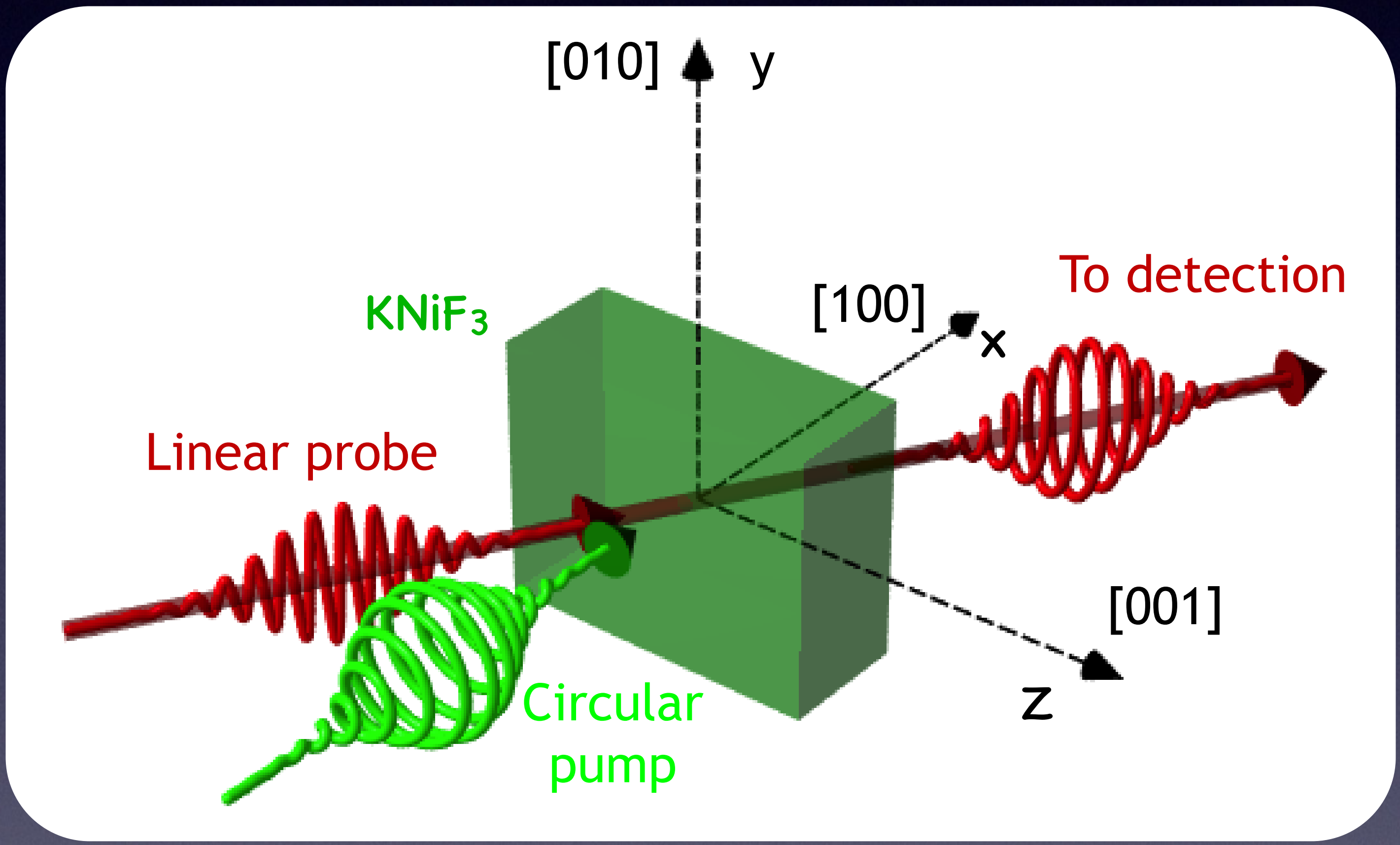


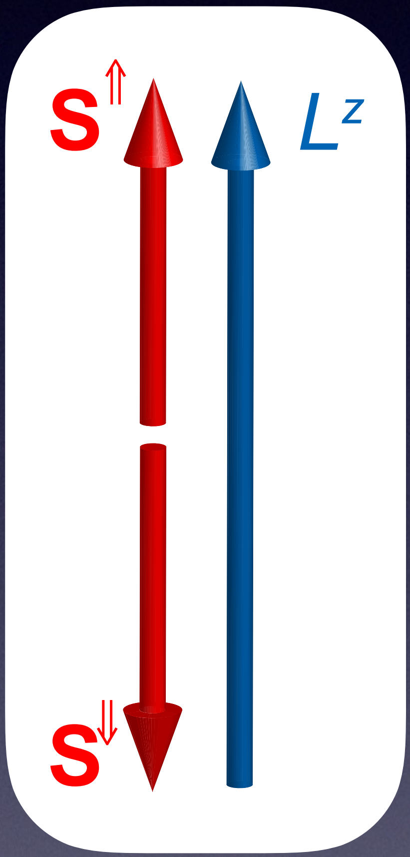
Cubic Heisenberg AF
(T_N = 246 K)



Experimental scheme

Spectral dependence of the ultrafast opto-magnetic effect





- ✓ Tunability range: (0.48 - 6.5) eV
- ✓ Quantity probed: ellipticity

$$L \equiv S^{\Uparrow} - S^{\Downarrow}$$

Antiferromagnetic vector

Magnetic linear birefringence (MLB)

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

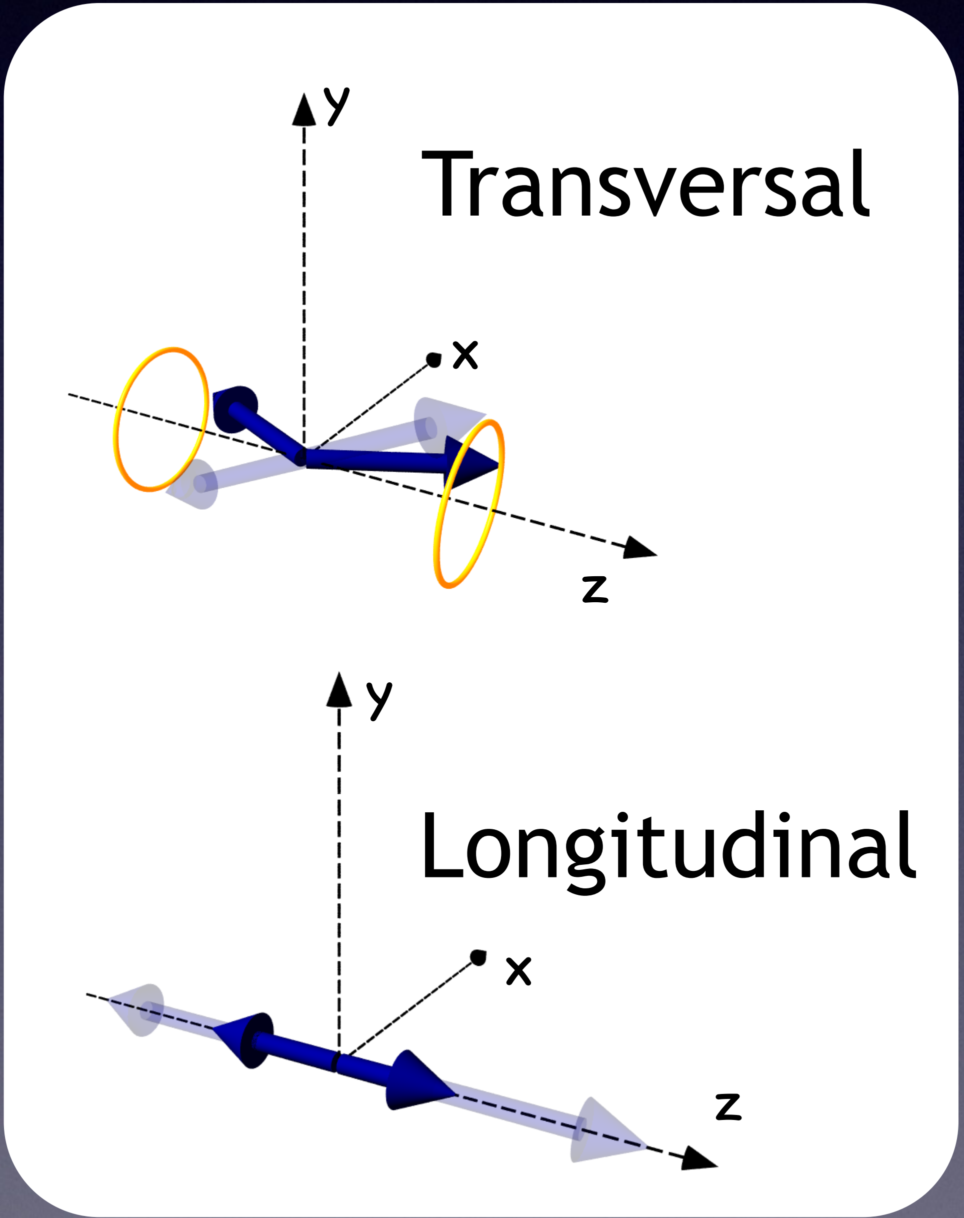
J. Ferre and G. Gehring, *Rep. Prog. Phys.* **47**, 513 (1984)

