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# *magnetism and light*

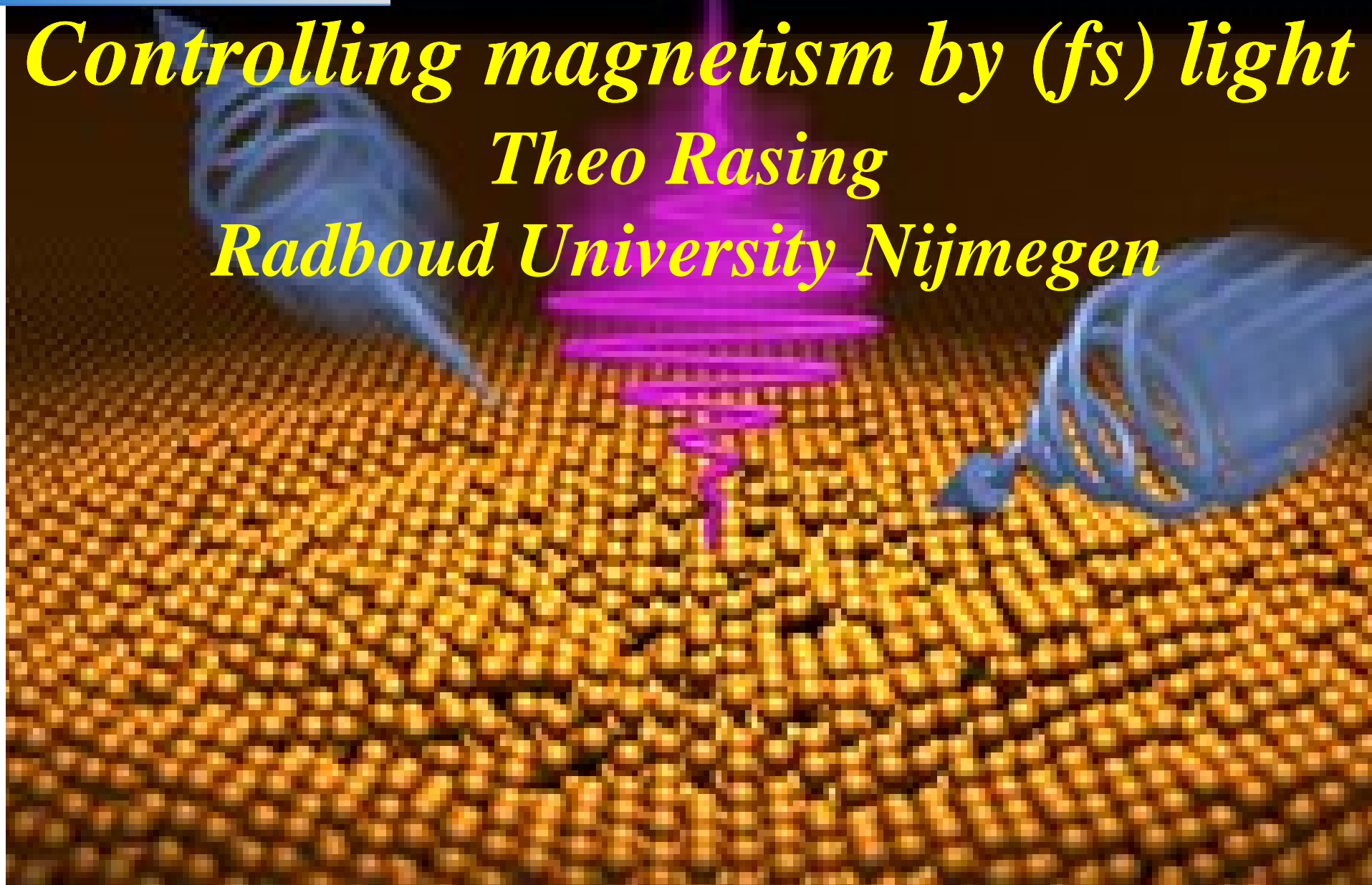


# *fast opto-magnetism*

## *Controlling magnetism by (fs) light*

*Theo Rasing*

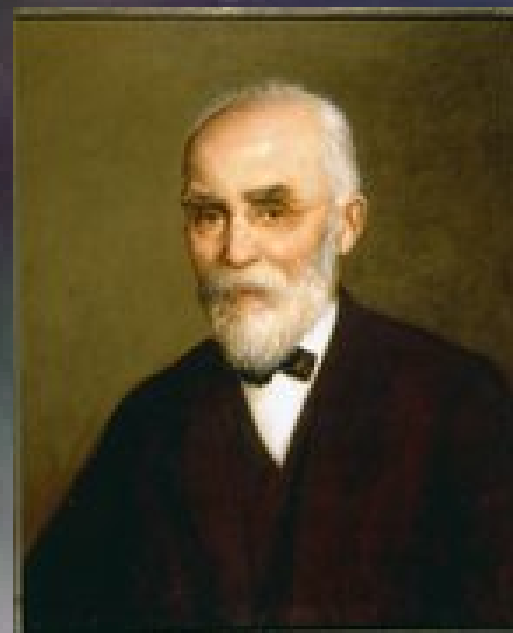
*Radboud University Nijmegen*



# Magnetism and light



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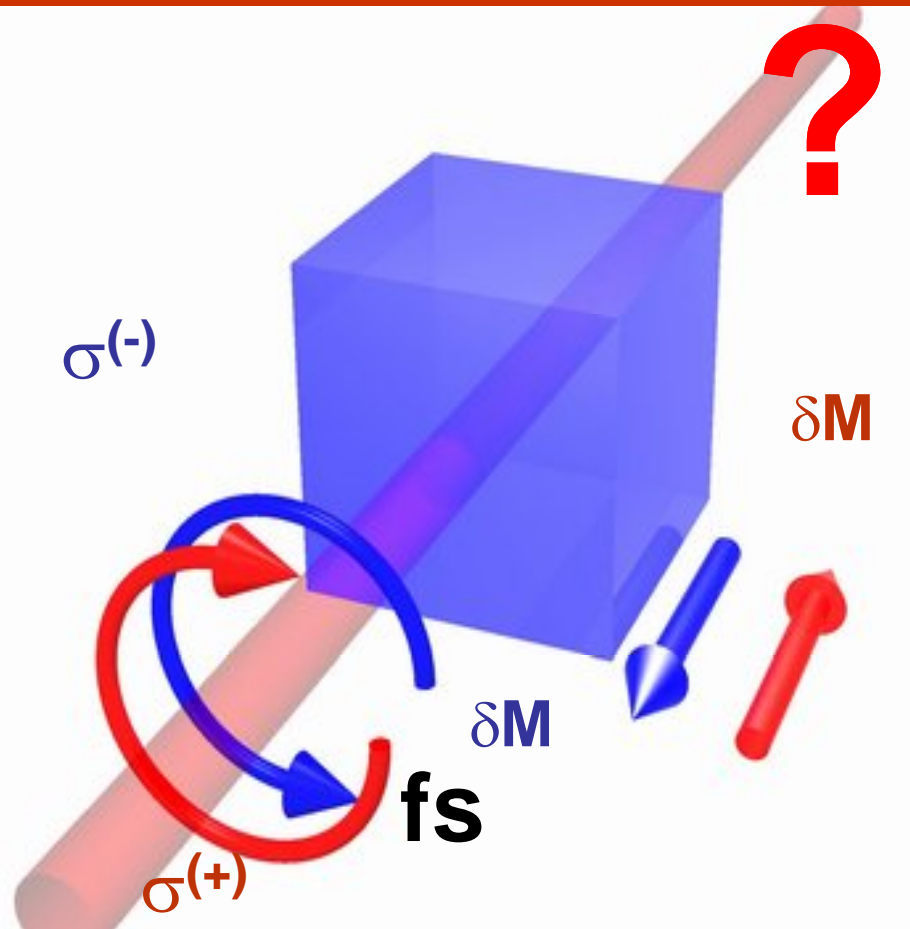


002

ary service they rendered by  
ce of magnetism upon  
(with Lorentz)



inverse?

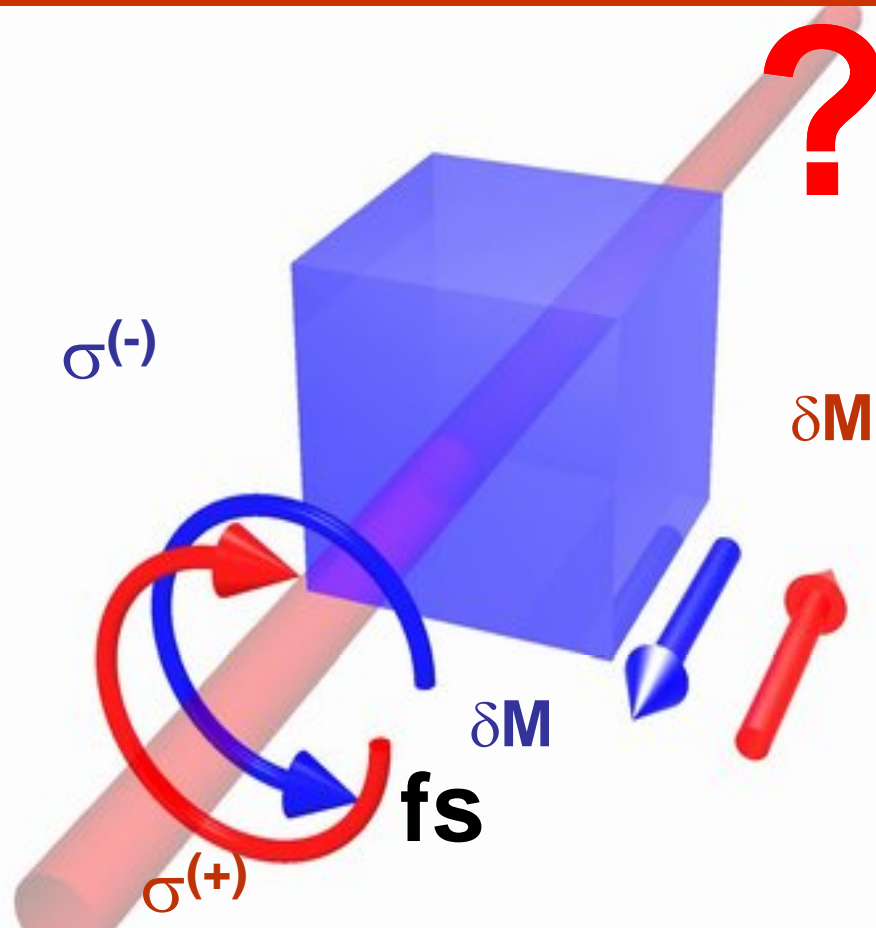


Faraday effect



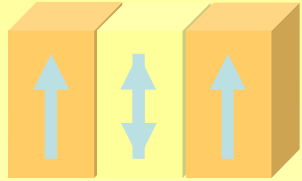


# opto-magnetism

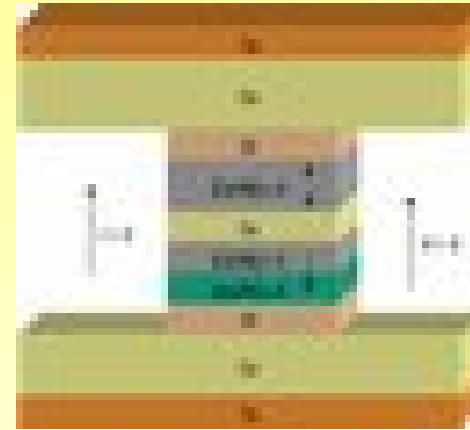


# magneto-optics

# Magnetization reversal



magnetic field



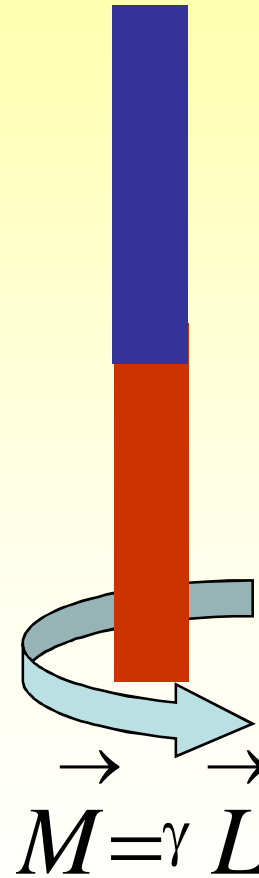
spin-polarized current

**both slow!**

$\sim 1\text{ns}$

## de Haas effect

# Magnetism and angular momentum



A. Einstein & W.J. de Haas,  
*Experimenteller Nachweis der Amperèschen Molekülströme,*  
 Verhandl. Deut. Phys. Ges. **17**, 152. 170 (1915).

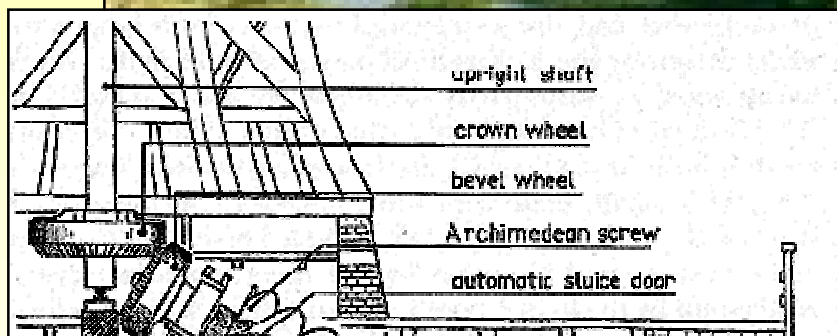


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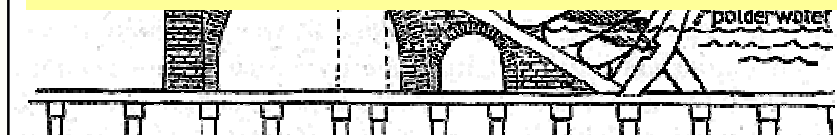
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# *Quality behind the Dykes*





**In traditional windmills the wings are connected to a wooden axle**



**Traditional windmills rotate counter-clockwise**

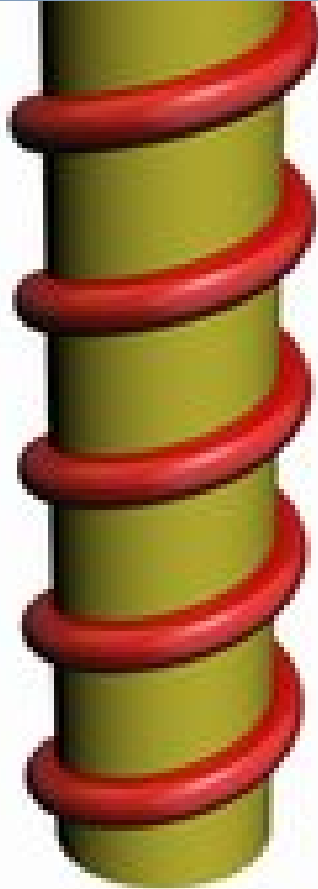
**Modern windmills rotate clock-wise !**



**Experience:  
counter-clock rotation increases lifetime of axle !**

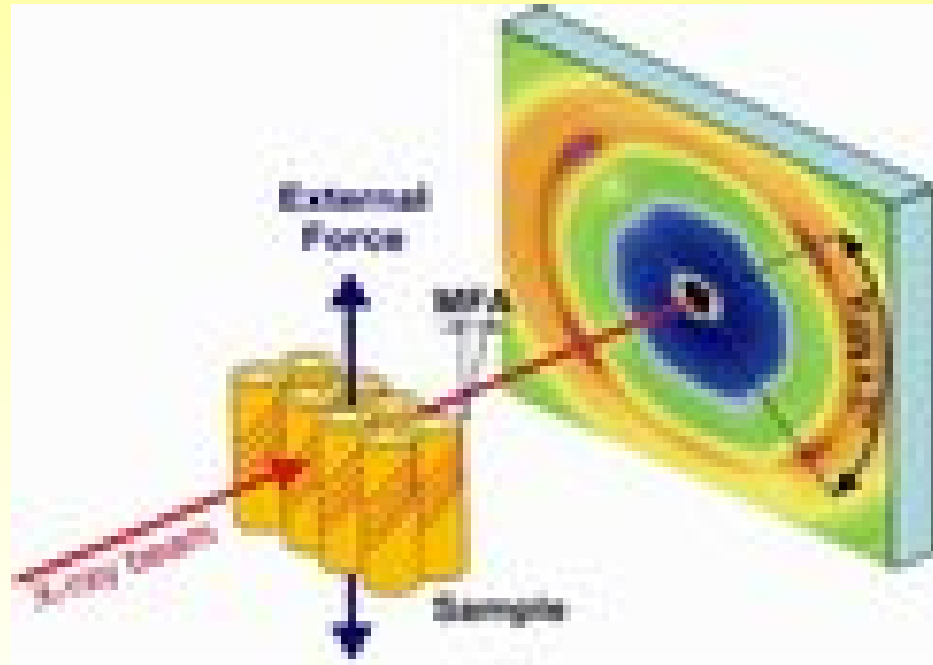
Lignin matrix

In situ wide-angle X-ray scattering (WAXS) on individual wood cells



Cellulose fibrils

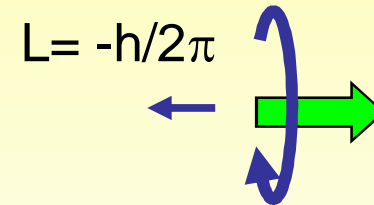
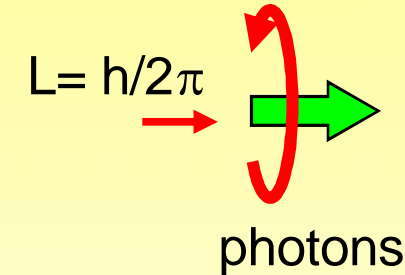
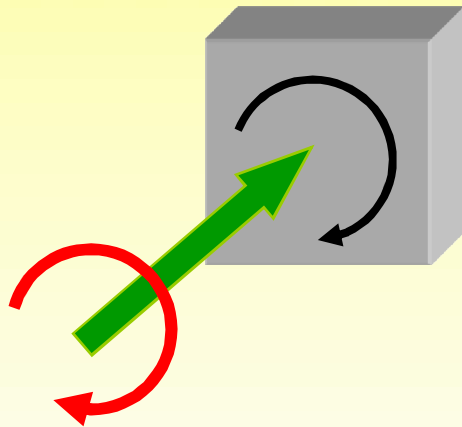
Wood cell



J. Keckes et al., Nature Materials 2, 811 (2003)

# momentum of photon !

## Beth's experiment



# Transfer of chiral information !

1998, 19, 1198

PHYSICAL REVIEW

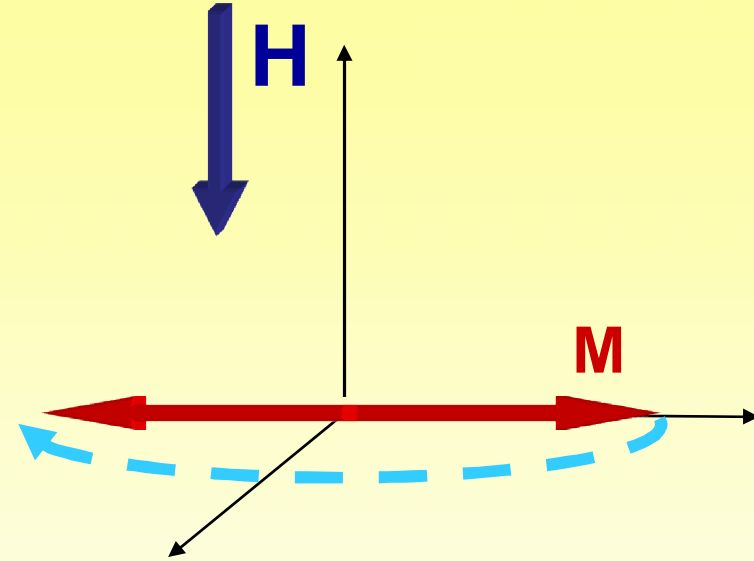
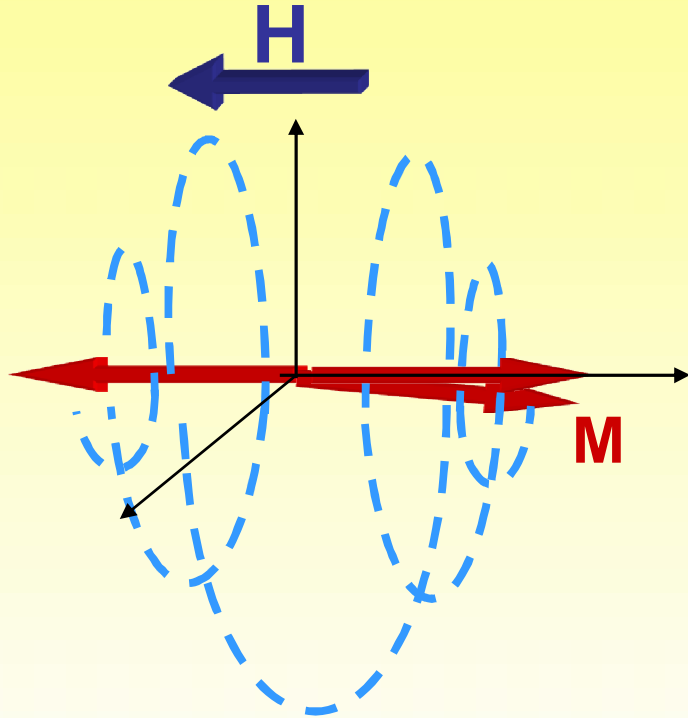
VOLUME 19

## Mechanical Detection and Measurement of the Angular Momentum of Light

Richard A. Beth,<sup>1</sup> *Worcester Polytechnic Institute, Worcester, Mass.* and *Palmer Physical Laboratory, Princeton University*

(Received May 1, 1998)

# Methods and time-scales of magnetization reversal



Th. Gerrits et al., *Nature* **418**, 509 (2002).  
S. Kaka, S. E. Russek, *Appl. Phys. Lett.* **80**, 2958 (2002).  
H. W. Schumacher et al., *Phys. Rev. Lett.* **90**, 017201 (2003).