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

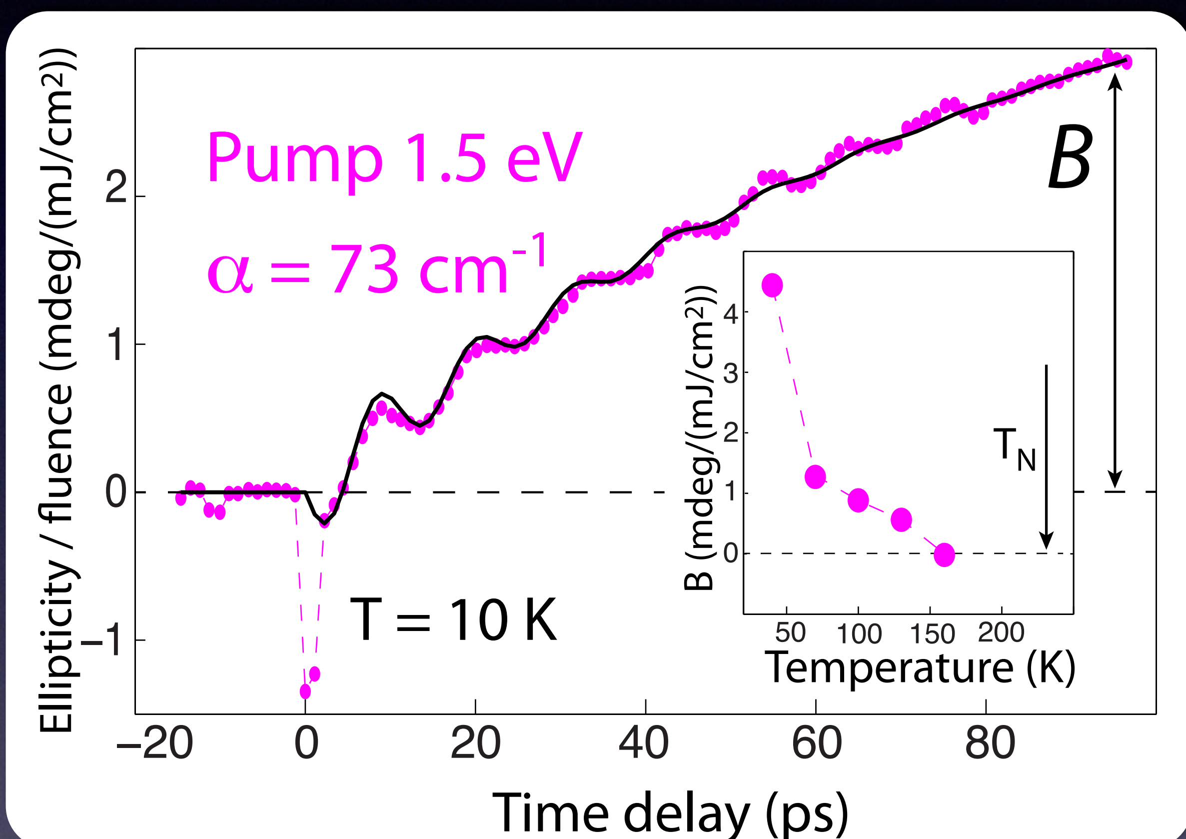
A.F Andreev et al Sov. Phys. Usp. **23**, 21 (1980)

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

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



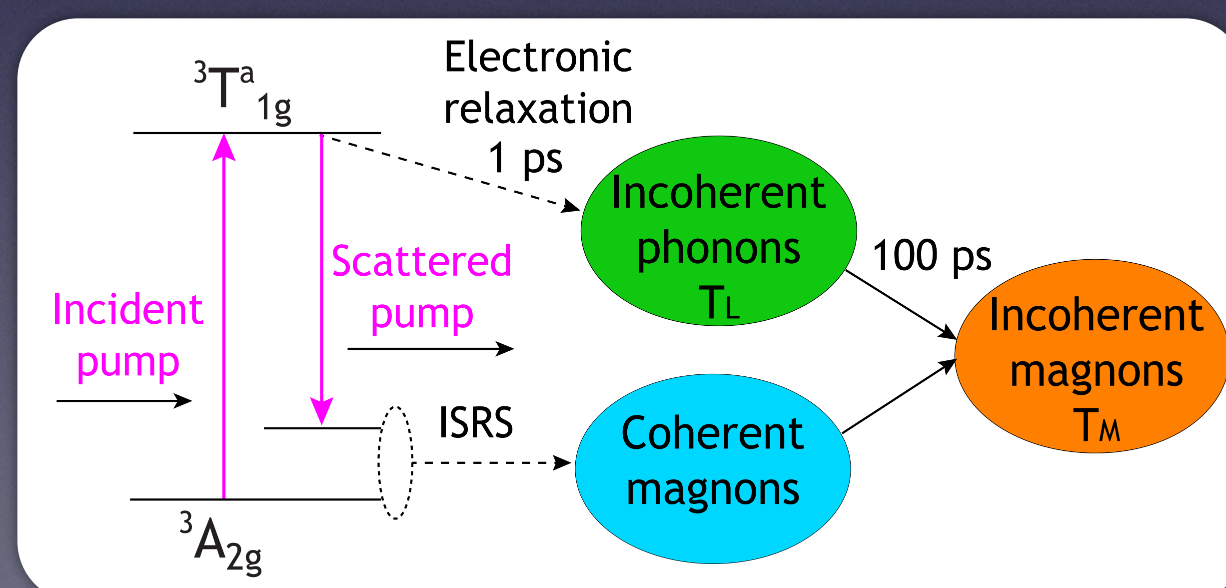
Laser-induced dynamics



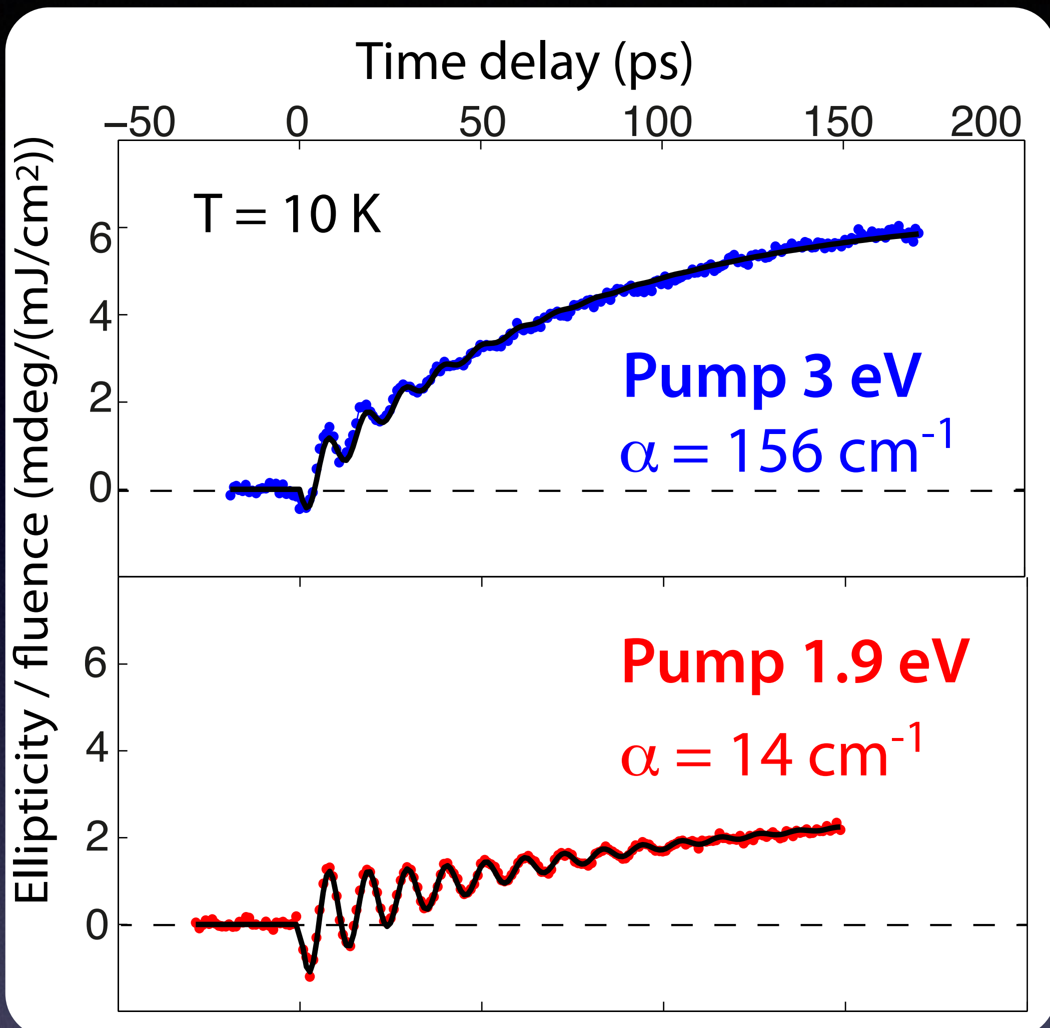
$$f(t) = \theta(t)[A\sin(2\pi\nu t)e^{(-t/\tau_d)} + B(1-e^{(-t/\tau_r)})]$$