**Reversal time ~nanoseconds**

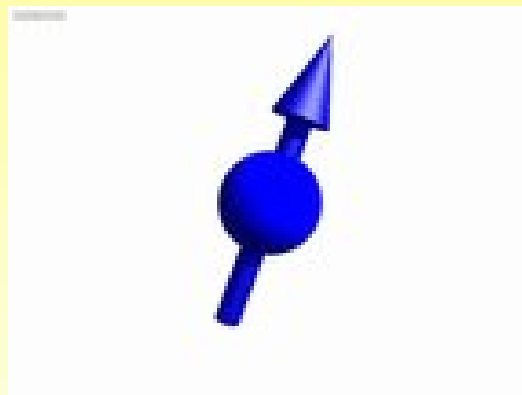
Reversal time is determined by damping (ns) and spin-lattice relaxation  
~ 100 ps

**The shortest time achieved is 100 ps**

**Faster?**



# Spin dynamics?



$$\gamma = g \cdot \frac{e}{2mc} = 2.8 \text{ GHz/kG}$$

100ps  $\longrightarrow$  0.36T

1ps  $\longrightarrow$  36T



34 Tesla →



# cales in magnetism

1 ns

100 ps

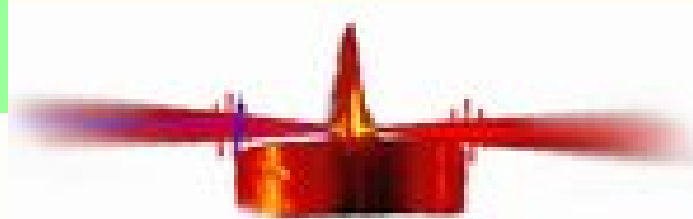
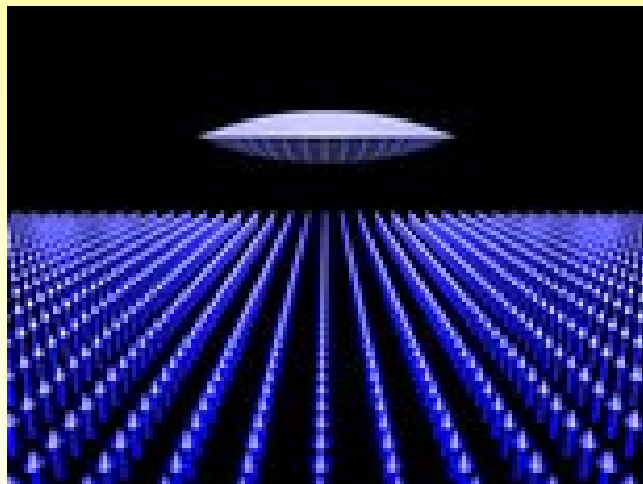
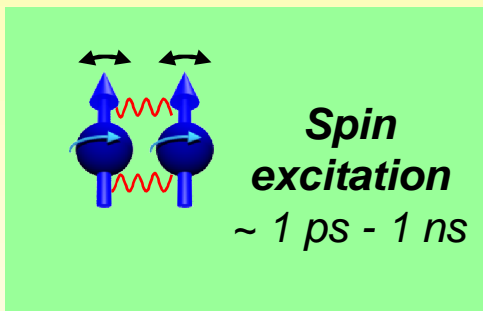
10 ps

1 ps

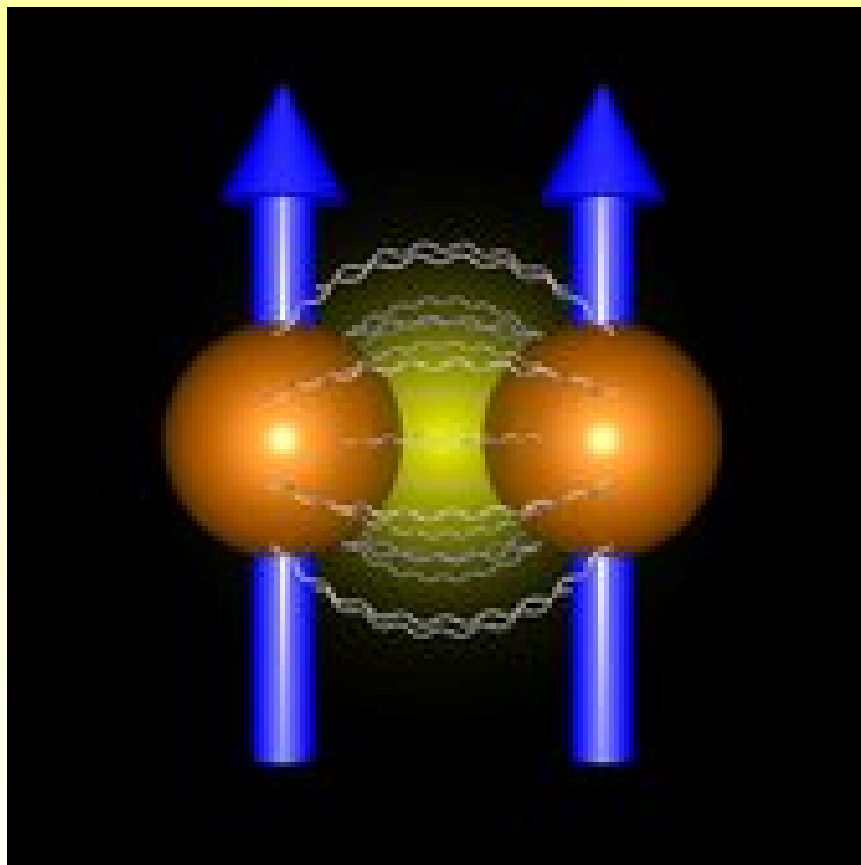
100 fs

10 fs

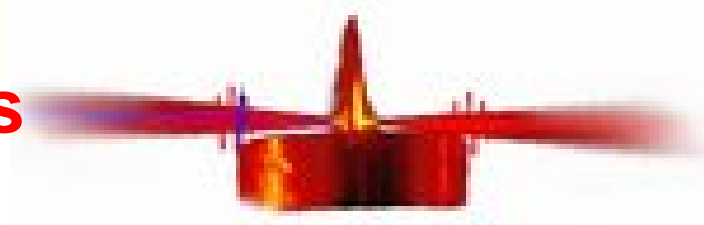
1 fs



# How = Mechanisms?



1-2eV, 10-100fs



# antosecond camera

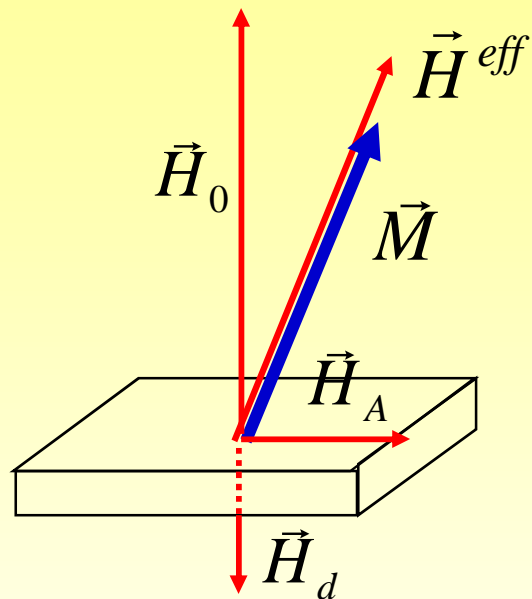


# Induced spin dynamics

of the measurements

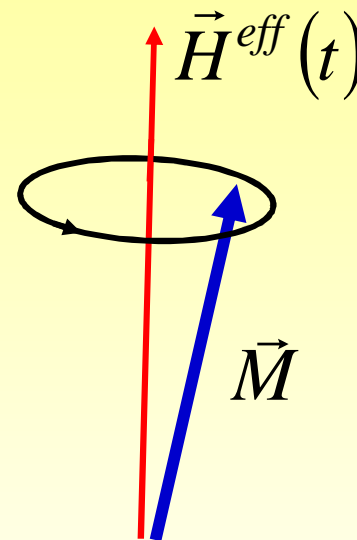
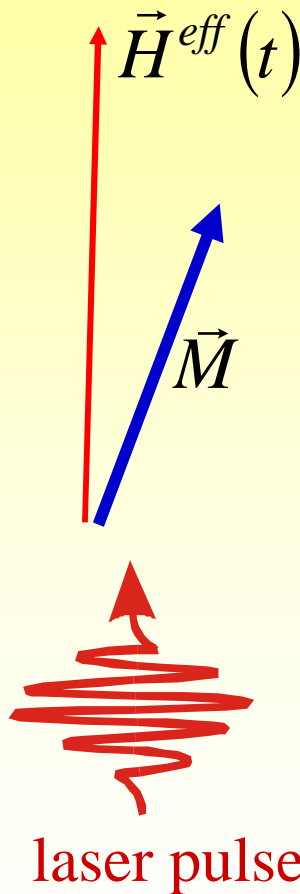
Ju et al., PRL **82**, 3705 (1999)

van Kampen et al., PRL **88**, 227201 (2002)



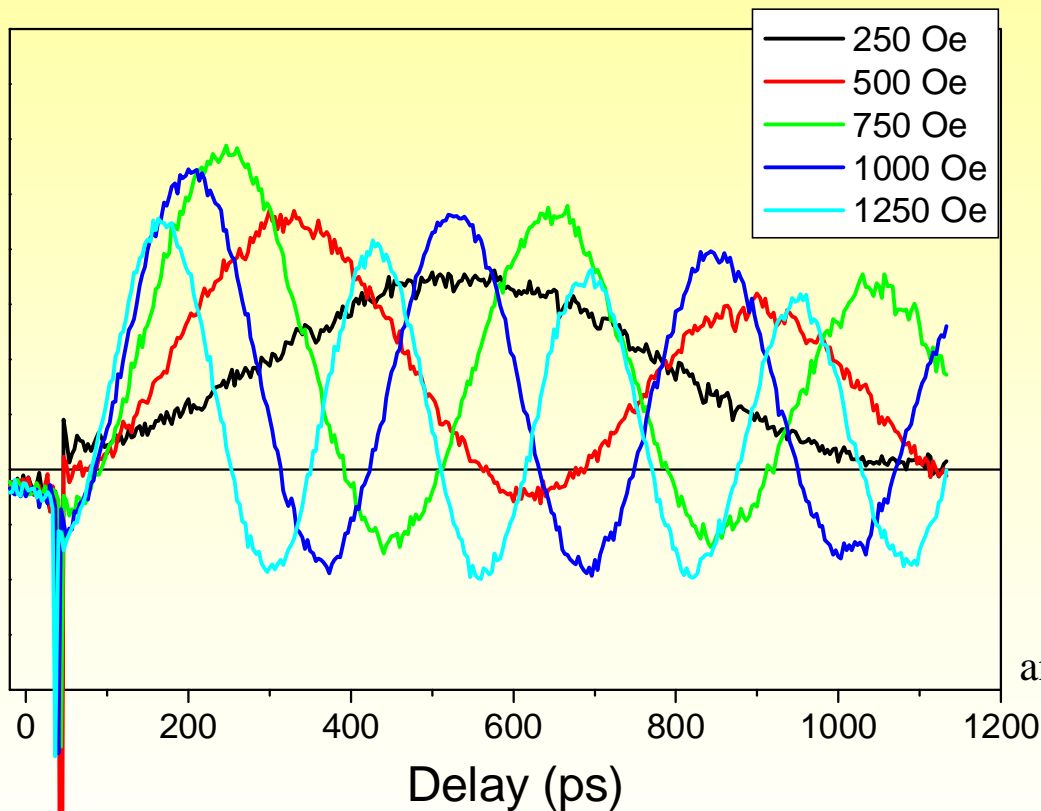
$$\vec{H}^{eff} = \vec{H}_0 + \underbrace{\vec{H}_A + \vec{H}_d}_{\text{laser-affected}}$$

↑  
applied field

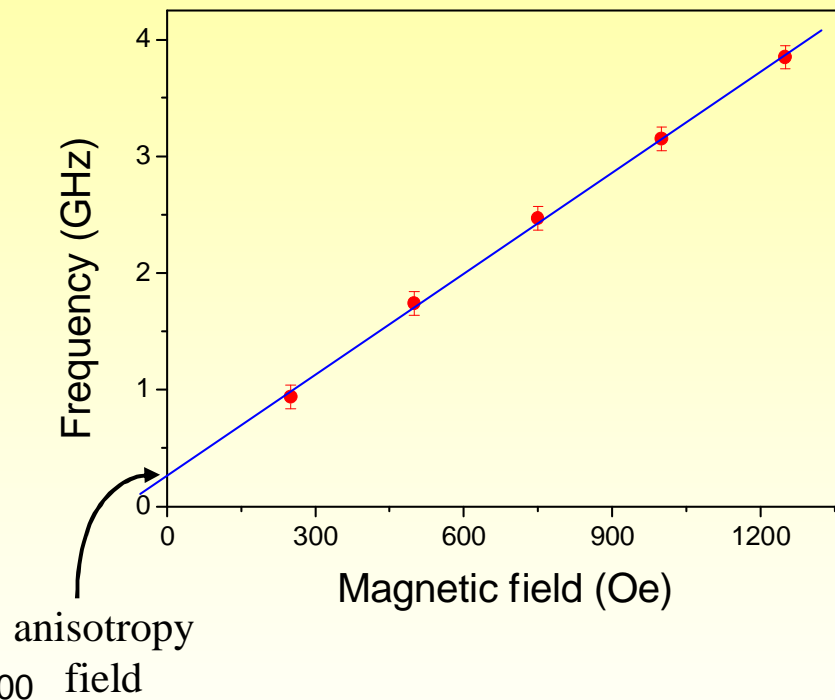


# Induced spin dynamics

thin film of magnetic garnet ( $\sim Y_3Fe_5O_{12}$ )



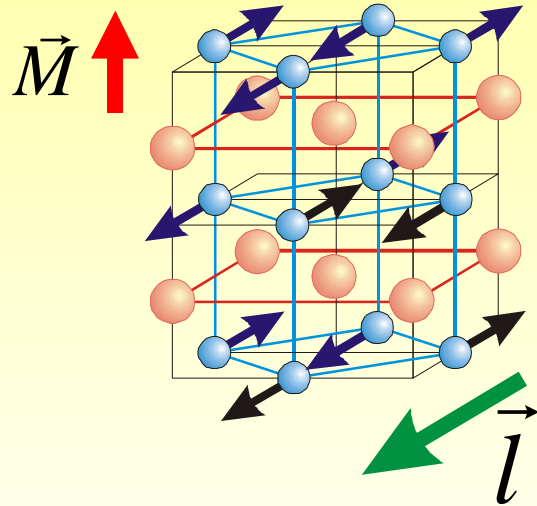
$$\omega = \gamma(H_A + H_0)$$



**~GHz**

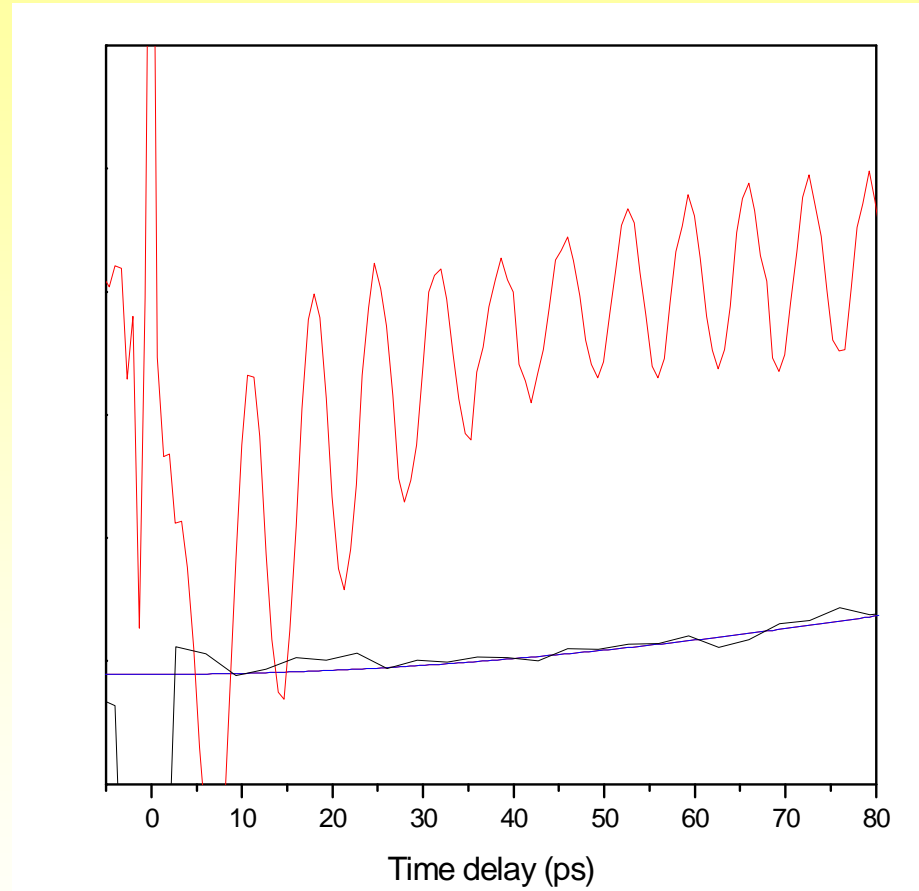
# Weak ferromagnet

**Orthoferrites (RE)FeO<sub>3</sub>**  
(canted antiferromagnets)



$$4\pi M \approx 200 \text{ G}$$

low damping  
large magneto-optical effects

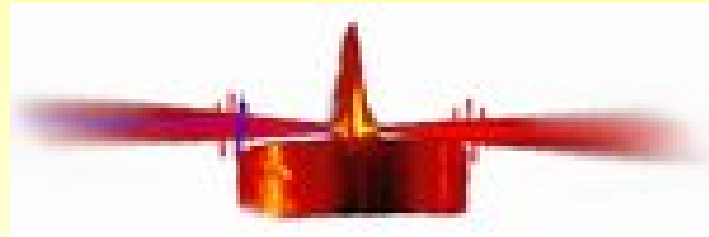


Kimel et al., Nature **429**, 850 (2004)