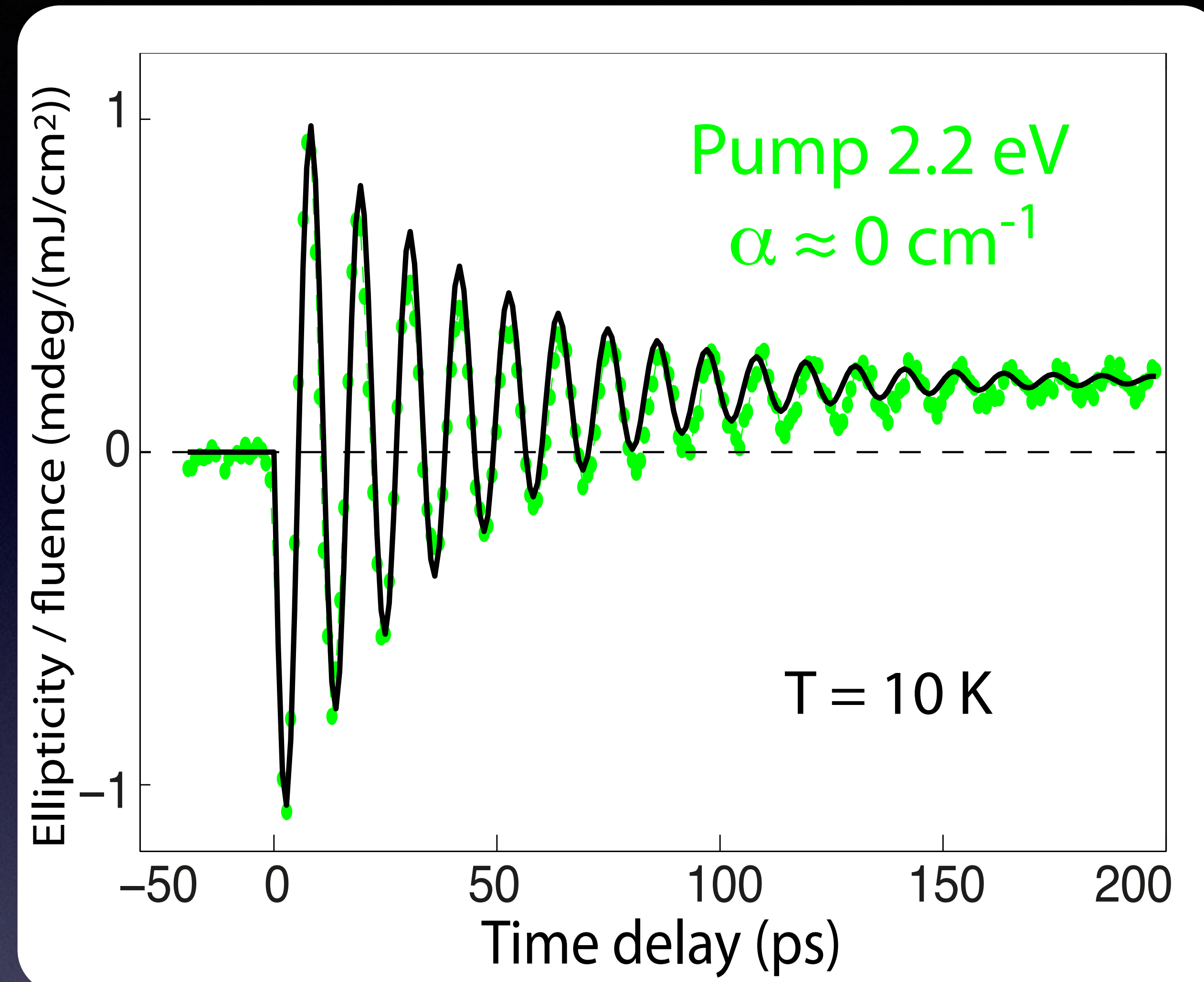
Transversal Longitudinal

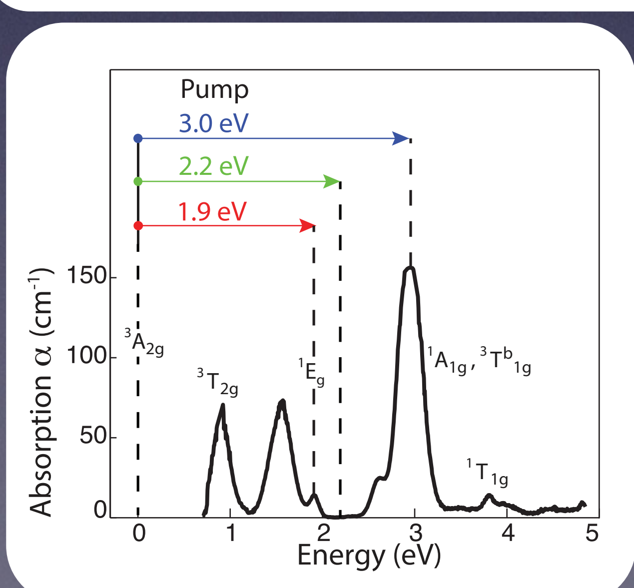
- ✓ Oscillations @ 90 GHz (AFM mode)
- ✓ Slower incoherent dynamics