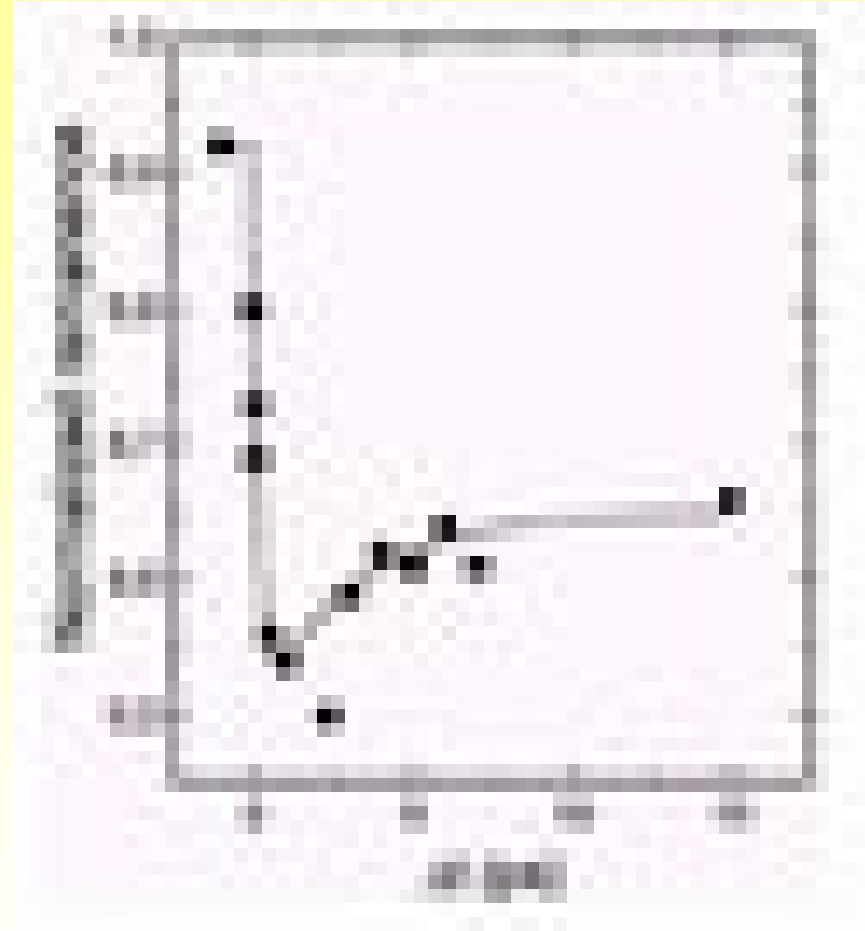
**Period: 6 ps vs 550 ps in garnets !**



# repaire et al:



60fs

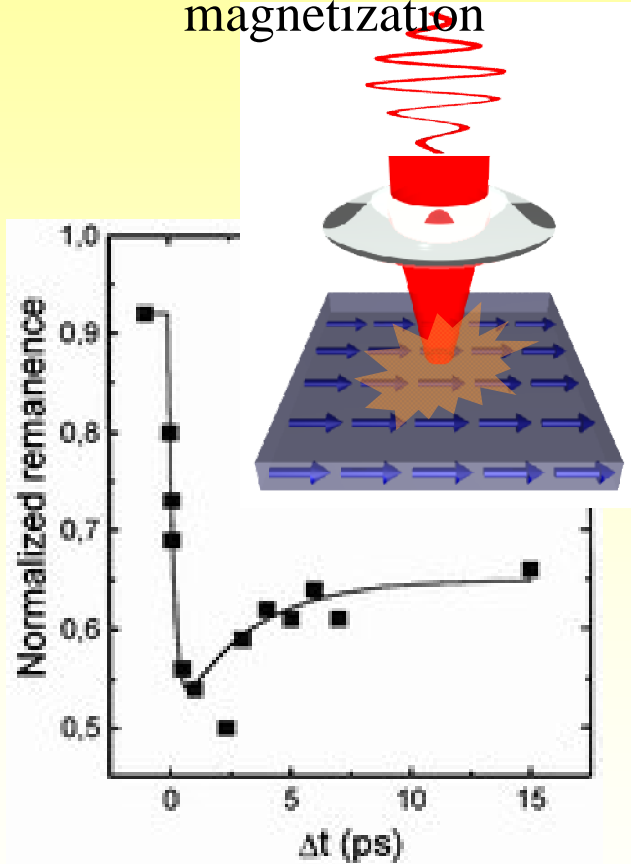


**Magnetization is changed within a picosecond!!!**

**How is that possible?**

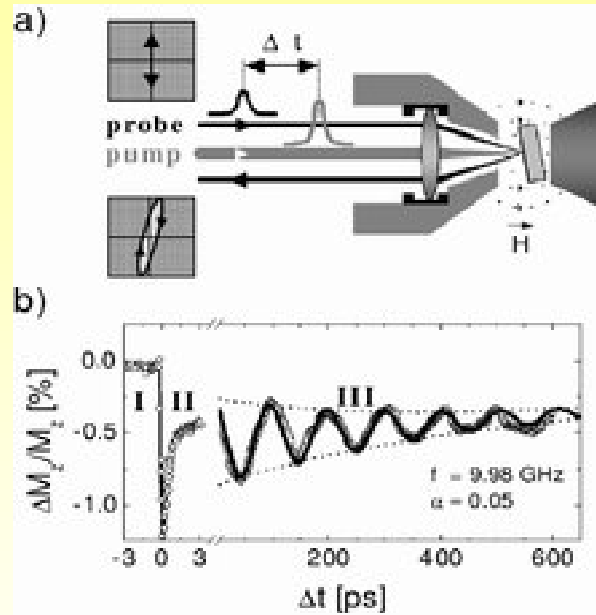
# laser-induced effects

laser-induced collapse of magnetization



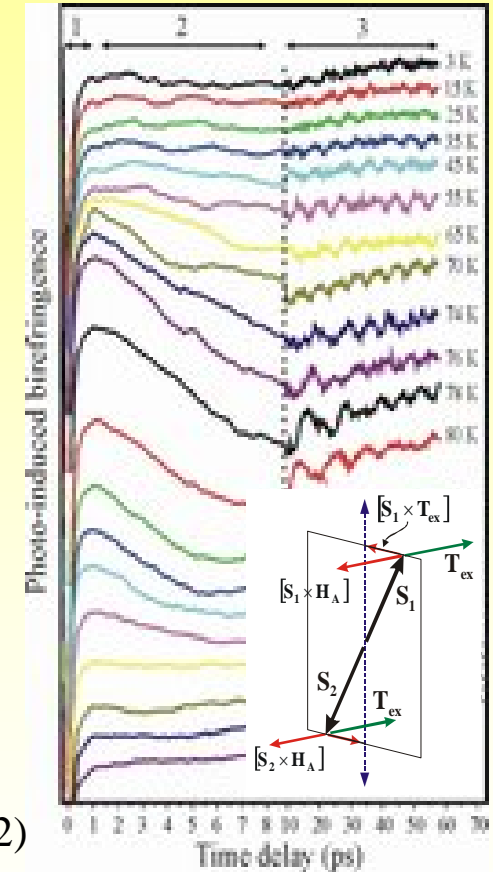
Beaurepaire et al, PRL **76**, 4250 (1996)

excitation and study of spin waves



Ju et al., PRL **82**, 3705 (1999)  
van Kampen et al, PRL **88**, 227201 (2002)

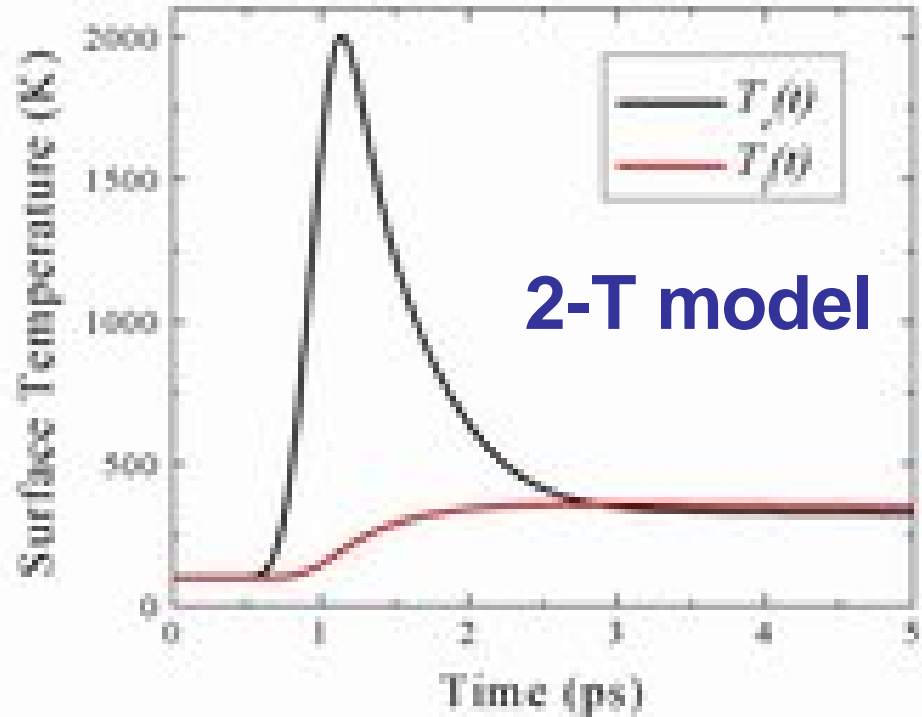
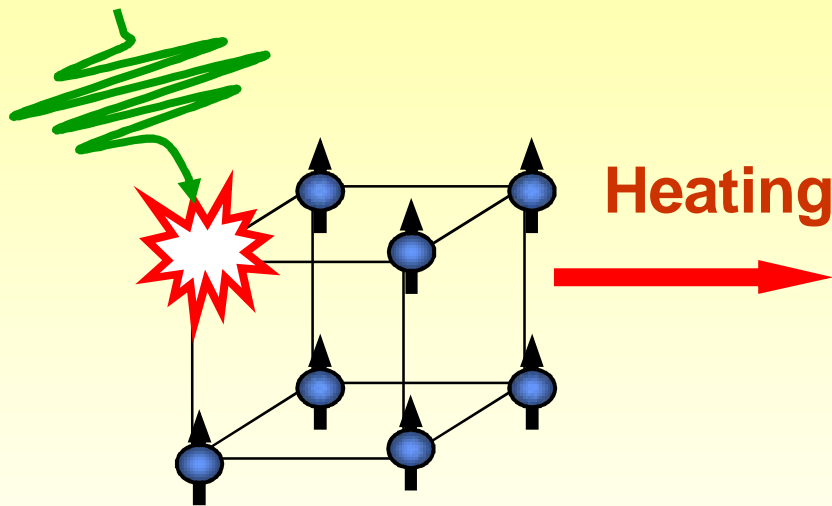
ultrafast phase transitions



Kimel et al., Nature **429**, 850 (2004)  
Ju et al, PRL **93**, 197403 (2004)  
Thiele et al, APL **85**, 2857 (2004)  
Kimel et al, Nature Phys **5**, 727 (2009)

# ed dynamics: Excitation mechanism?

## Ultrashort laser pulse



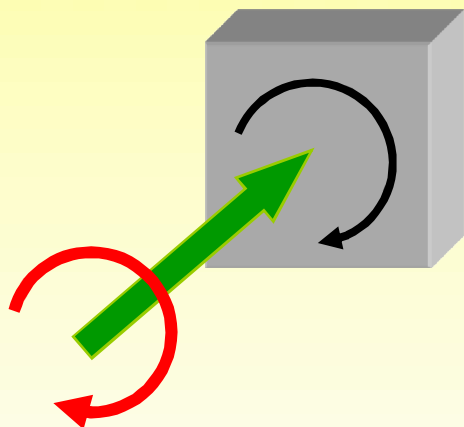
## Solution:

(only quenching !)

**A nonthermal effect of photons on spins**

# momentum of photon !

## Beth's experiment



$$L = h/2\pi$$
A green arrow points to the right. A red circular arrow around it indicates clockwise rotation. A small red arrow points to the left.

photons

$$L = -h/2\pi$$
A green arrow points to the right. A blue circular arrow around it indicates counter-clockwise rotation. A small blue arrow points to the left.

1936, 19, 1048

PHYSICAL REVIEW

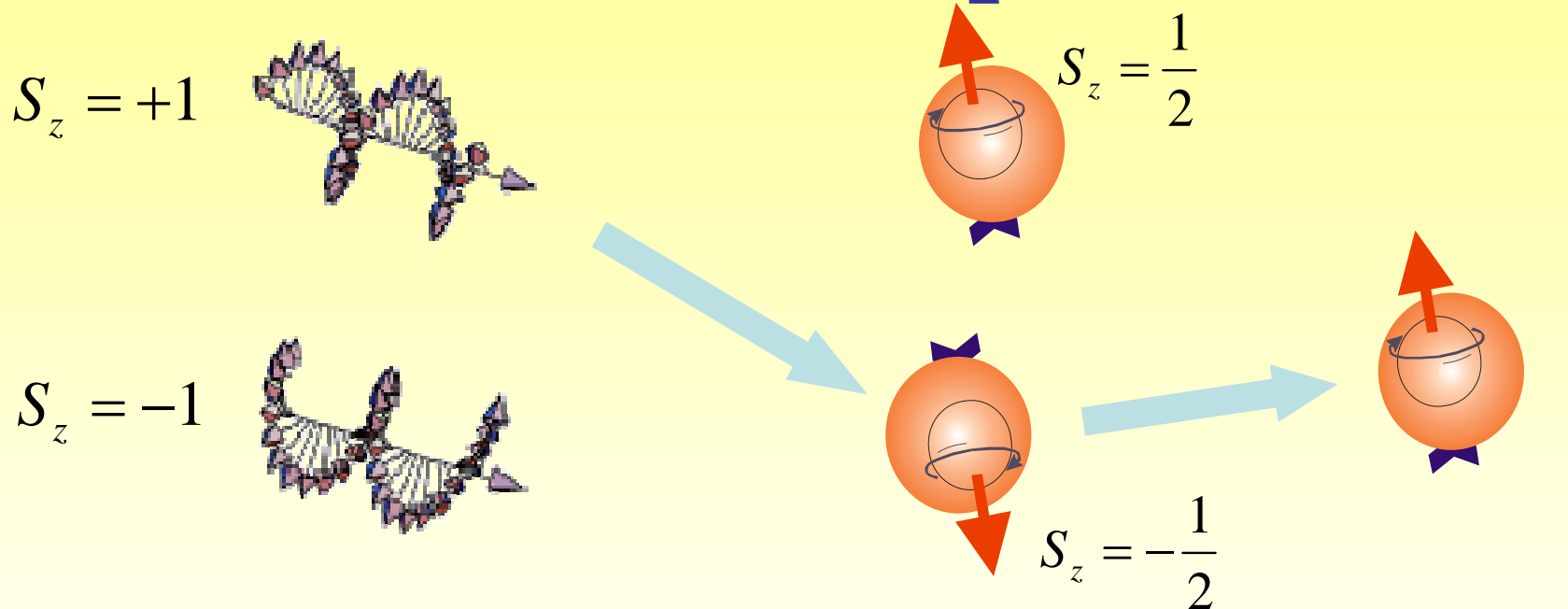
VOLUME 43

### Mechanical Detection and Measurement of the Angular Momentum of Light

LEONARD A. BETH,<sup>1</sup> *Wentworth-Polytechnic Institute, Worcester, Mass. and Palmer Physical Laboratory, Princeton University*

(Received May 1, 1936)

# polarization, photon spin, and absorption

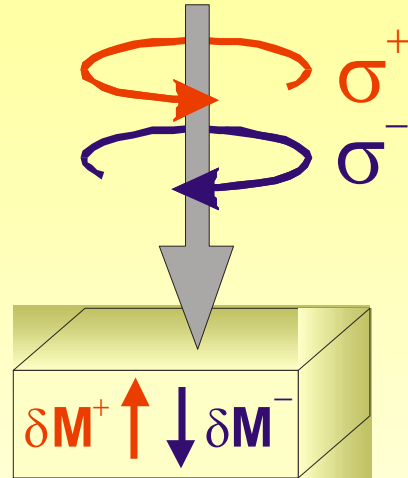


1 photon / site = 20000 K  $\Delta T$

$\left. \begin{array}{l} \sim 0.01 \text{ phot/site max} \\ \sim 0.01 \text{ efficiency} \end{array} \right\} \text{effect } \sim 10^{-4}$

~~very fast and easy?~~

# ions magnetize a medium ?



Yes, they can!!!

$$\mathbf{M} = \frac{\epsilon_0}{\mu_0} a [\mathbf{E} \times \mathbf{E}^*]$$

inverse Faraday effect !

L.P.Pitaevski: Electric forces in a transparent dispersive medium, Sov.Phys.JETP 12,1008-1013 (1961)

J.P.van der Ziel,P.S.Pershan&L.D.Malmstrom: Optically induced magnetization resulting from the inverse Faraday effect,Phys.Rev.Lett.15,190-193(1965)

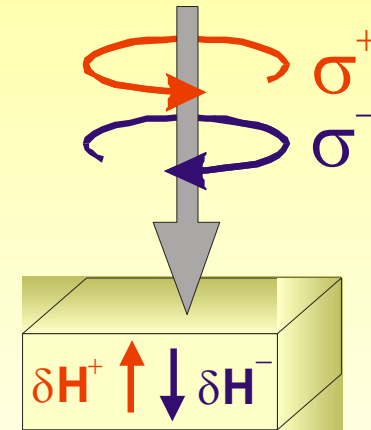
# Dynamics

$$W = \epsilon \epsilon_0 E(\omega) E^*(\omega)$$

$$H(0) = -\frac{1}{\mu_0} \frac{\partial W}{\partial M(0)} = -\frac{\epsilon_0}{\mu_0} E(\omega) E^*(\omega) \frac{\partial \epsilon}{\partial M}$$

$$\epsilon' = \begin{pmatrix} \epsilon_{xx} & -i\alpha M & 0 \\ +i\alpha M & \epsilon_{yy} & 0 \\ 0 & 0 & \epsilon_{zz} + o(M^2) \end{pmatrix}$$

$$\vec{H}(0) = \frac{\epsilon_0}{\mu_0} \alpha [\vec{E}(\omega) \times \vec{E}^*(\omega)]$$



**Inverse Faraday effect**

Pitaevskii, *Sov. Phys. JETP* **12**, 1008 (1961).  
van der Ziel *Phys. Rev. Lett.* **15**, 190 (1965).