Spectral dependence







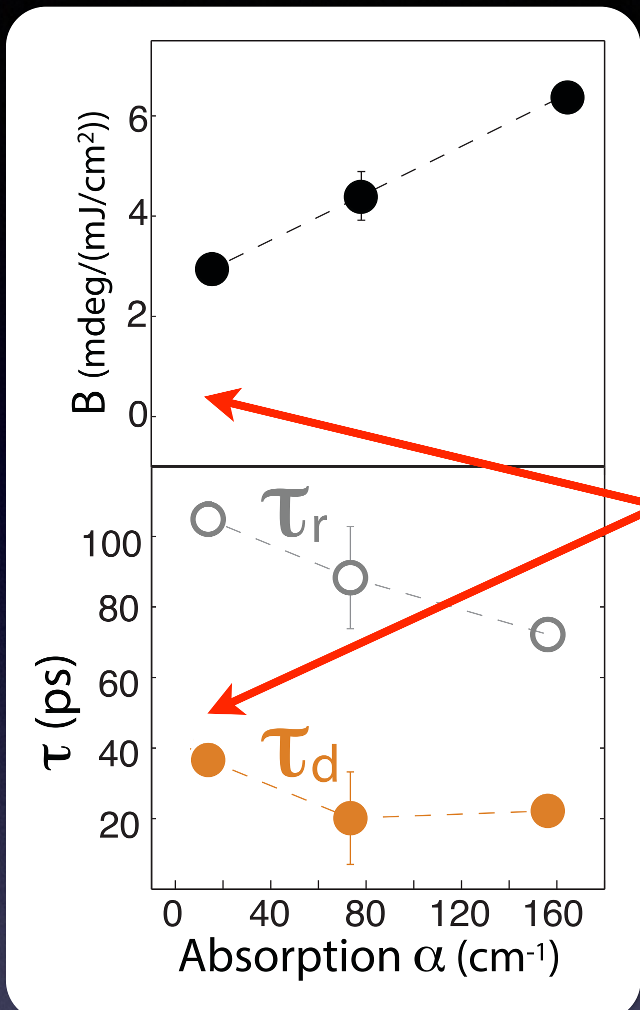
✓ Dissipative regime: picture confirmed

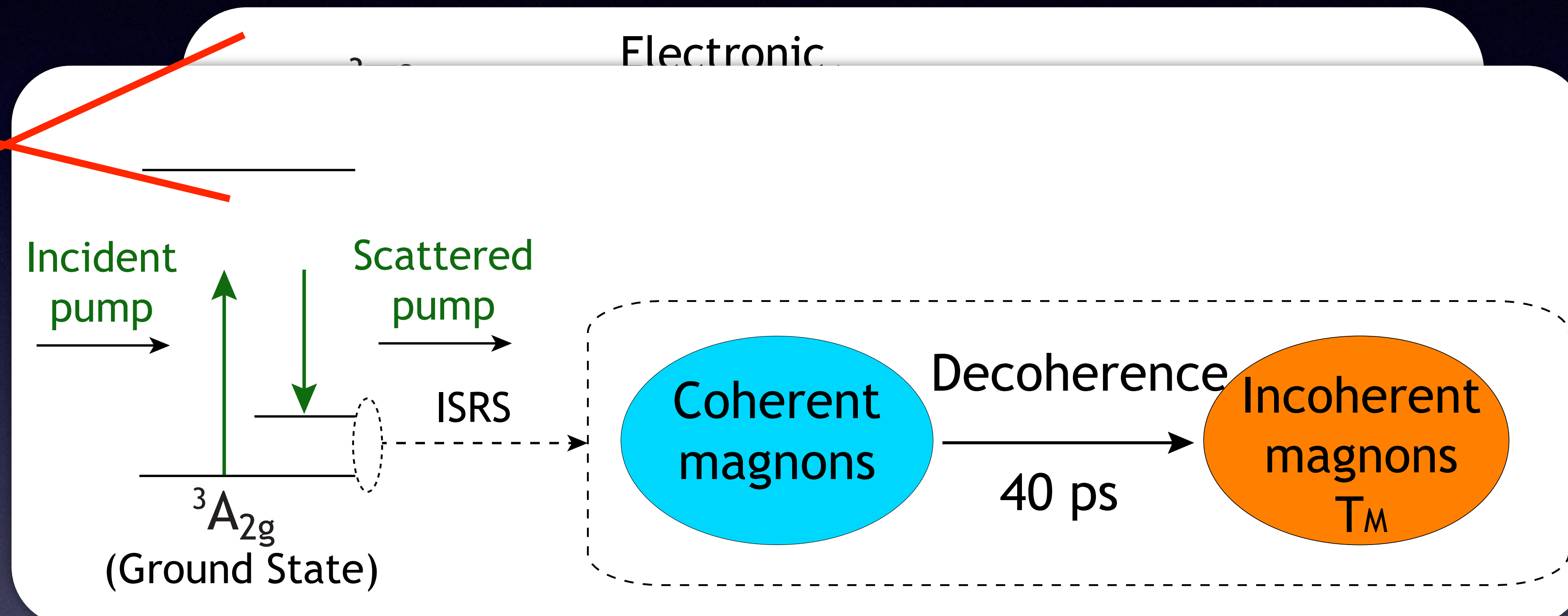
✓ Non-dissipative regime: amplitude of the oscillations unaffected

✓ Non-zero incoherent signal

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

$\tau_d = \tau_r$ → novel regime!



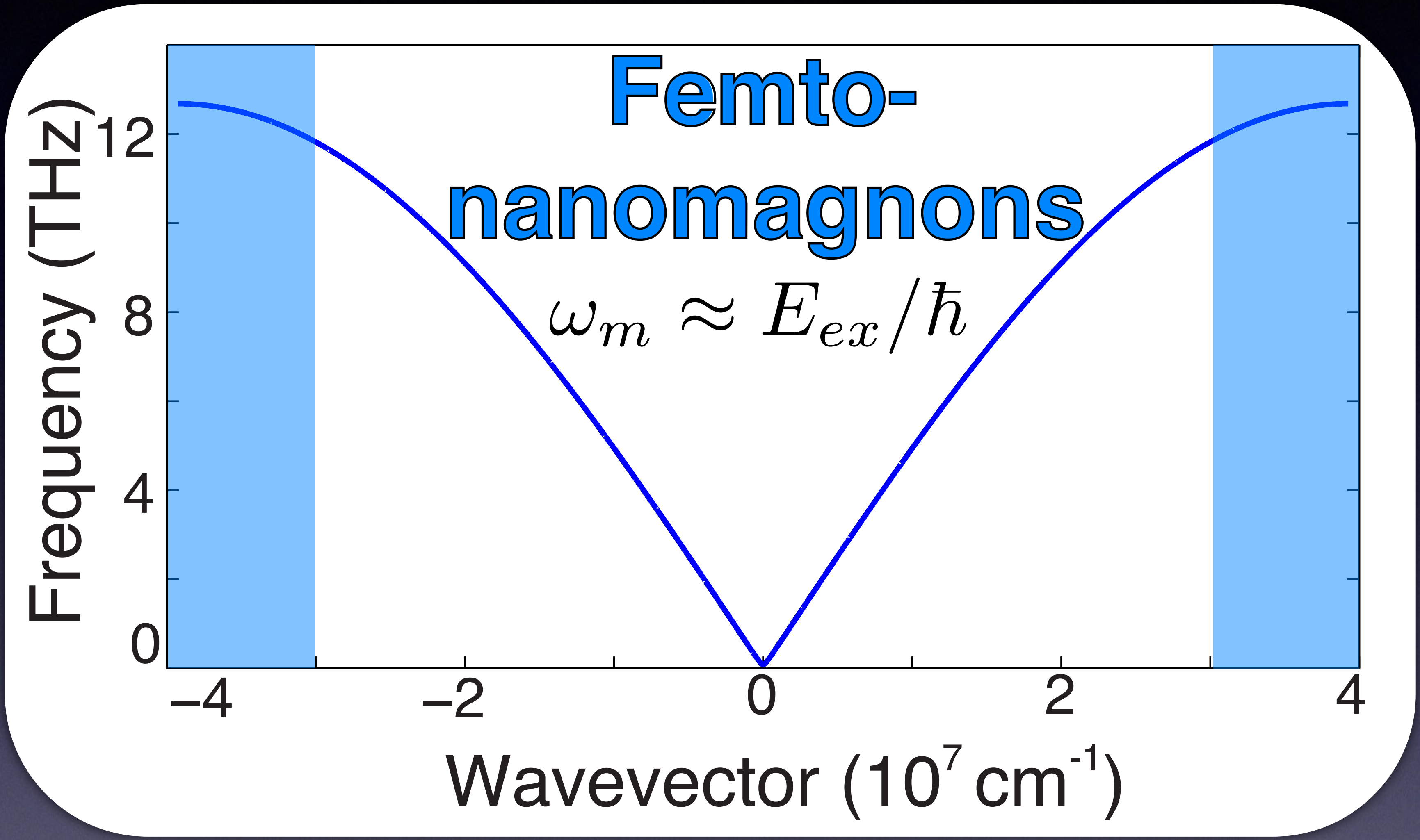


D.Bossini et al PRB. **89** (R), 060405 (2014)

- ✓ No electrons are photo-excited
- ✓ T_M increases only via magnetic interactions

Zero Absorption Criterion: $\tau_d = \tau_r$

Magnon dispersion in AF

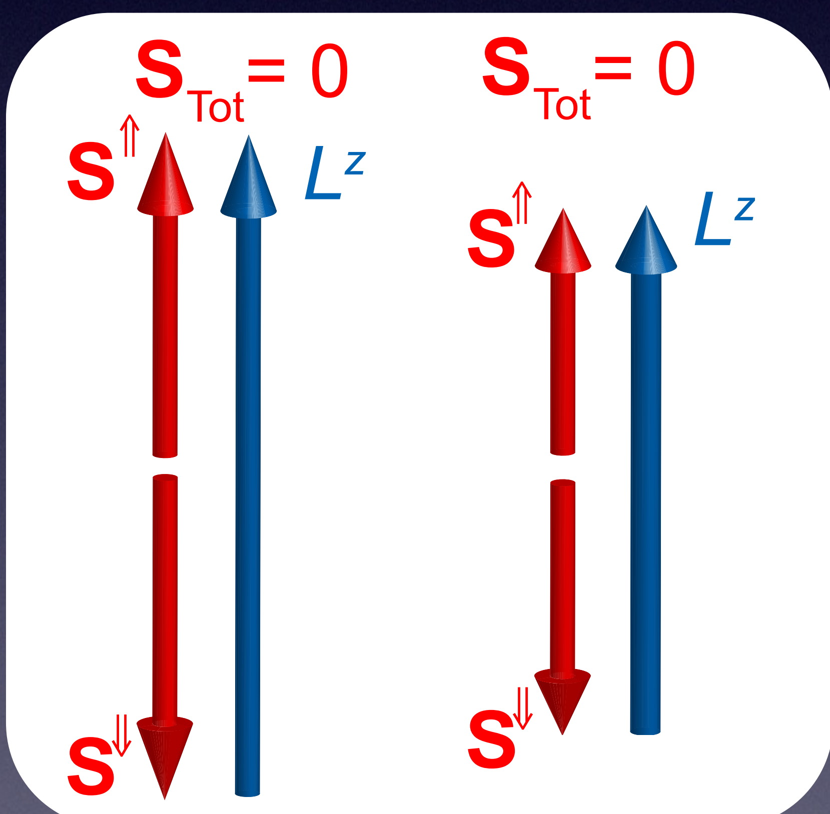


- ✓ Femtosecond period
- ✓ Nanometer wavelength
- ✓ Defined by E_{ex}

Measure the macrospin dynamics triggered by femto-nanomagnons

Generation

Problem: high-wavevector magnons are usually unaccessible

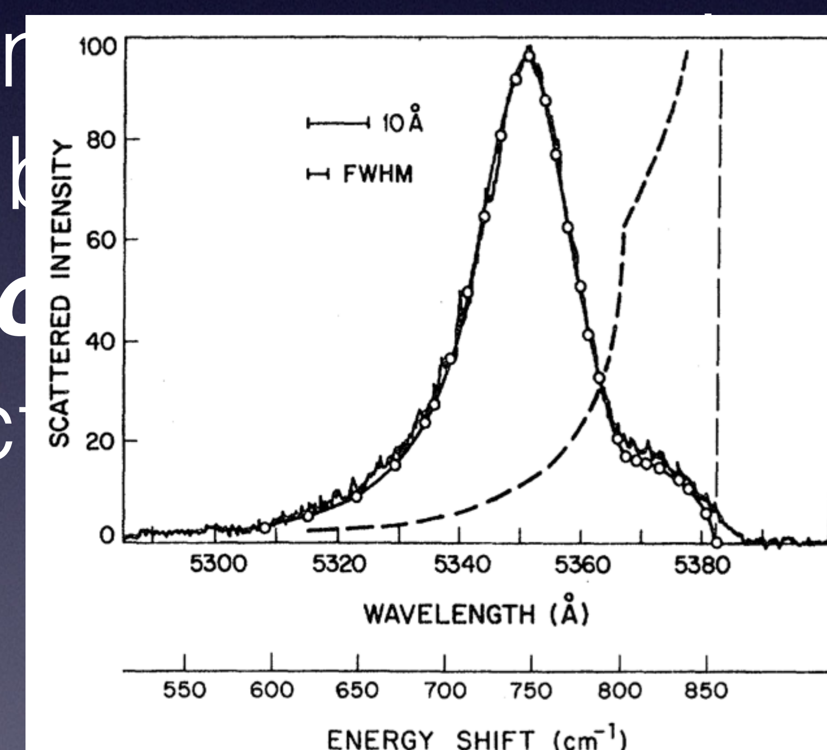


$$L \equiv S^{\Uparrow} - S^{\Downarrow}$$

- ✓ Spin and momen
- ✓ Light-induced b
magnon pair: *two*
- ✓ High-wavevect

$KNiF_3$

$$E_{2M} = E_{ex} + \Delta$$



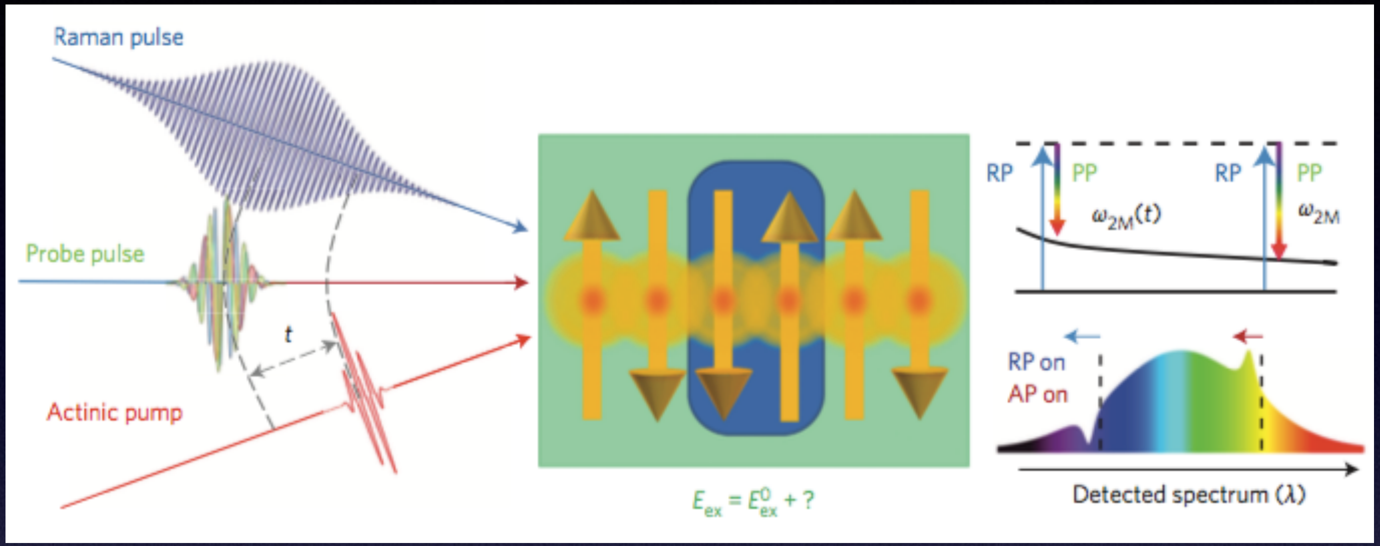
S. Chinn et al. PRB **3**, 1709 (1971)