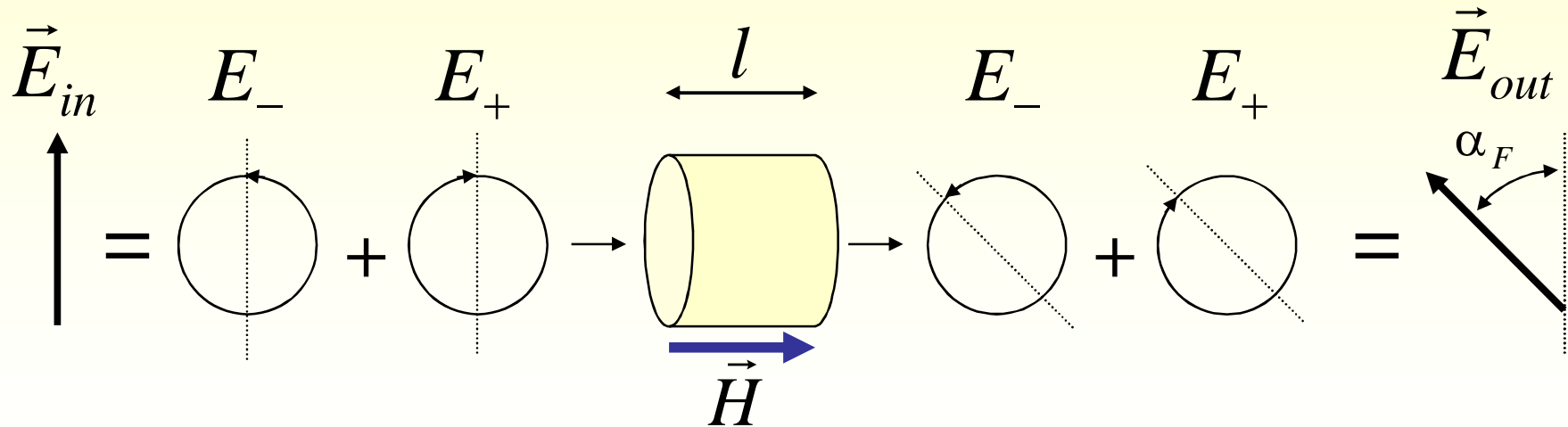
# Faraday effect

$$\text{Faraday rotation: } \theta_F = \frac{2\pi l}{\lambda} \frac{\alpha M}{\epsilon_0}$$

$$\vec{H}(0) = \frac{\epsilon_0}{\mu_0} \alpha [\vec{E}(\omega) \times \vec{E}^*(\omega)]$$

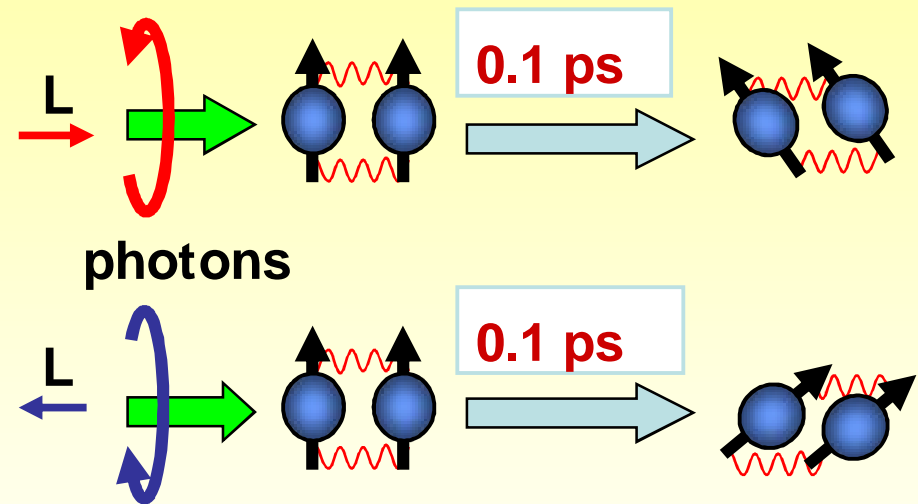
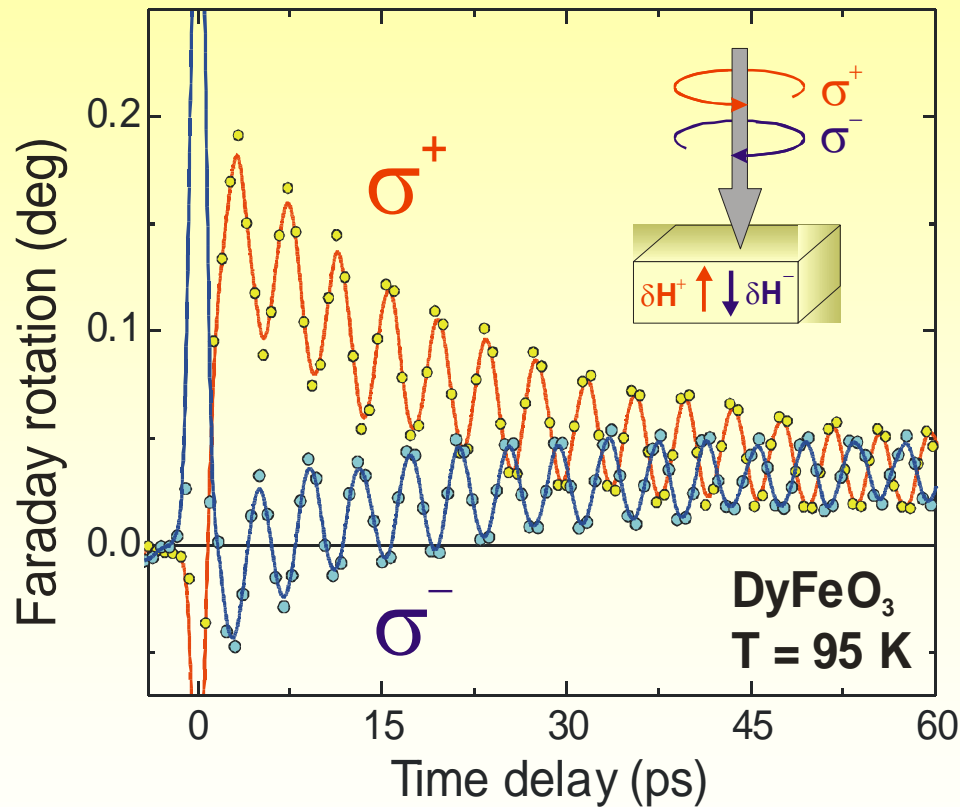
no absorption required!

no angular momentum transfer!



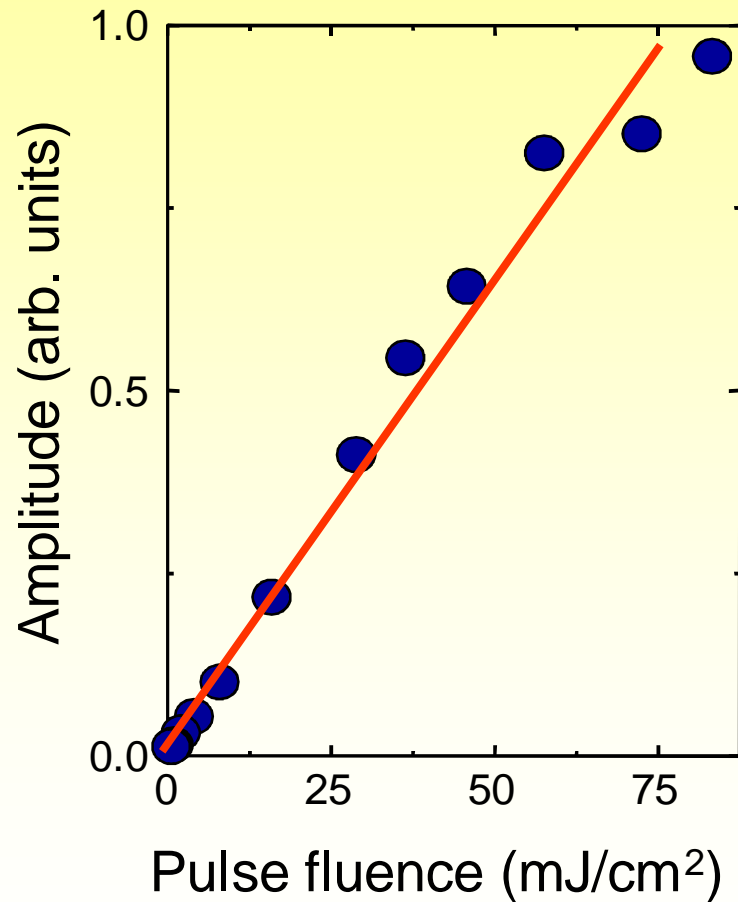


# Excitation of spins in DyFeO<sub>3</sub>



as strong as 1 T!

# of the laser-induced spin-waves

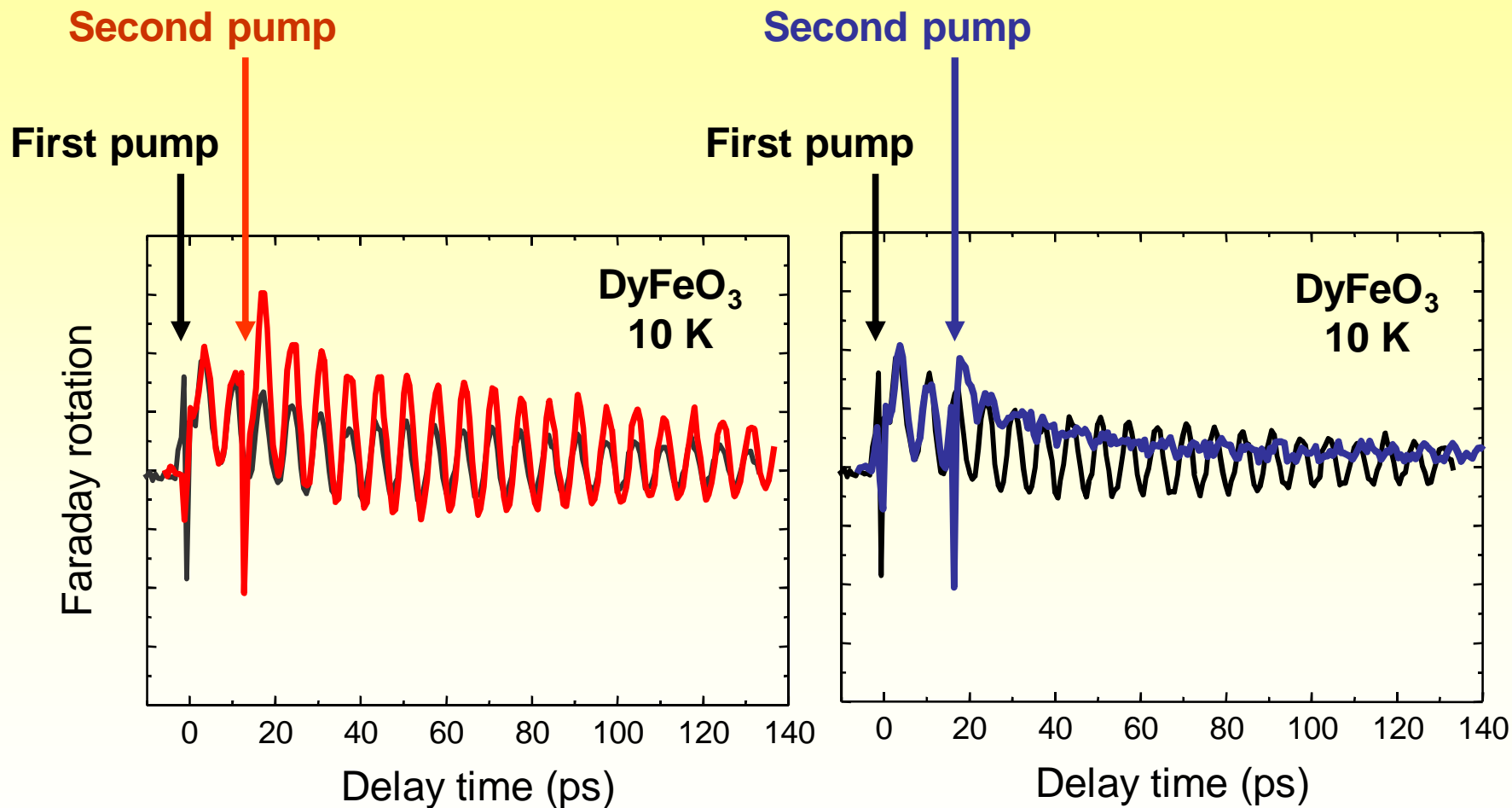


## Inverse Faraday effect

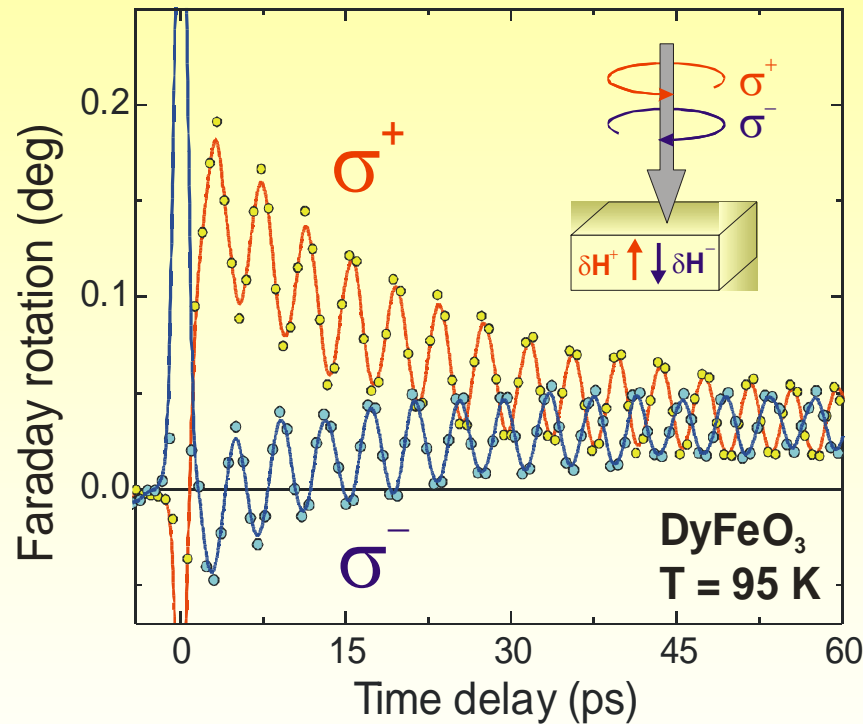
$$H_{eff}(0) = \alpha \frac{\epsilon_0}{\mu_0} E(\omega) E^*(\omega)$$

**Fields up to 5 T!**

# pump coherent control



# Excitation of spins in DyFeO<sub>3</sub> via the Inverse Faraday effect



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

## Referee A:

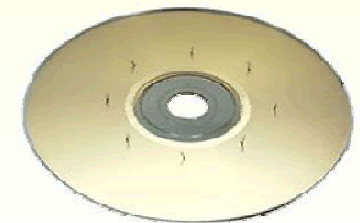
*But, unfortunately, the observed signal is so small that it seems impractical to utilize the inverse Faraday effect for the purpose of ultrafast control of magnetization in metallic materials.*

# Magneto-optical imaging + pulsed laser

**Circularly polarized 40fs laser pulses**



**20 nm GdFeCo film**



**Magneto-Optical microscope**

# Manipulation of magnetization

40 fs pulses, 1 kHz

GdFeCo

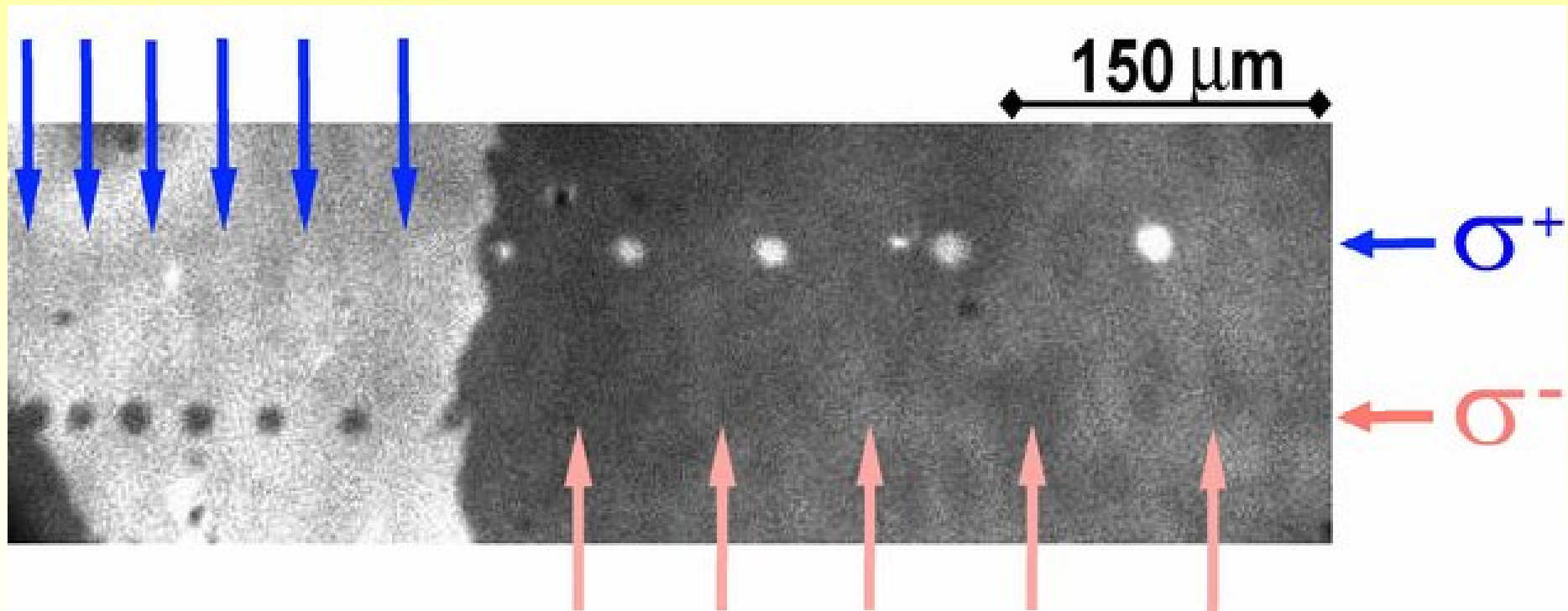
$H_{\text{ext}} = 0$



*Reversal by 40fs laser pulses!*

# Magnification reversal in FeGdCo induced by a single 40 fs laser pulse

Sweeping the pulsed laser beam at high speed across the sample



***“ Each domain written with single 40 fs laser pulse***

C.D. Stanciu et al., PRL 99,047601 (2007)

# Ultrafast all optical recording



C.D. Stanciu et al., *patent* #P77323PC00, PRL **99**, 047601 (2007)



*So,*

*É Light can reverse magnetization!*

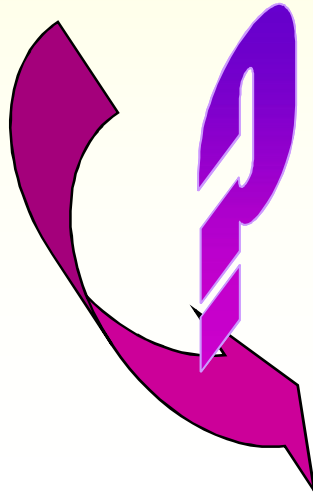
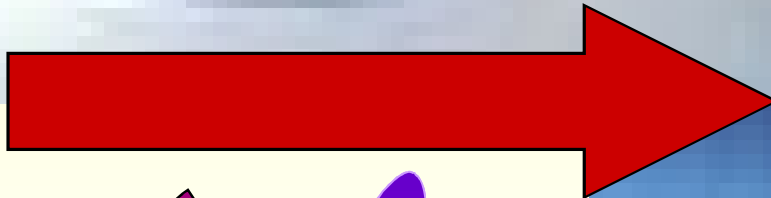
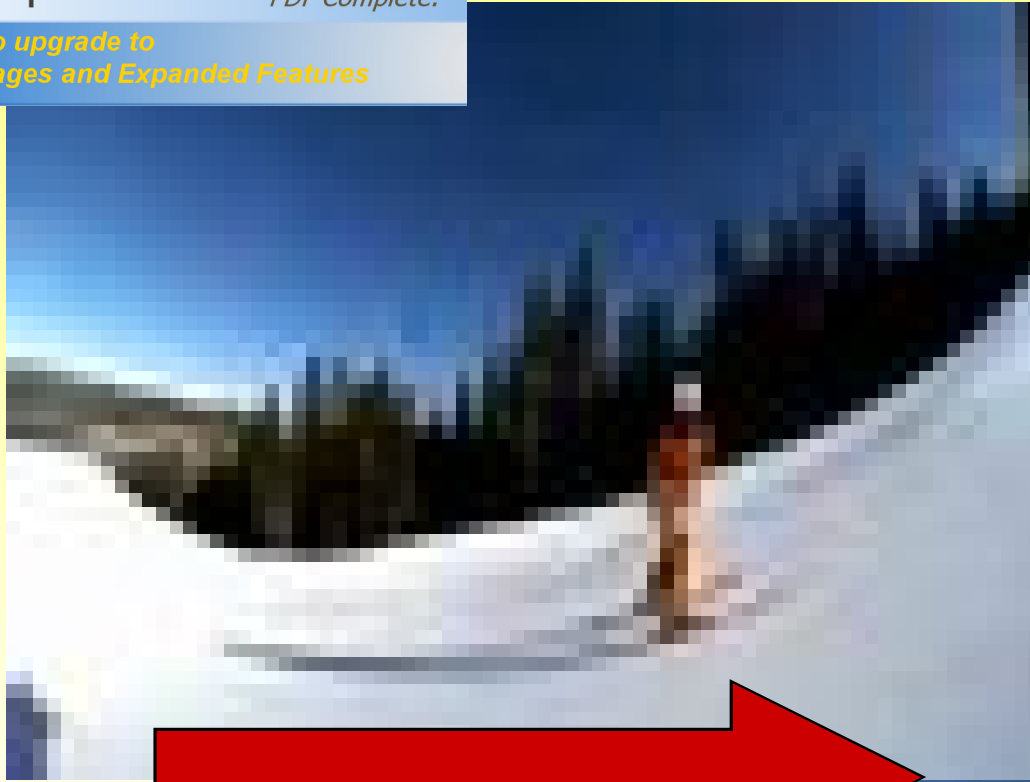
*É..and even a single 40fs pulse !*