U. Balucani et al. PRB **8**, 4247 (1973)

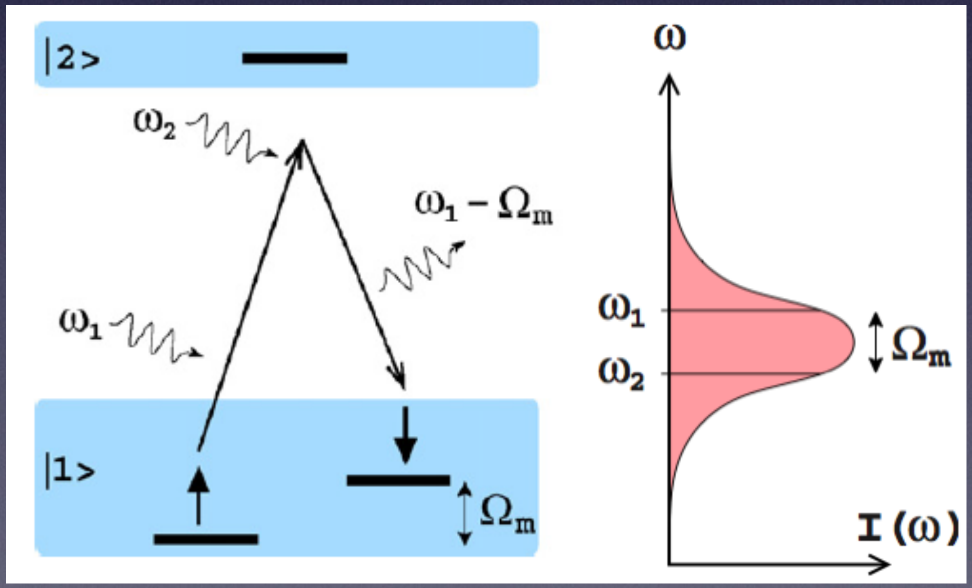
Two options

G. Batignagni, D.Bossini et al Nat. Phot. **9,** 506 (2015)

Time-resolved stimulated Raman spectroscopy



ISRS

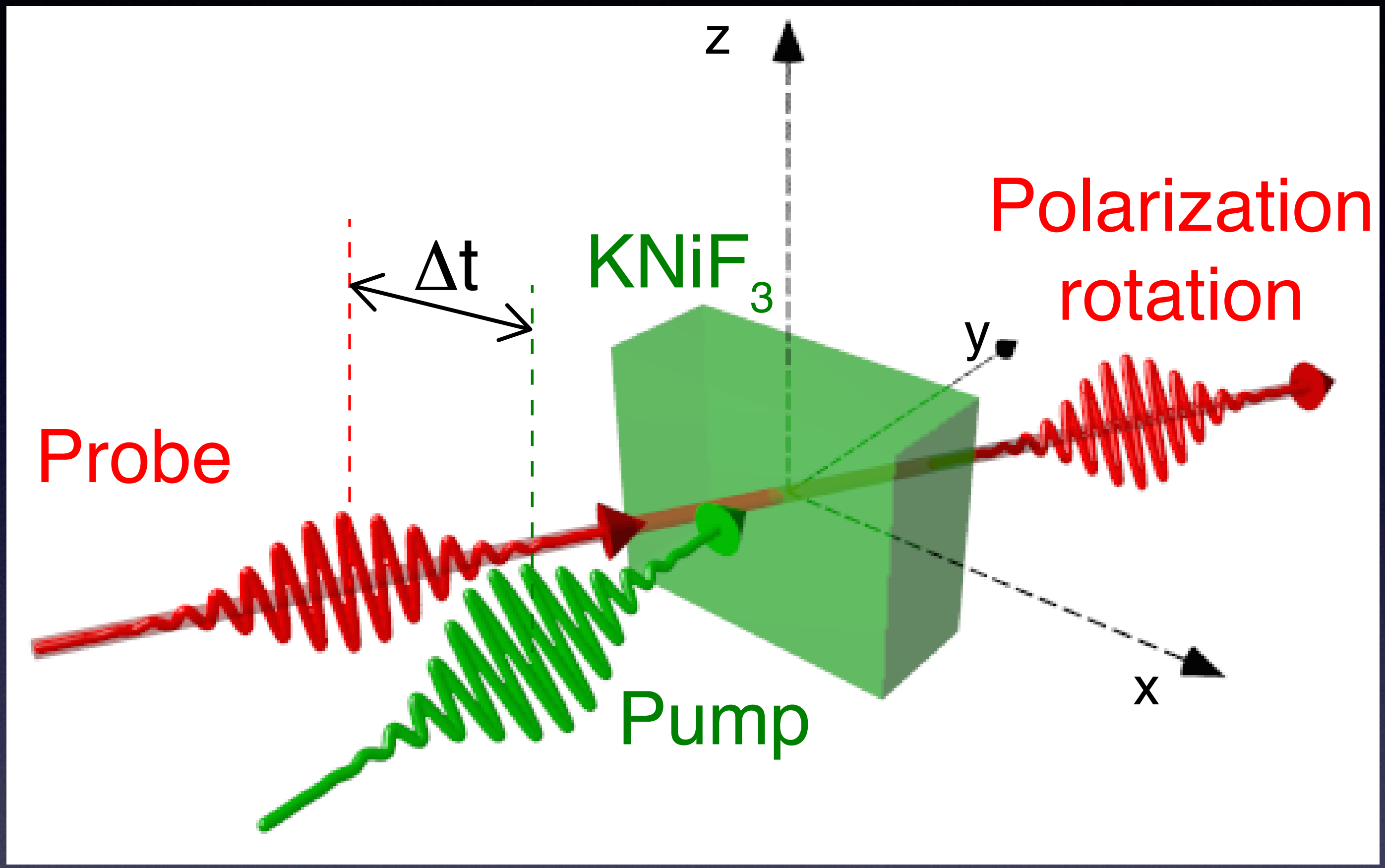


2M period in $KNiF_3$: 45 fs

Time domain → Pulses shorter than period

10 fs laser pulses

Detection



Pump-probe technique

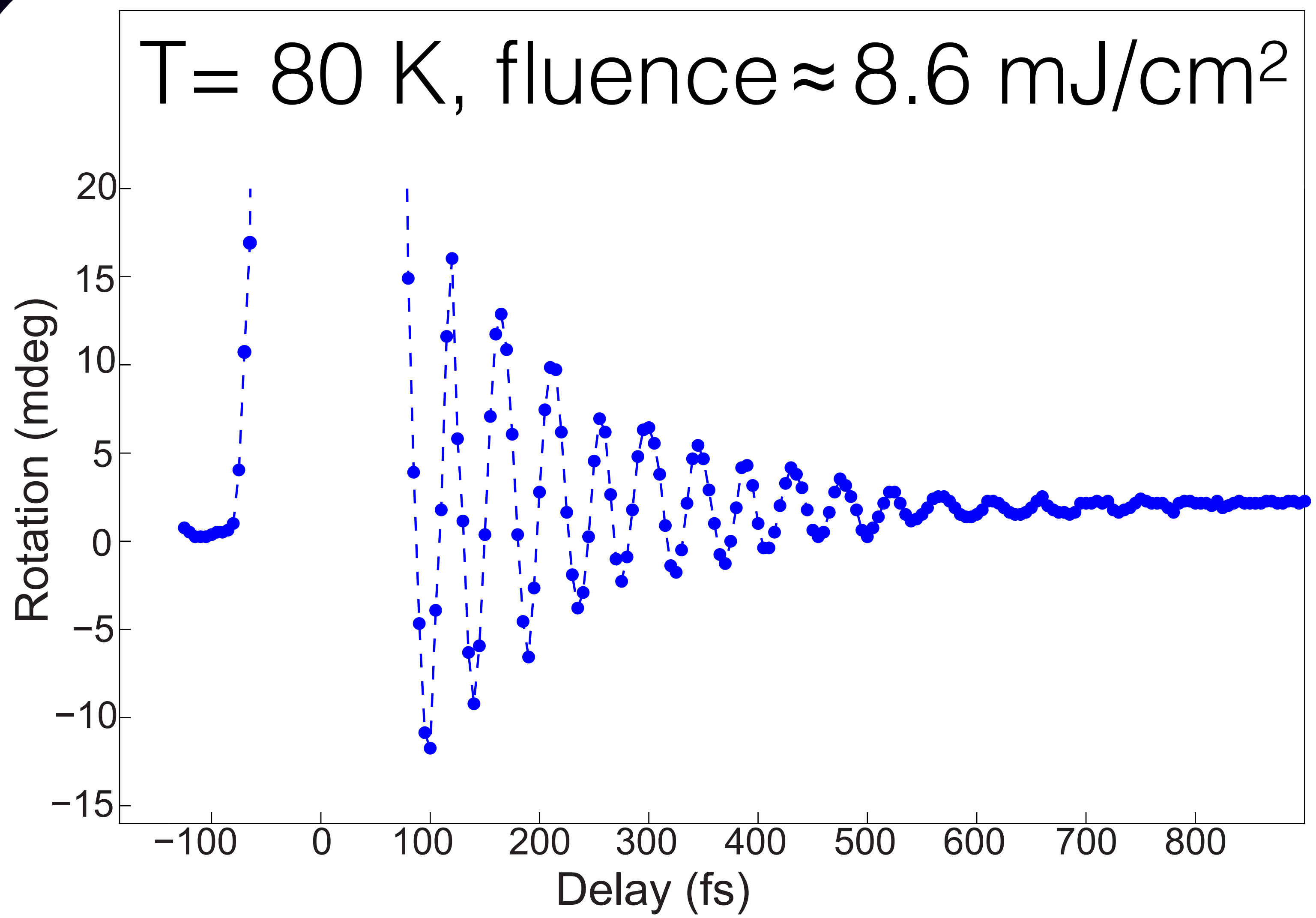
Magneto-optical response to the photo-excitation measured as a function of the delay