*É But what is the mechanism?*

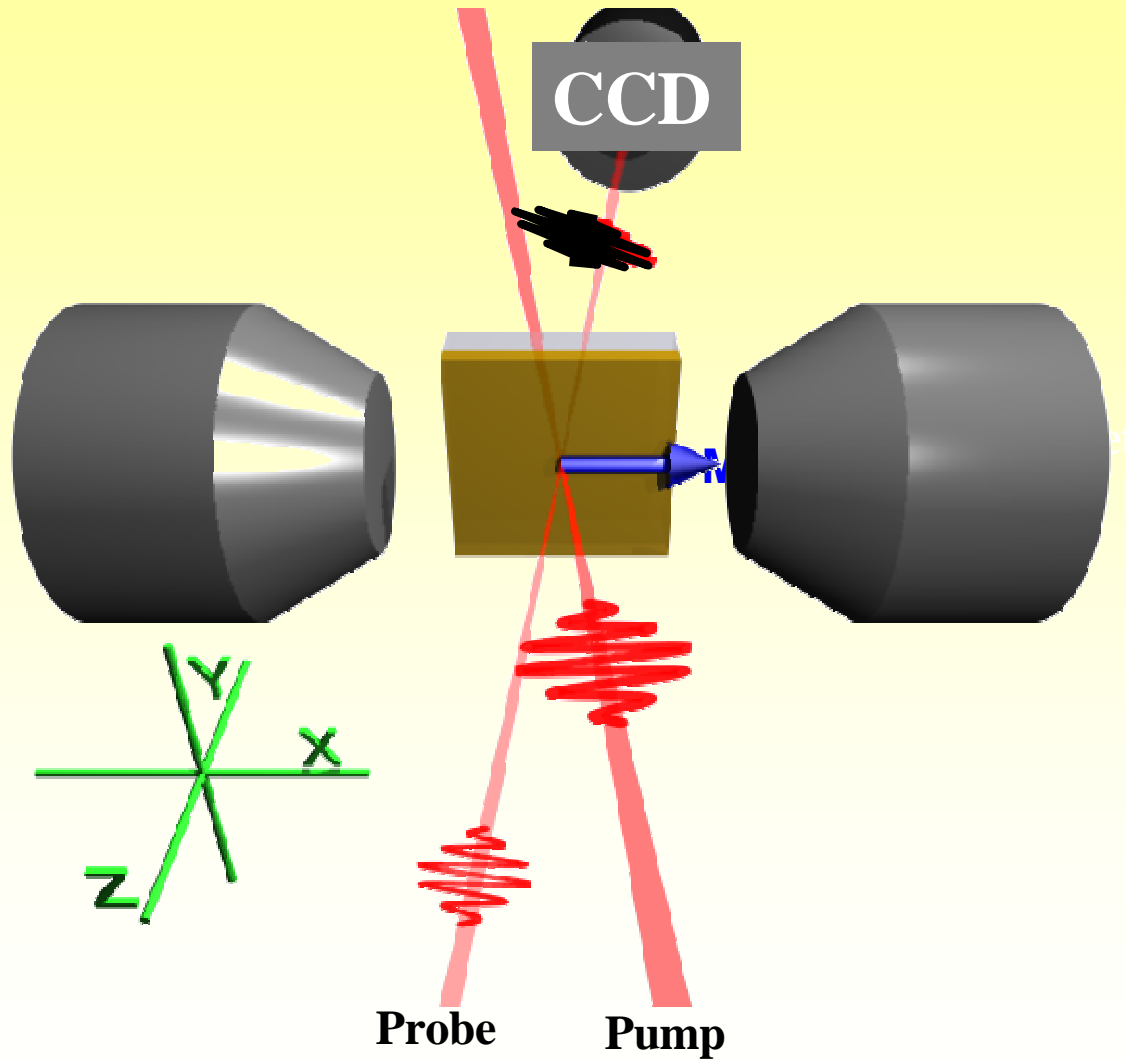


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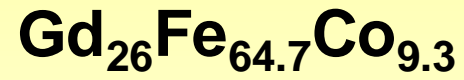
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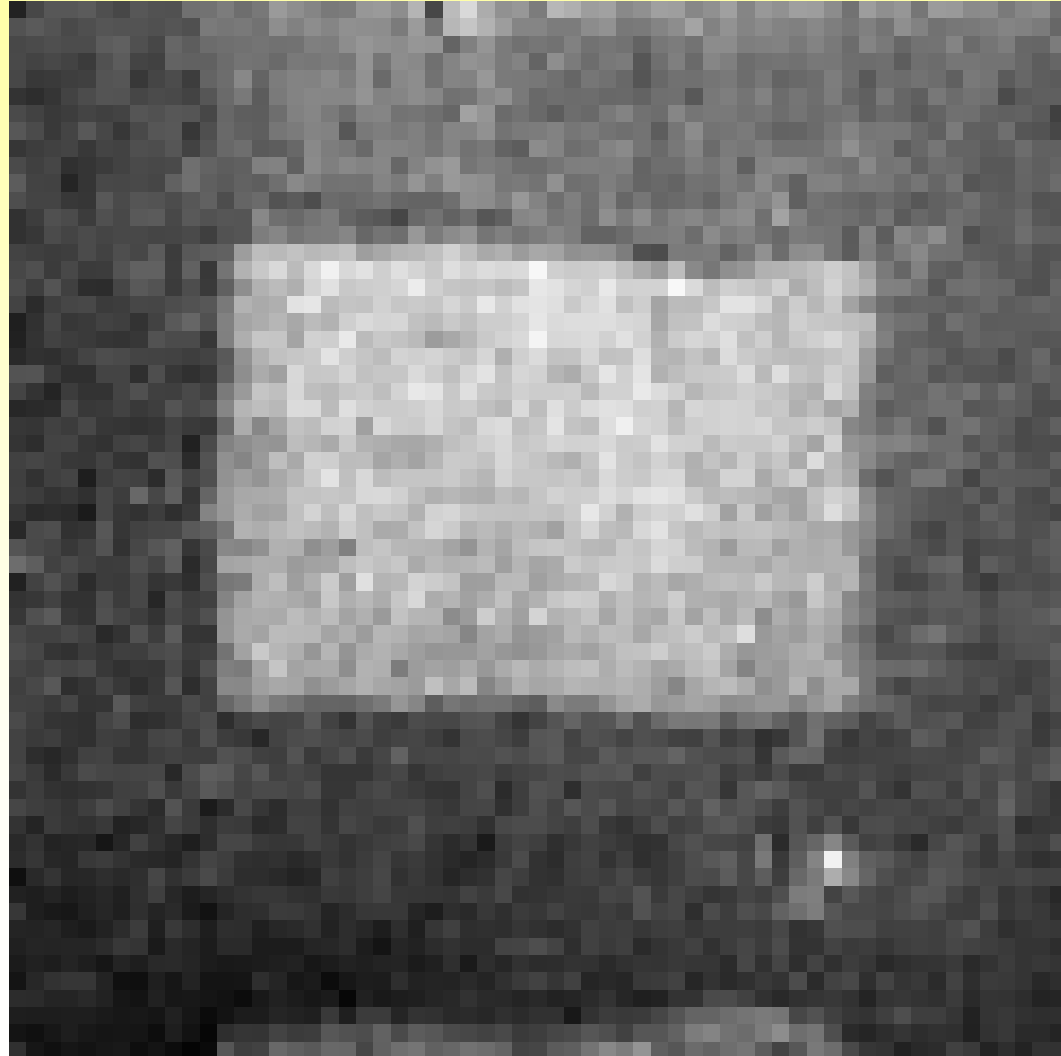
# Optical magneto-optical imaging



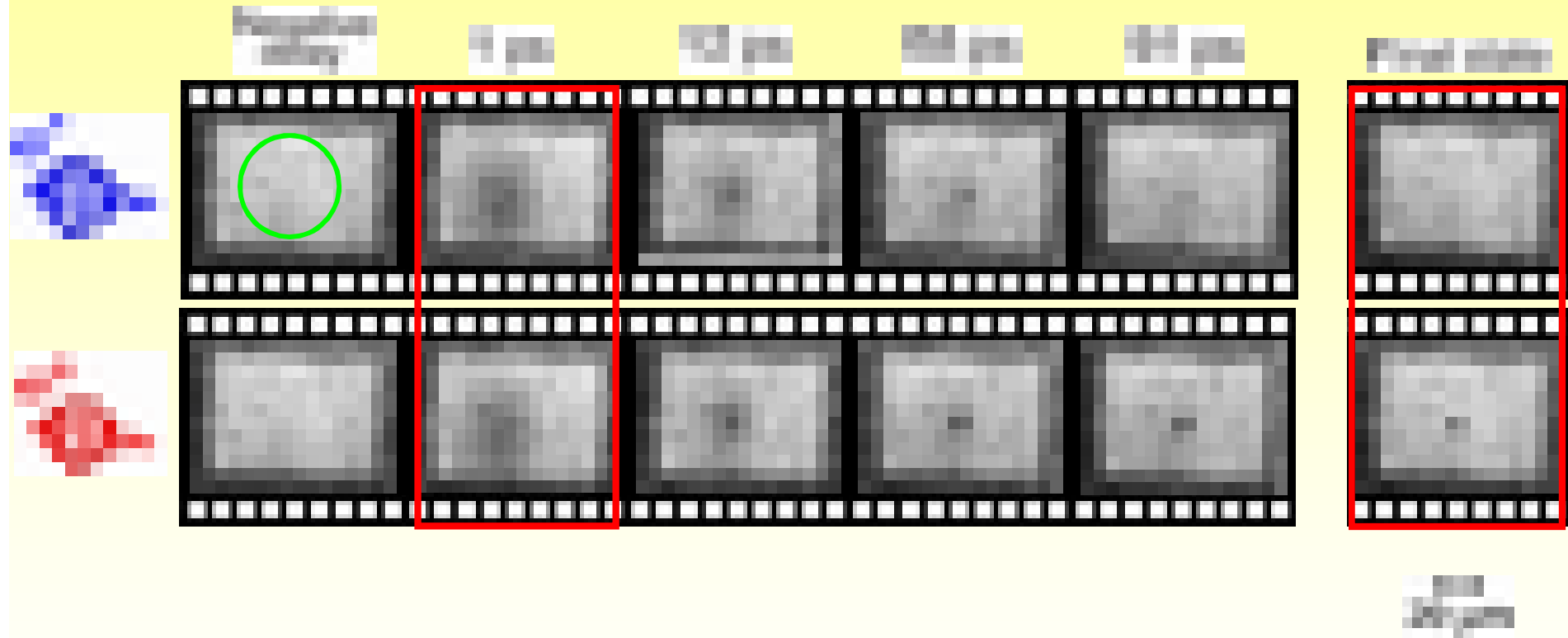
# Physics of all-optical reversal



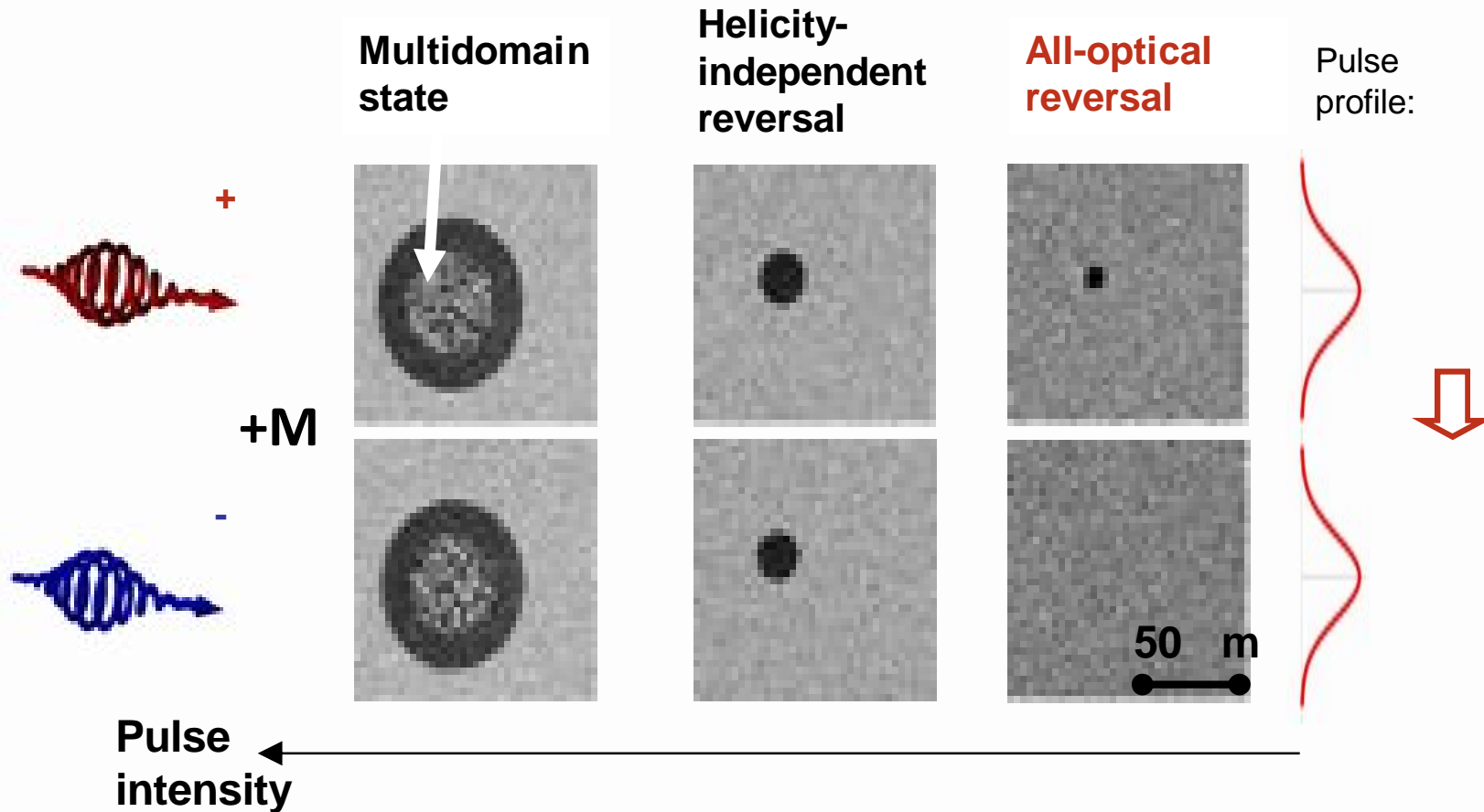
-



# olution of the domains



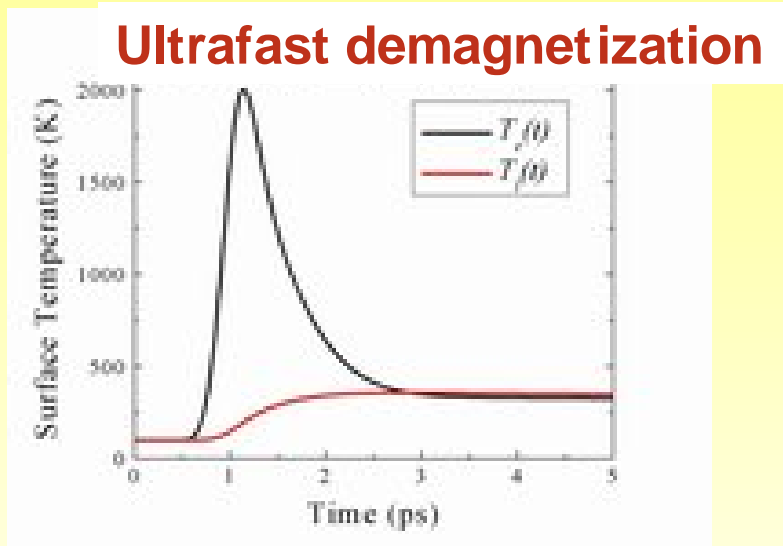
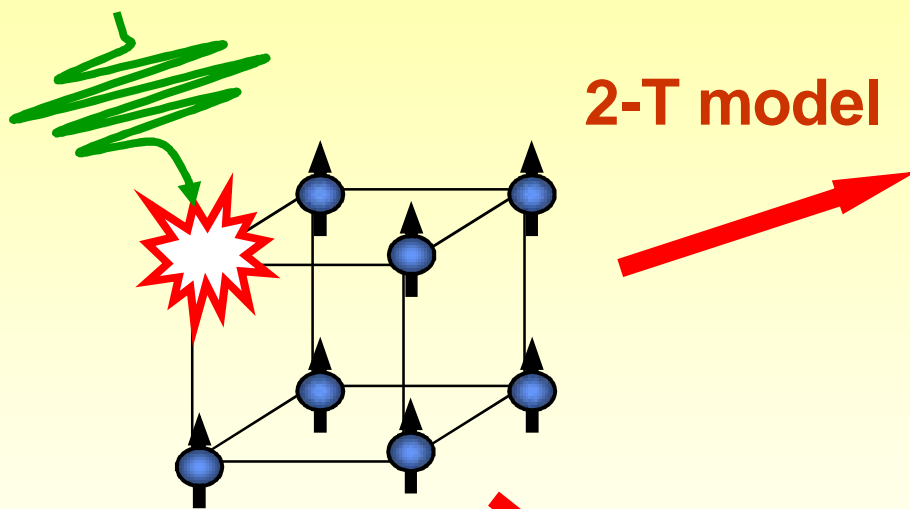
# Dependence of the light intensity on all-optical magnetization reversal



**All-optical reversal in narrow (~5%) intensity range**

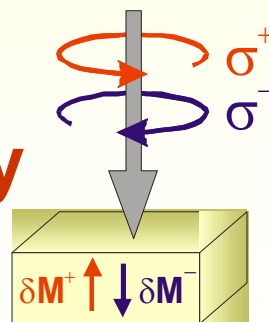
# Ultrafast dynamics: theory/simulations

## Ultrashort laser pulse



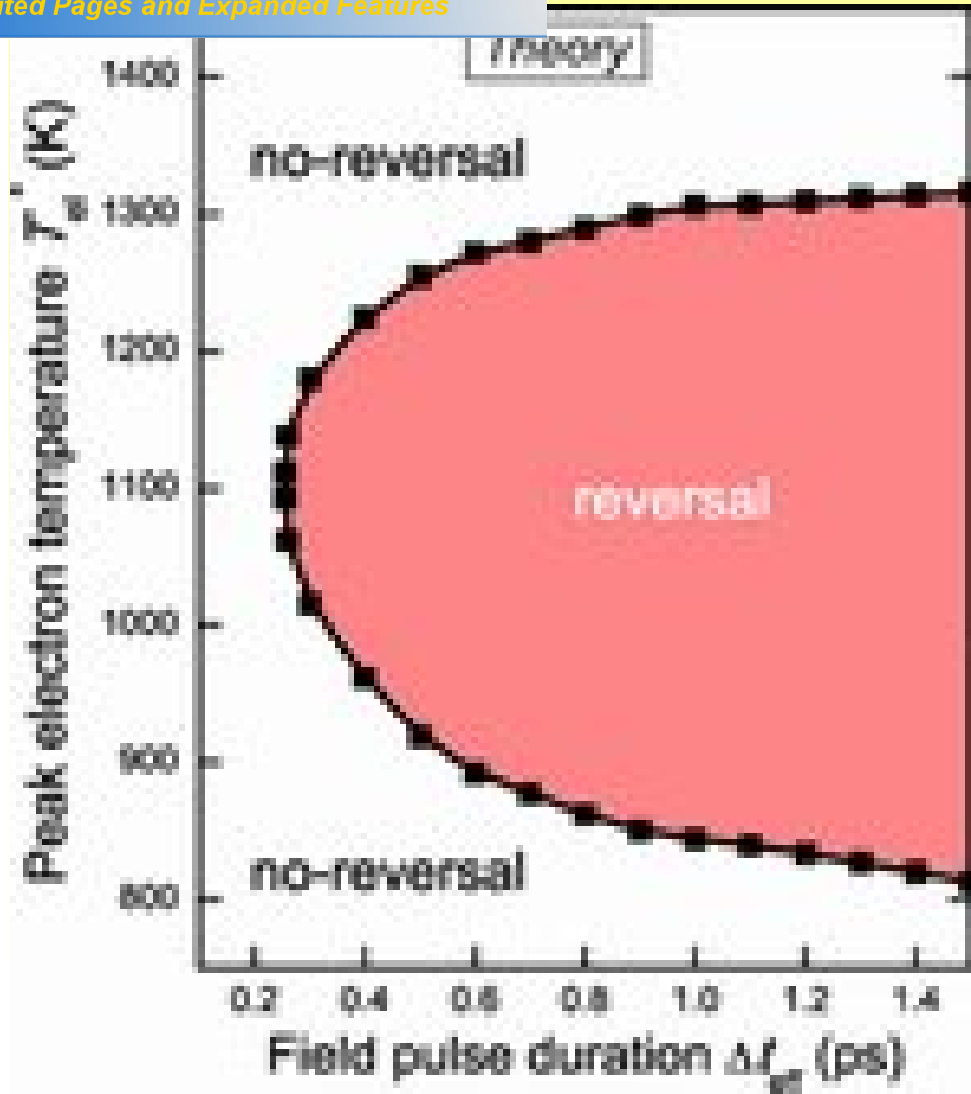
## Inverse Faraday

[A. V. Kimel et al., Nature 435, 655 (2005)]



**GdFeCo: 20T!**

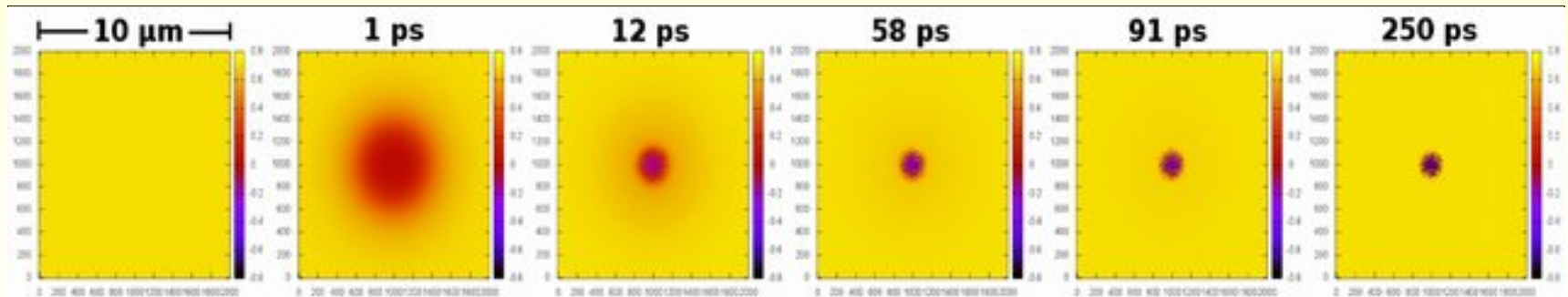
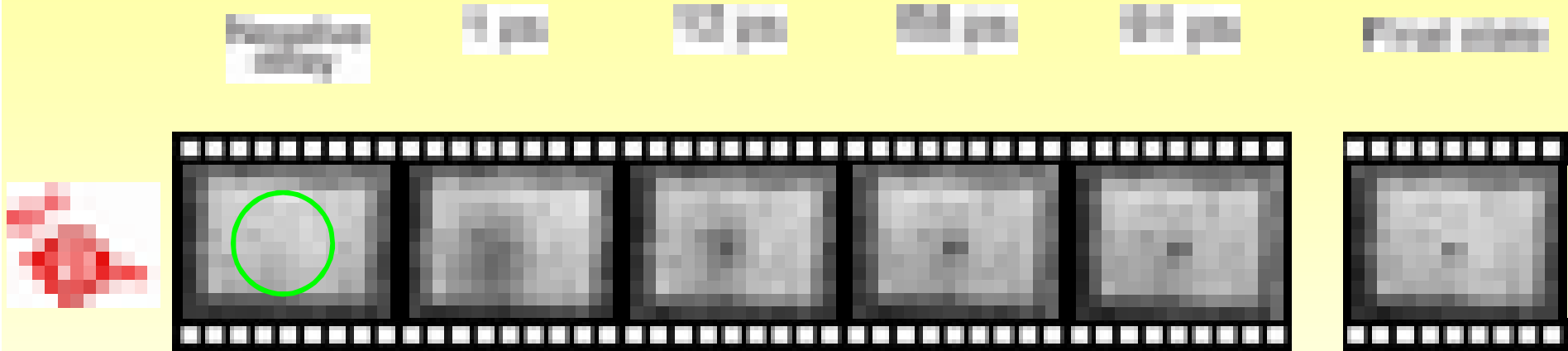
# Theory - Experiment



K. Vahaplar et al., PRL 103, 117201 (2009)



# ion of the domains

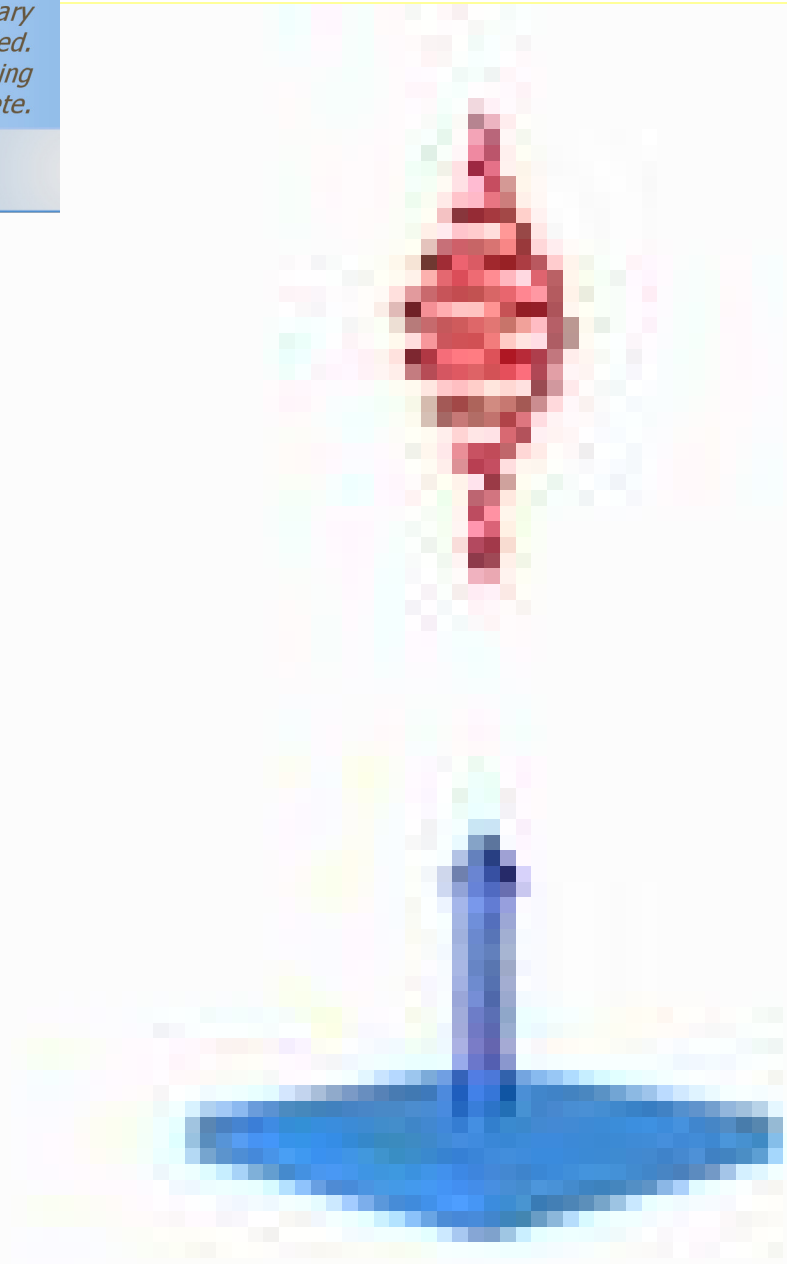


K. Vahaplar et al., Phys. Rev.B , 2010

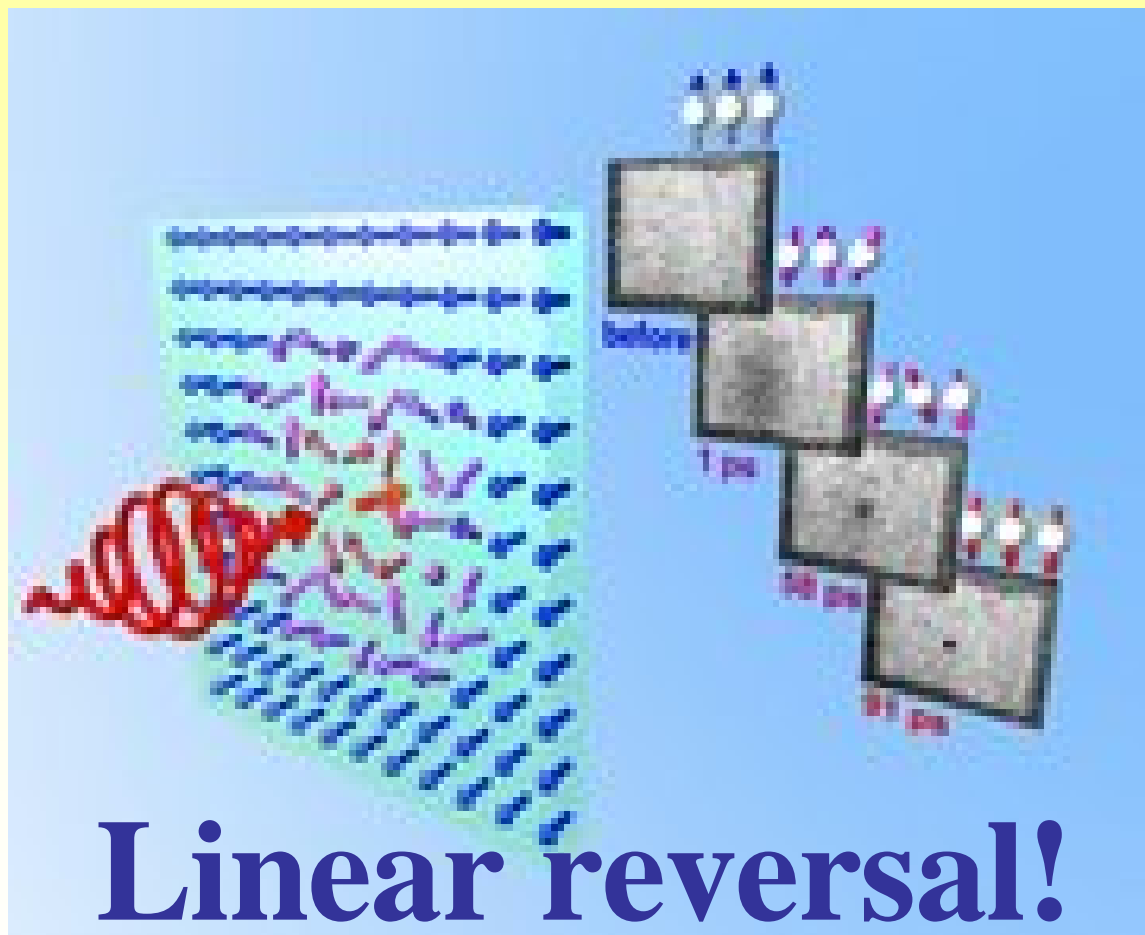


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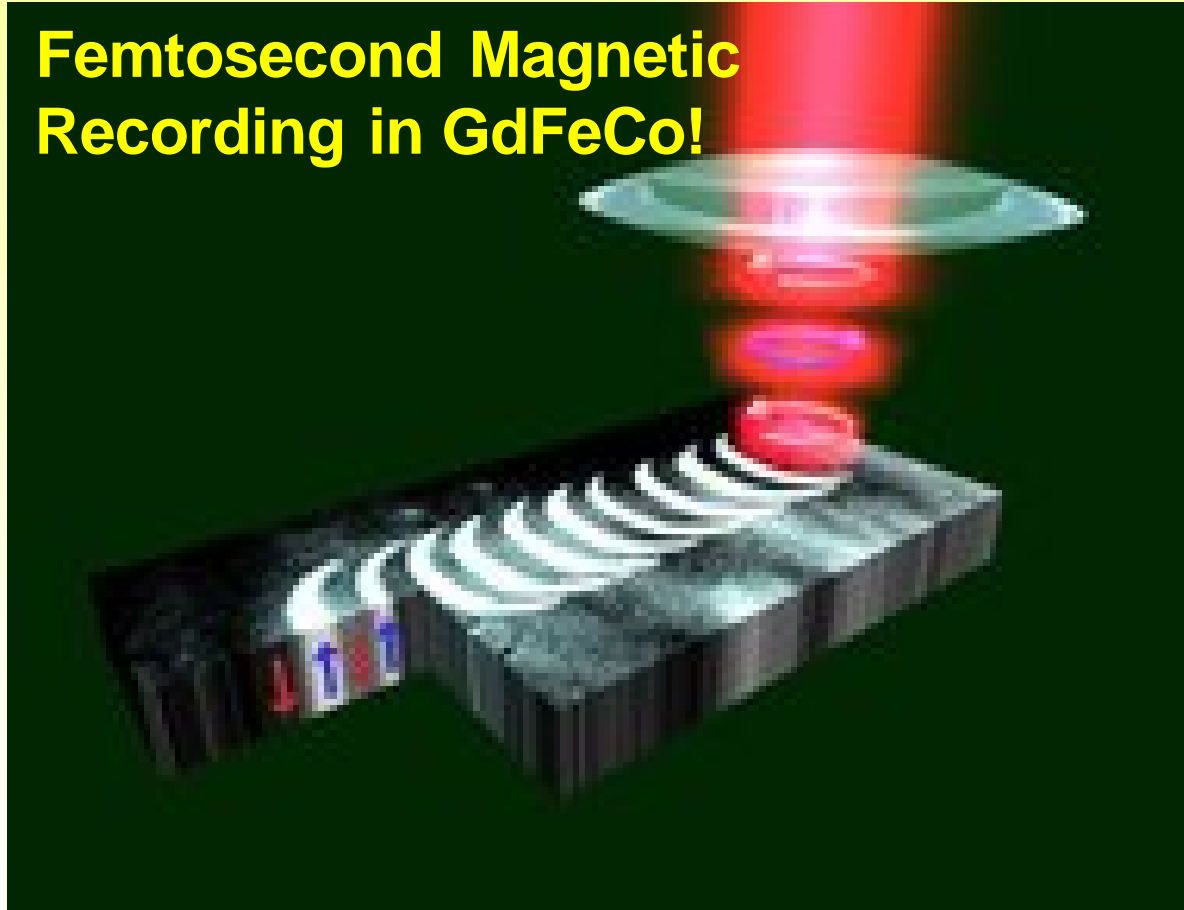


# ew Mechanism!

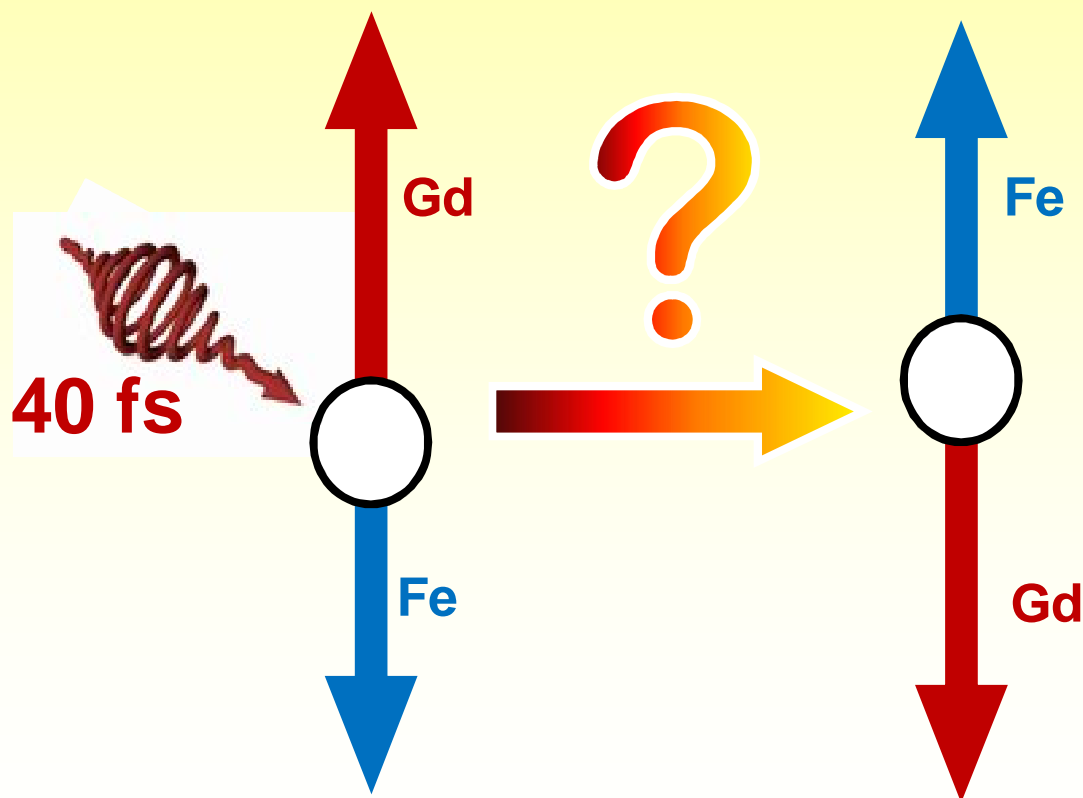


# near-reversible: how? multi-sublattice ferrimagnet!

## Femtosecond Magnetic Recording in GdFeCo!

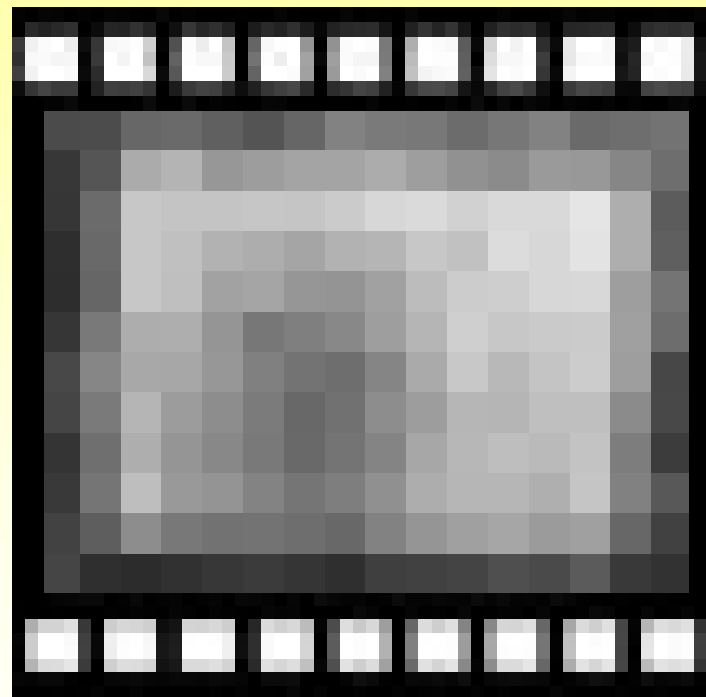
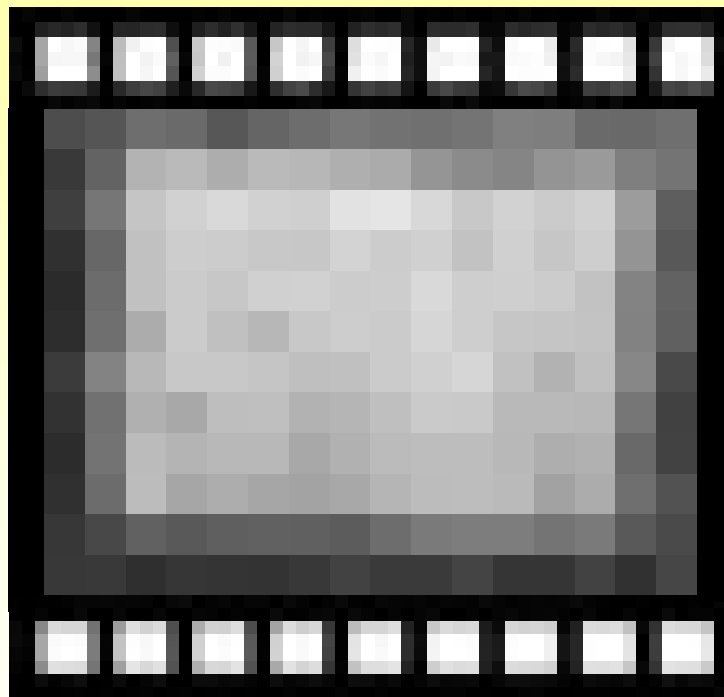