All-optical detection via a **second-order magneto-optical effect**

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

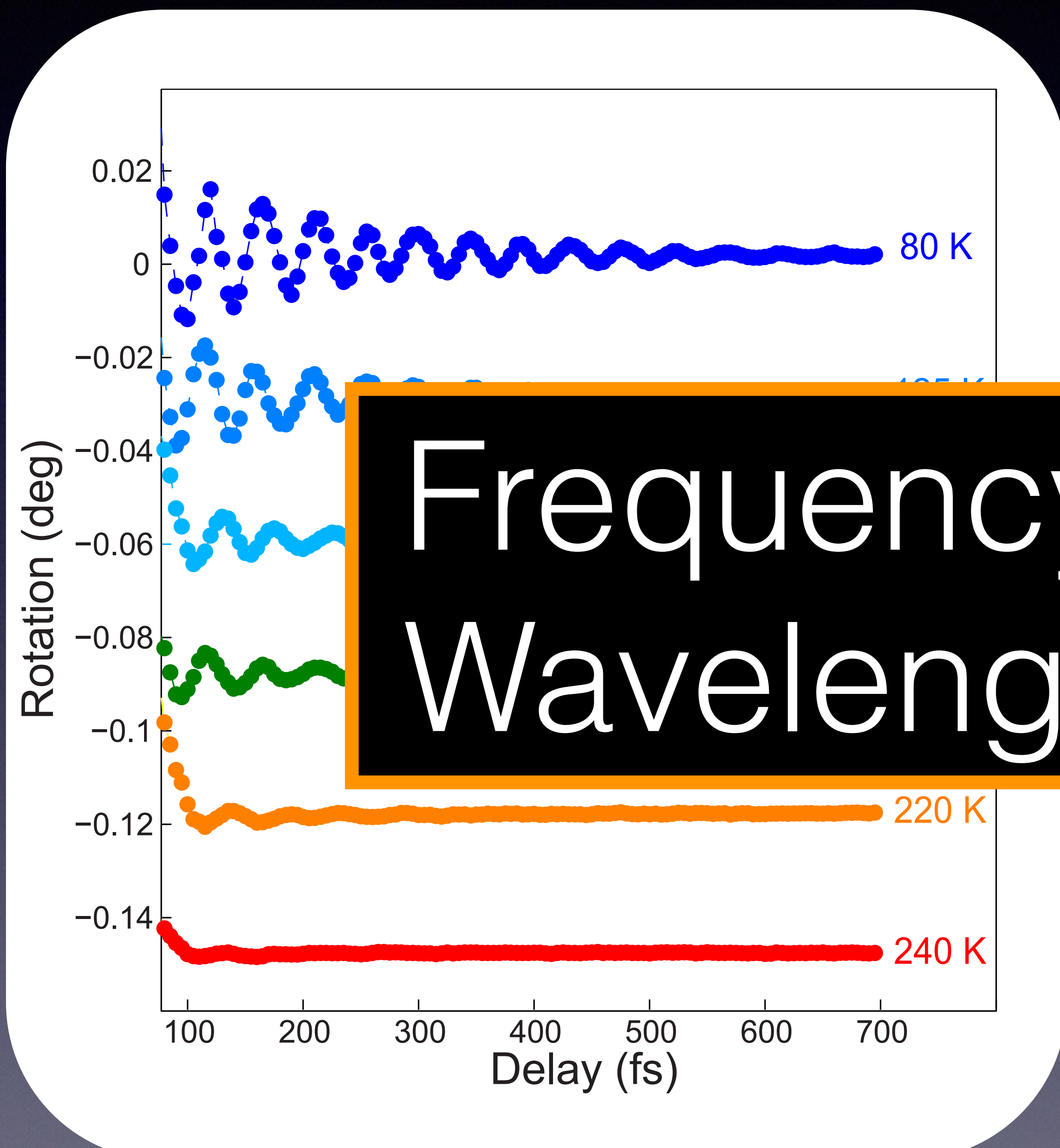
J. Ferrè *et al.* Rep. Prg. Phys **47**, 513 (1984)

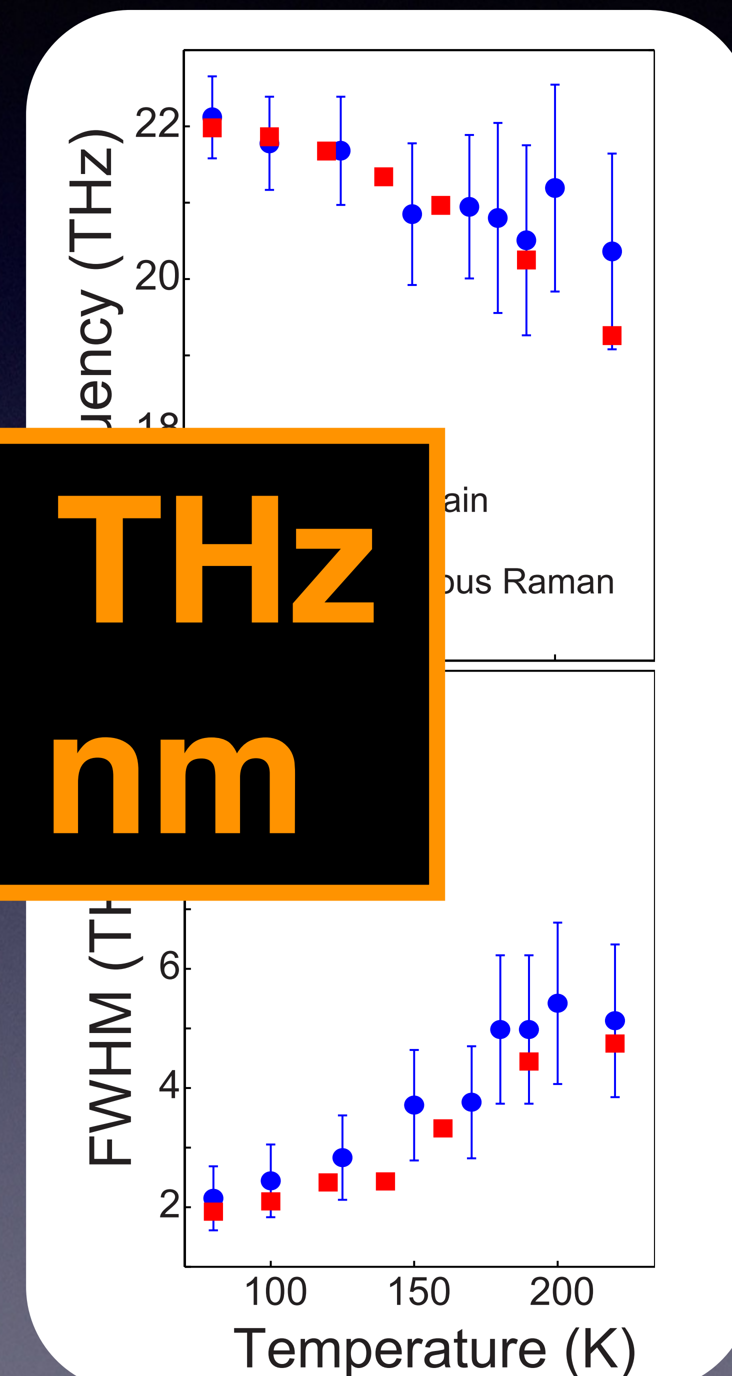
Laser-induced dynamics



- ✓ Pump and probe linearly and orthogonally polarized
- ✓ Oscillations @ 22 THz (T=45 fs)
- ✓ Lifetime ≈ 500 fs

Temperature dependence





Frequency: 22 THz
Wavelength: 1 nm

Model: approach

$$\hat{H} = \hat{H}_0 + \hat{H}_1$$

$$\hat{H}_0 = J \sum_{\langle i,j \rangle} \hat{\boldsymbol{S}}_i^{\Uparrow} \cdot \hat{\boldsymbol{S}}_j^{\Downarrow}$$

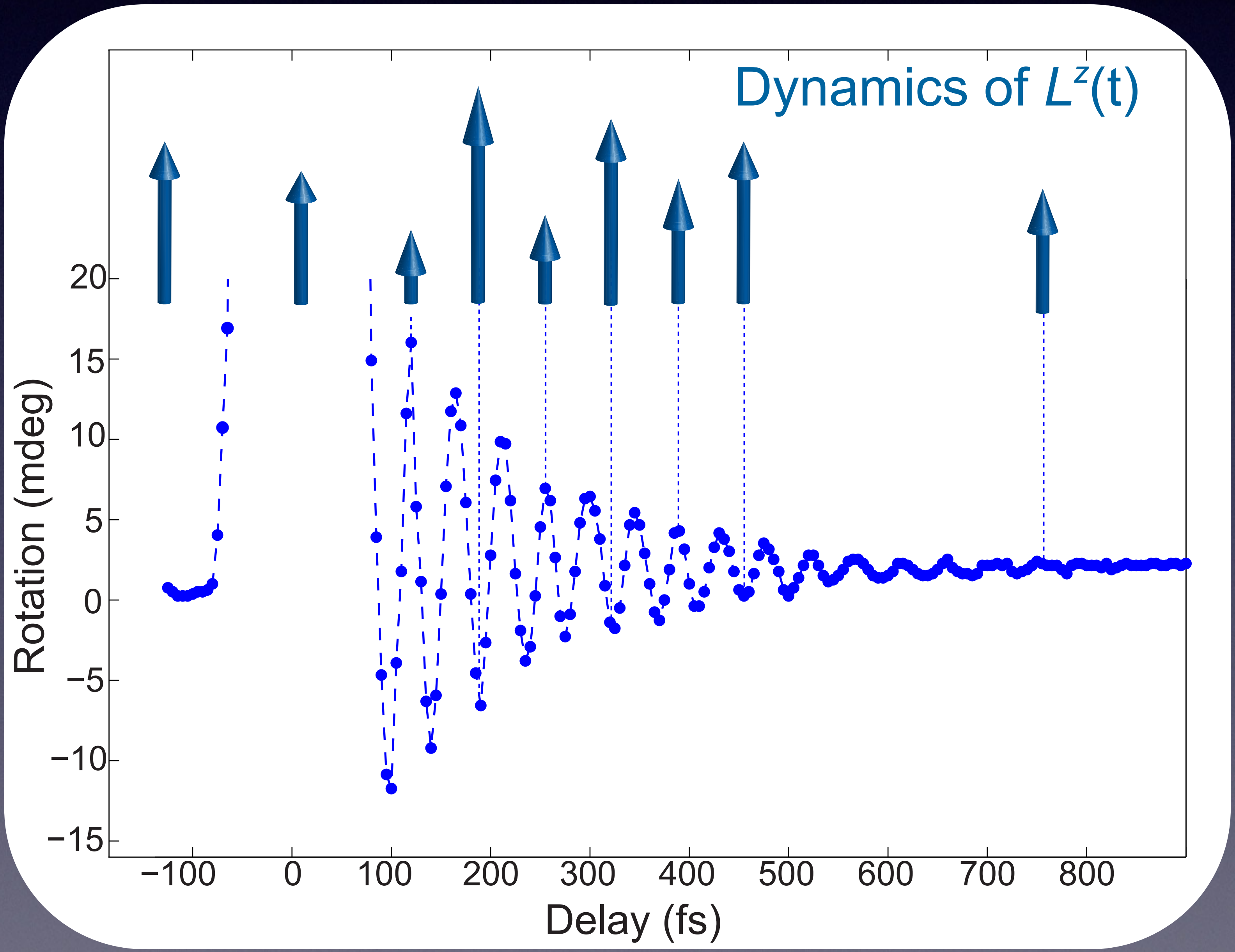
$$\hat{H}_1(t) = \delta(t) \frac{4\pi I_1}{n_R c} \sum_{\langle i,j \rangle} \Xi_{ij} \left(\frac{\hat{S}_i^{+\Uparrow} \hat{S}_j^{-\Downarrow} + \hat{S}_i^{-\Uparrow} \hat{S}_j^{+\Downarrow}}{2} + A \hat{S}_i^{z\Uparrow} \hat{S}_j^{z\Downarrow} \right)$$

$|\psi(t)\rangle$ → Expectation values

$\langle \hat{S}_i^{z\Uparrow} \hat{S}_j^{z\Downarrow} \rangle$ $L^z(t)$ **Same time-dependence**

Macrospin dynamics

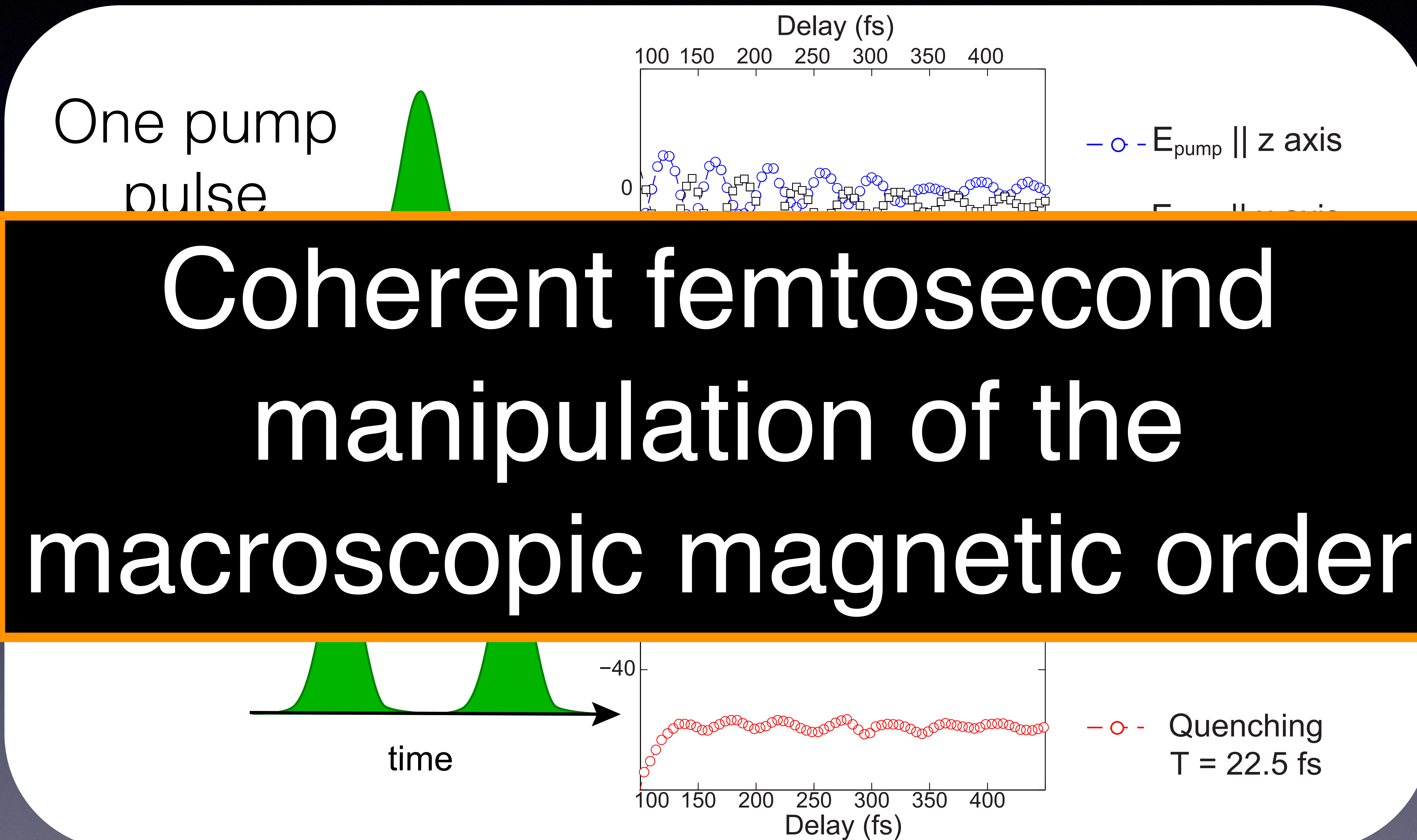
$$\langle \hat{S}_i^{z\Uparrow} \hat{S}_j^{z\Downarrow} \rangle \qquad L^z(t)$$



Same time-dependence

Macroscopic probe of the femtosecond dynamics of nanometer spin correlations

Control the dynamics



Coherent femtosecond manipulation of the macroscopic magnetic order

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

What's next

Dynamics induced by femto-nanomagnons

J. Zhao et al. PRL **93**, 107203 (2004)

$$\hat{H}_1(t) = \delta(t)\frac{4\pi I_1}{n_R c}\sum_{\langle i,j\rangle}\Xi_{ij}\left(\frac{\hat{S}_i^{+\Uparrow}\hat{S}_j^{-\Downarrow}+\hat{S}_i^{-\Uparrow}\hat{S}_j^{+\Downarrow}}{2}+A\hat{S}_i^{z\Uparrow}\hat{S}_j^{z\Downarrow}\right)$$

Symmetric in x-y plane

Precession forbidden: dynamics purely longitudinal

No classical equation of motions!

What's next

- ✓ Criteria for quantum mechanical effect ($KNiF_3$ VS K_2NiF_4)
- Pure manipulation of the phase, no classical counterpart !!
- ✓ Issues on the Néel point (K_2NiF_4)
- "Artificial" semiclassical equation of motion (generalized potential)

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D. Bossini et al. *in preparation*

Conclusions

1. Control of the photo-induced energy flow: zero absorption regime
2. Macroscopic probe of the femtosecond dynamics of E_{ex}
3. Coherent femtosecond manipulation of the magnetic order on the nanometer scale

Femto-nanomagnonics!

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

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

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