# Ultrafast **laser** induced reversal in *multi*-sublattice **magnets**



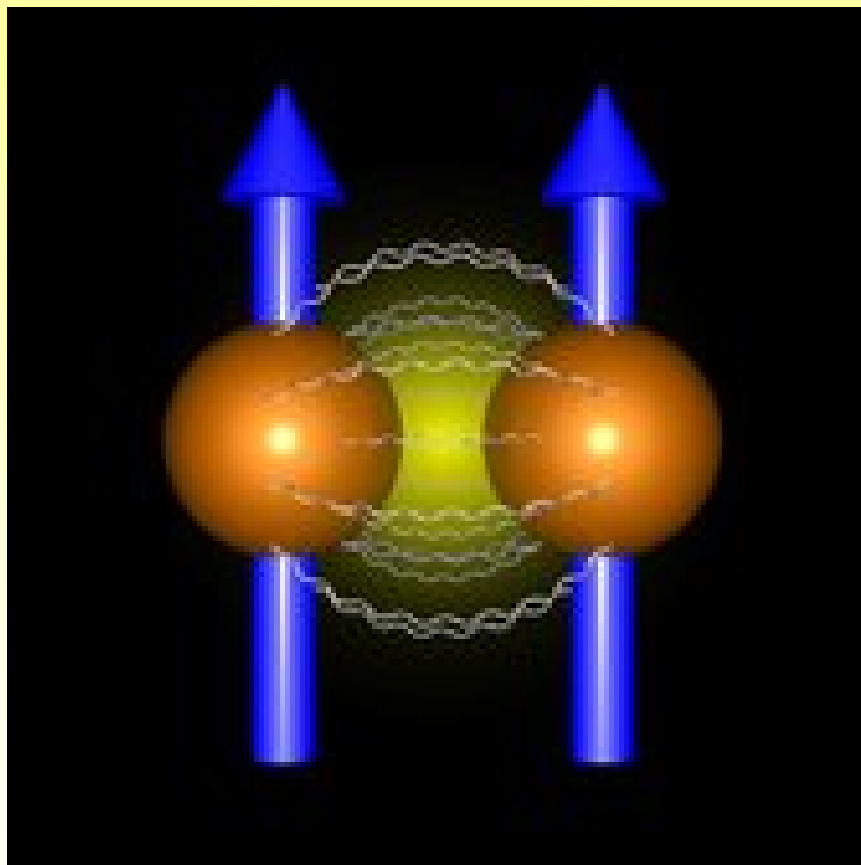


$0 \text{ í } \text{ í } \text{ í } \text{ í } 1 \text{ ps?}$

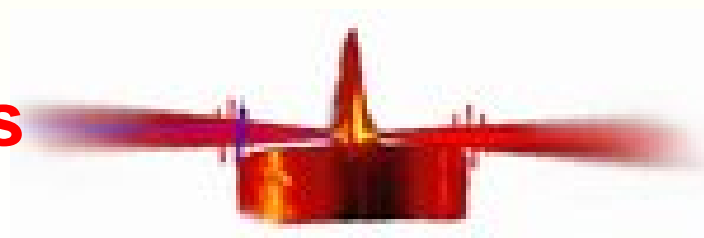


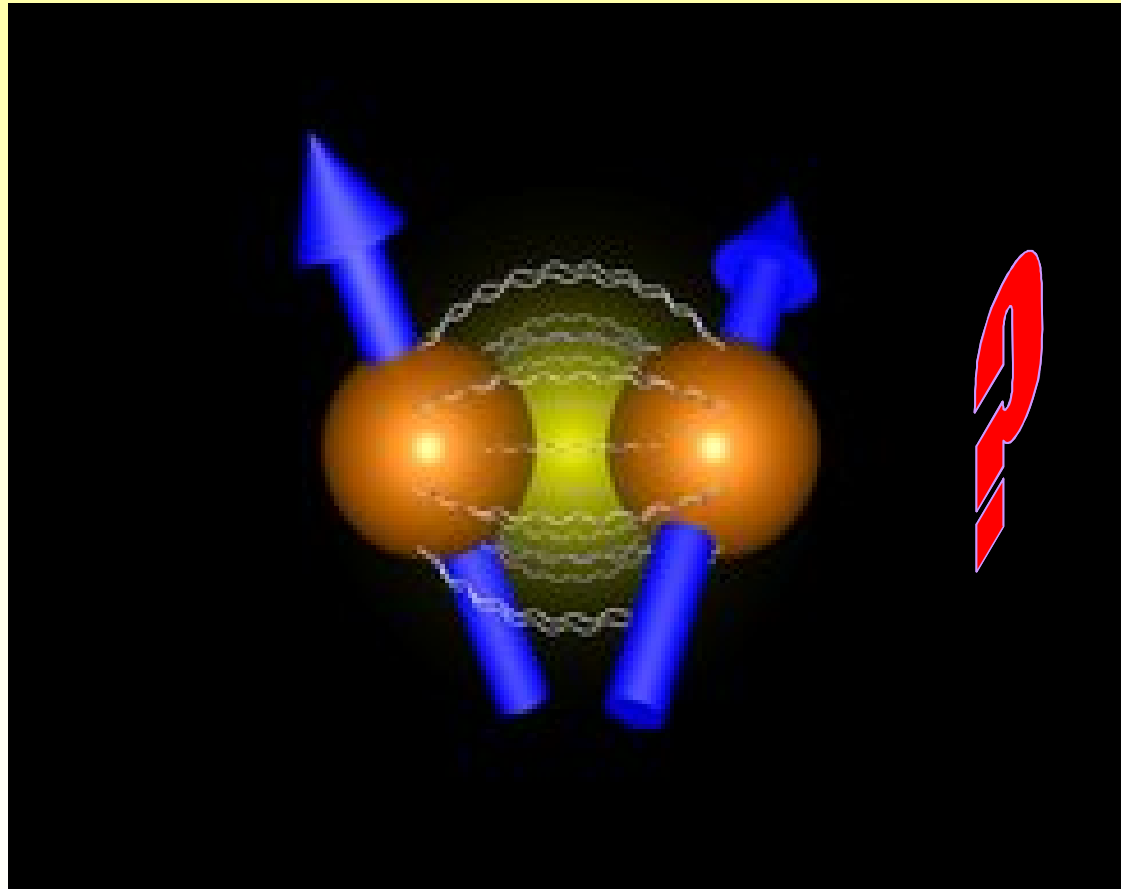
**Magnetism at the timescale  
of the exchange interaction?**

$\text{Fe-Fe} = 1.4 \times 10^{-19} \text{ J} \sim 40 \text{ fs}$



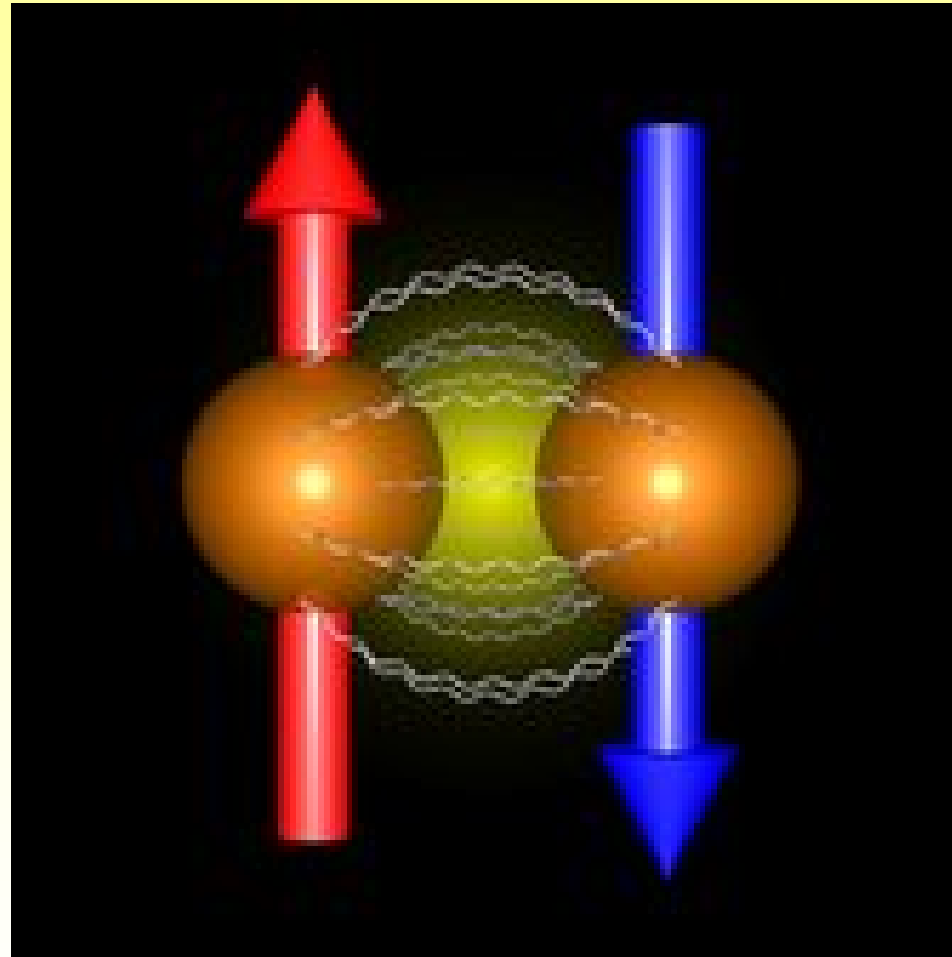
**1-2eV, 10-100fs**





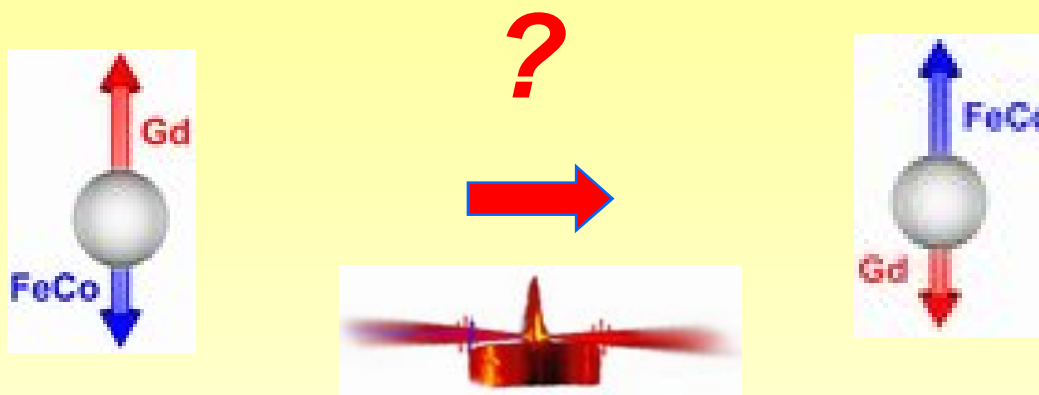


# ferrimagnet



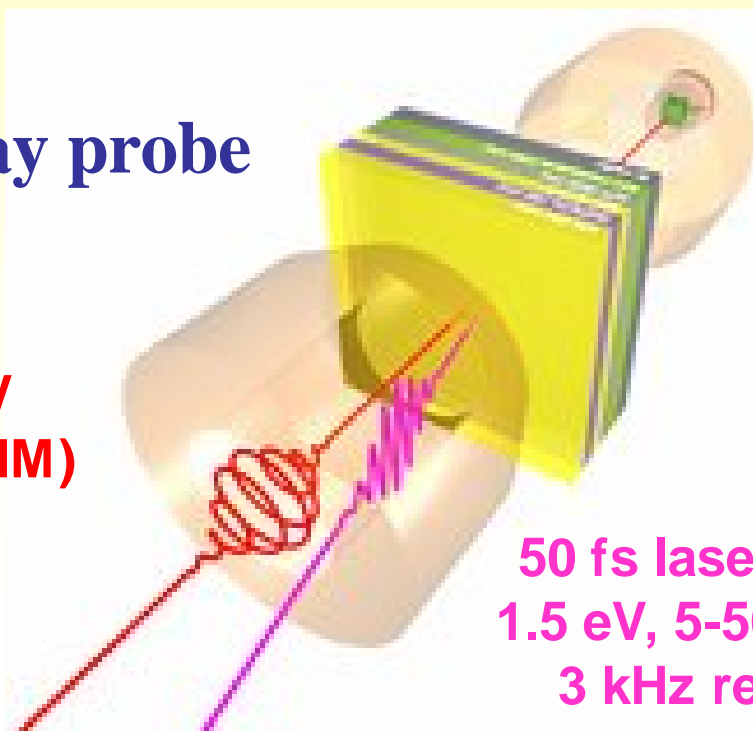
ōspins colouredö!

# Element specific view



Laser pump ó X-ray probe

X-rays  
400-1400 eV  
10-50 ps (FWHM)



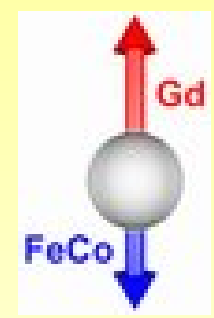
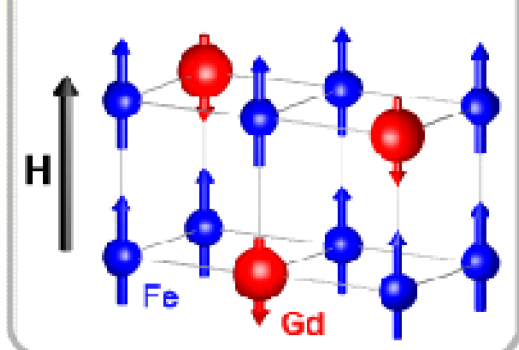
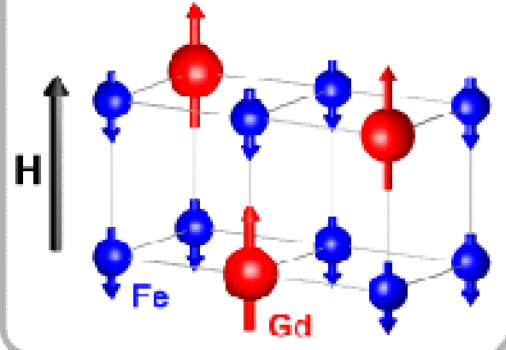
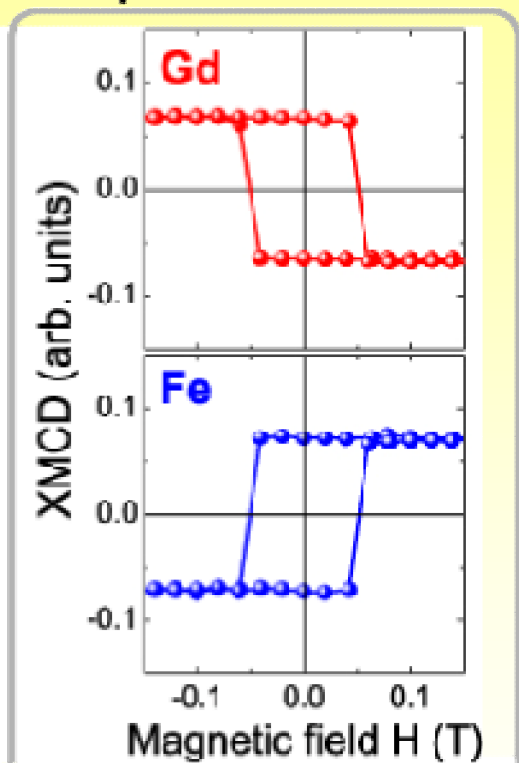
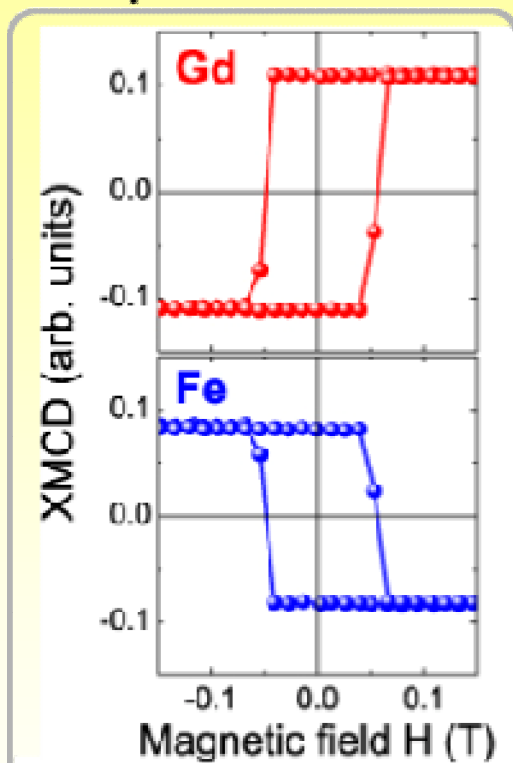
BESSY

50 fs laser pulses  
1.5 eV, 5-50 mJ/cm<sup>2</sup>  
3 kHz rep. rate

# Antiferromagnetic Rare Earth-Transition Metal

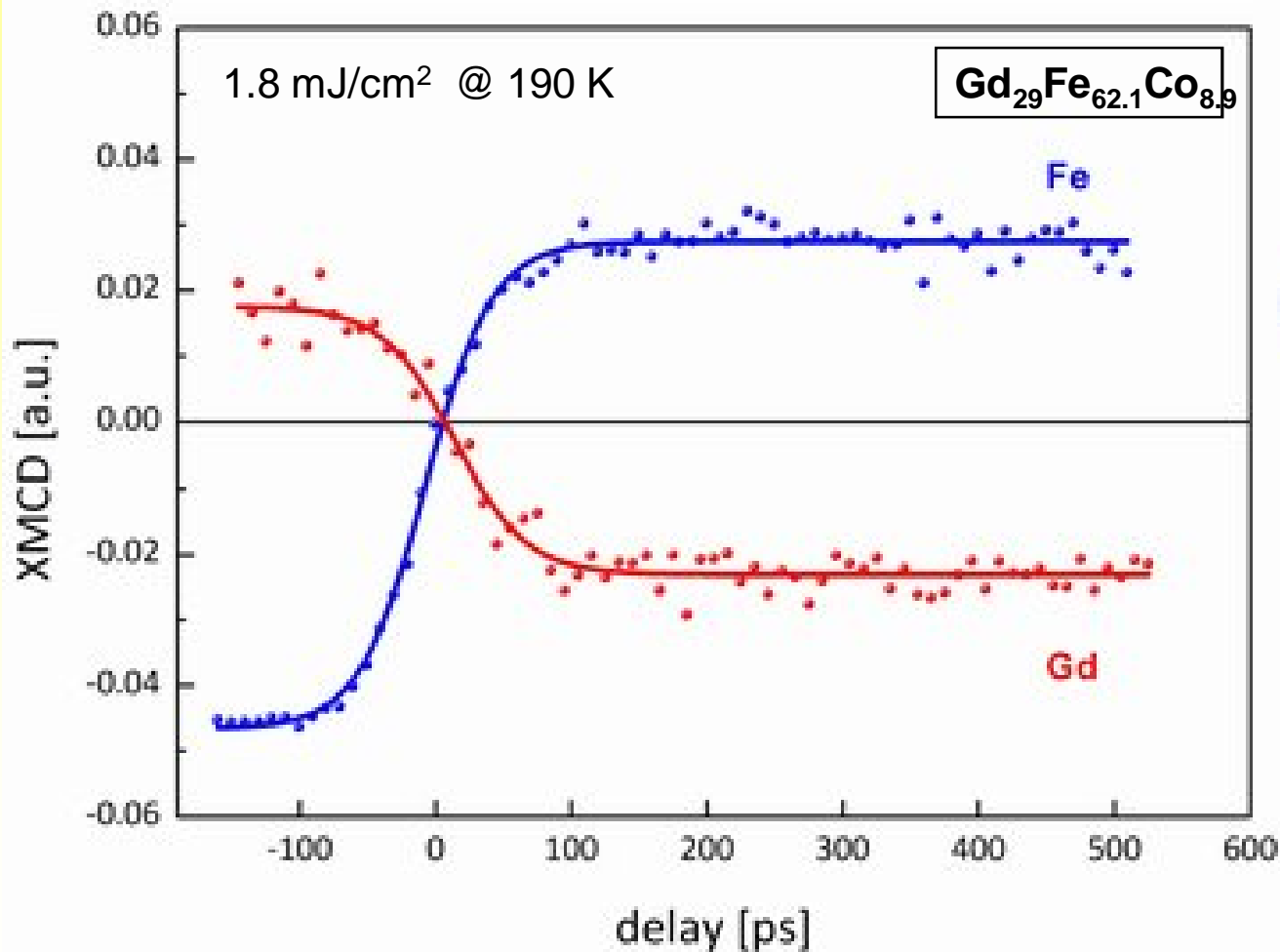
Below compensation point  $T = 82$  K

Above compensation point  $T = 350$  K



# TR-XMCD @ BESSY

» Fe and Gd sub-lattices switch simultaneously within 50 ps.





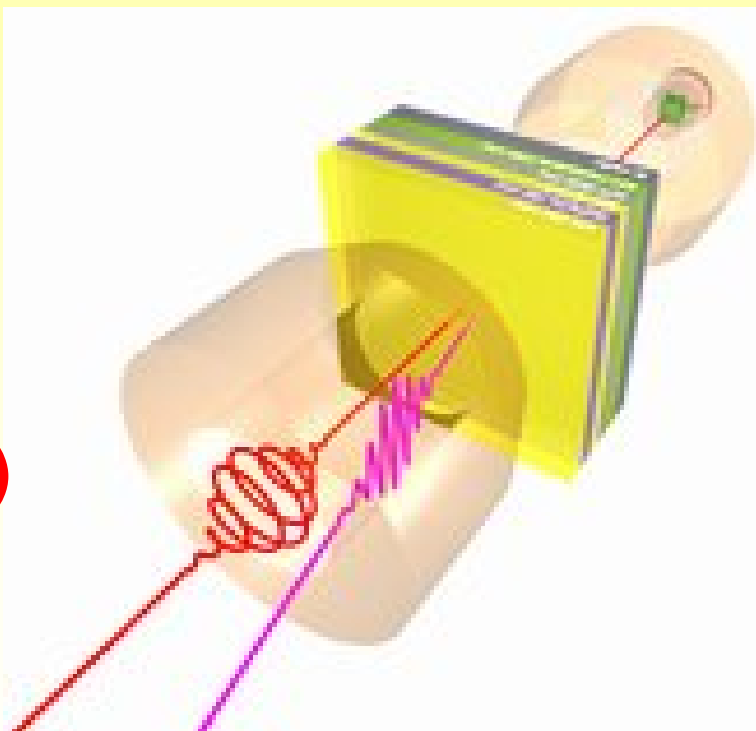
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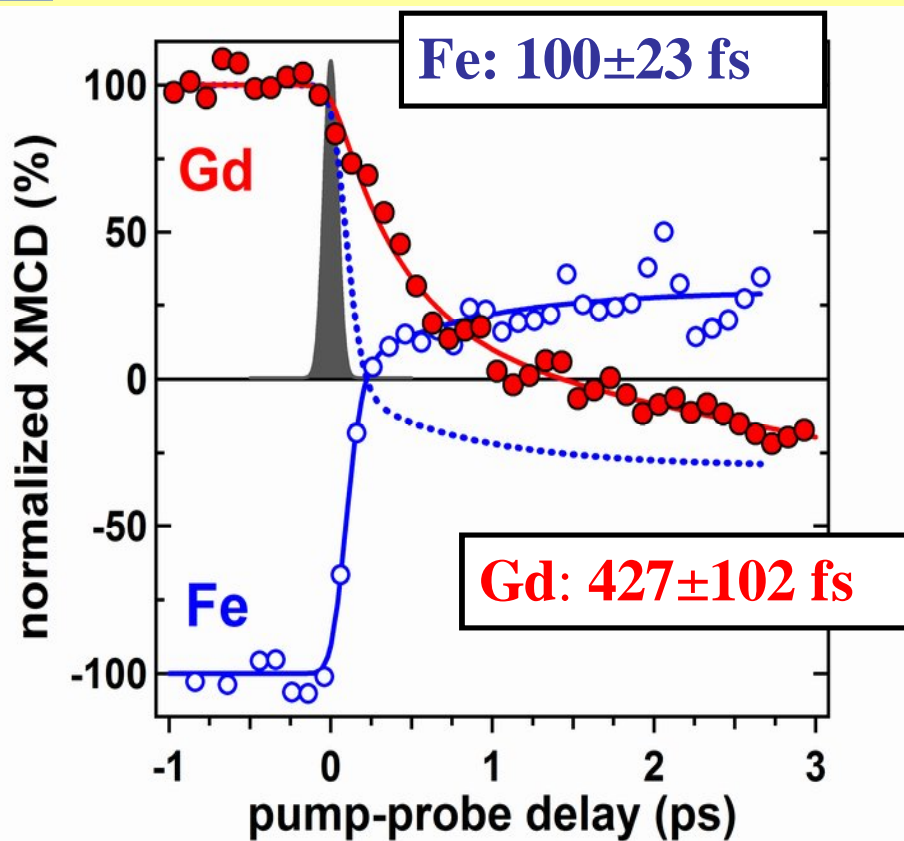
# Canøt we look faster?

# FEMTO-SLICING!

**X-rays**  
**400-1400 eV**  
**100 fs (FWHM)**

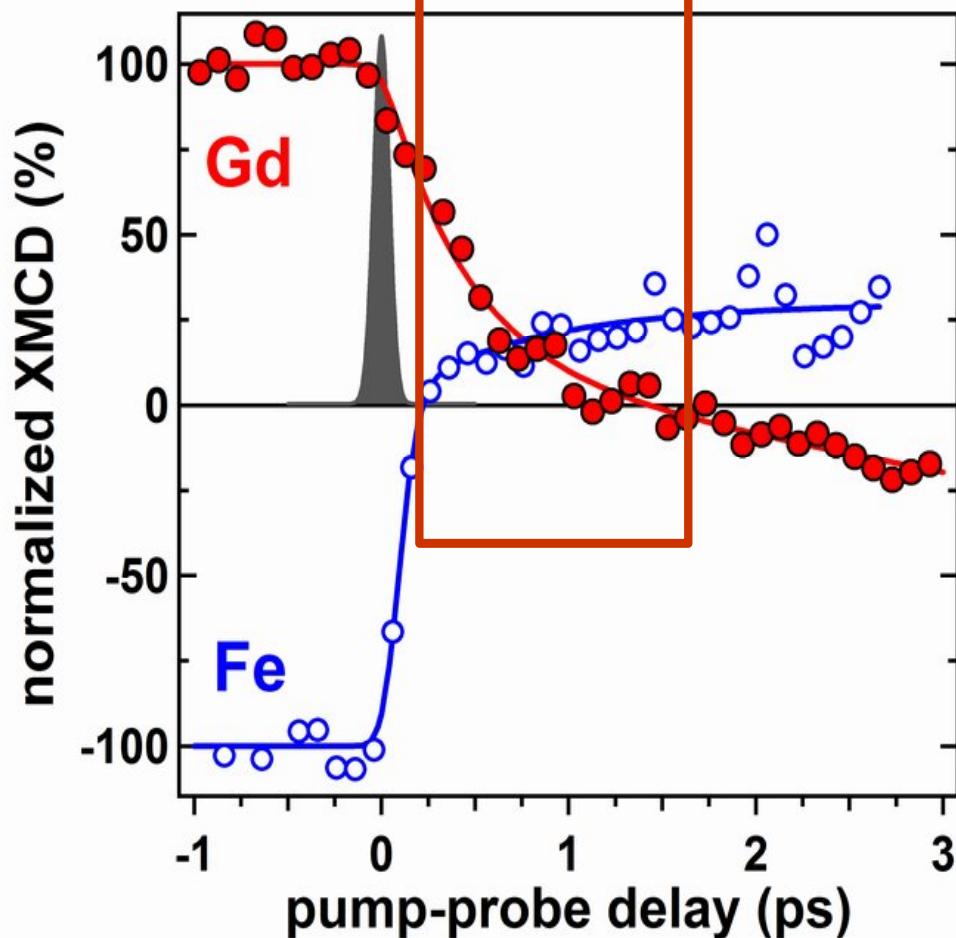
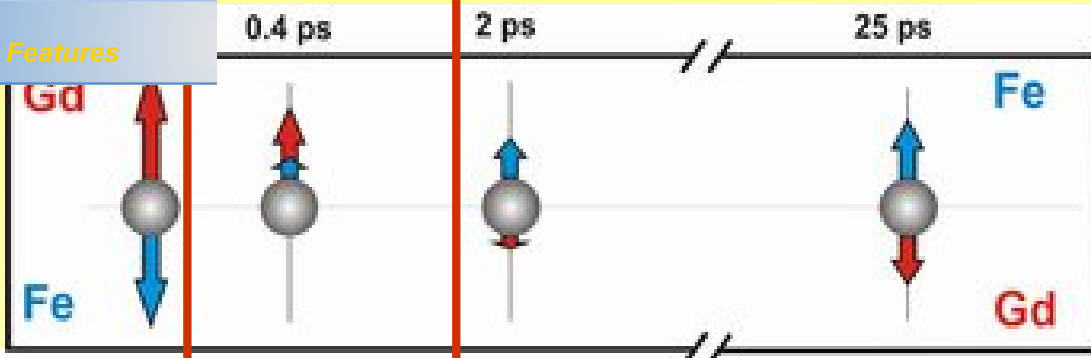


# specific magnetization reversal



**Different magnetization switching dynamics Fe and Gd !!!**

# via a novel non-equilibrium state



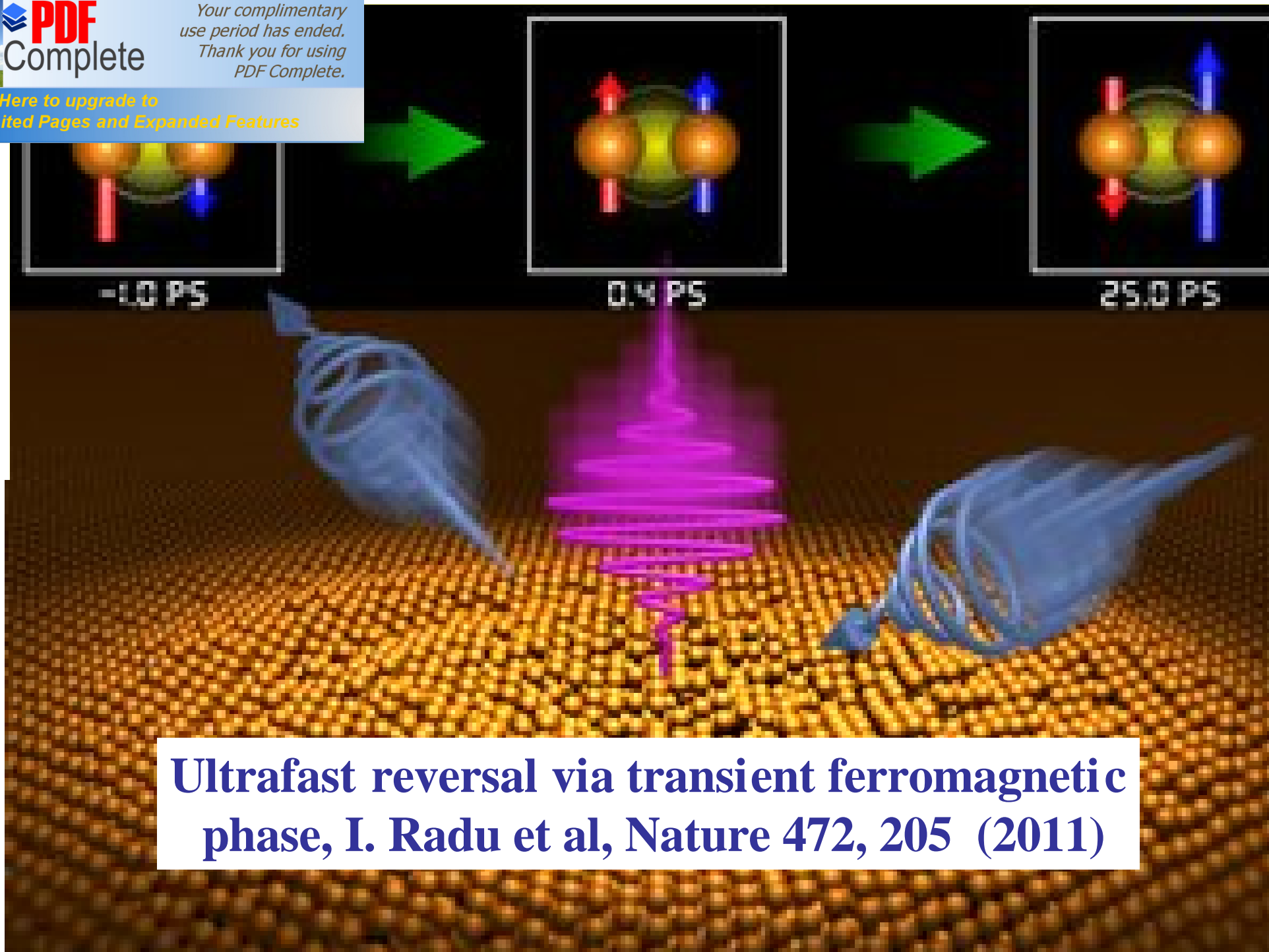
“Transient **FERROMAGNETIC ALIGNMENT** for ~1.2 ps



“The system evolves against the exchange interaction !!!

$$J_{\text{Fe-Gd}} = 4.77 \times 10^{-20} \text{ J} \sim 140 \text{ fs}$$







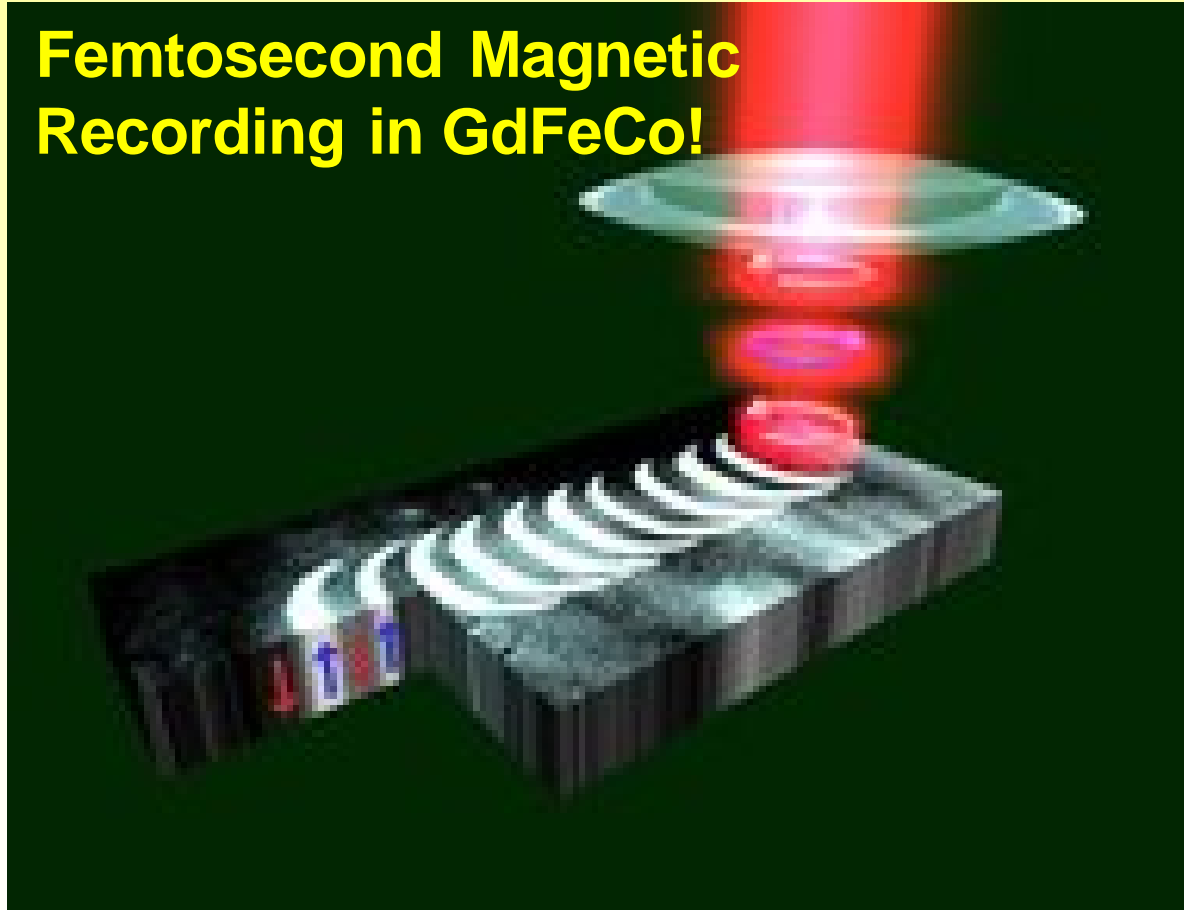
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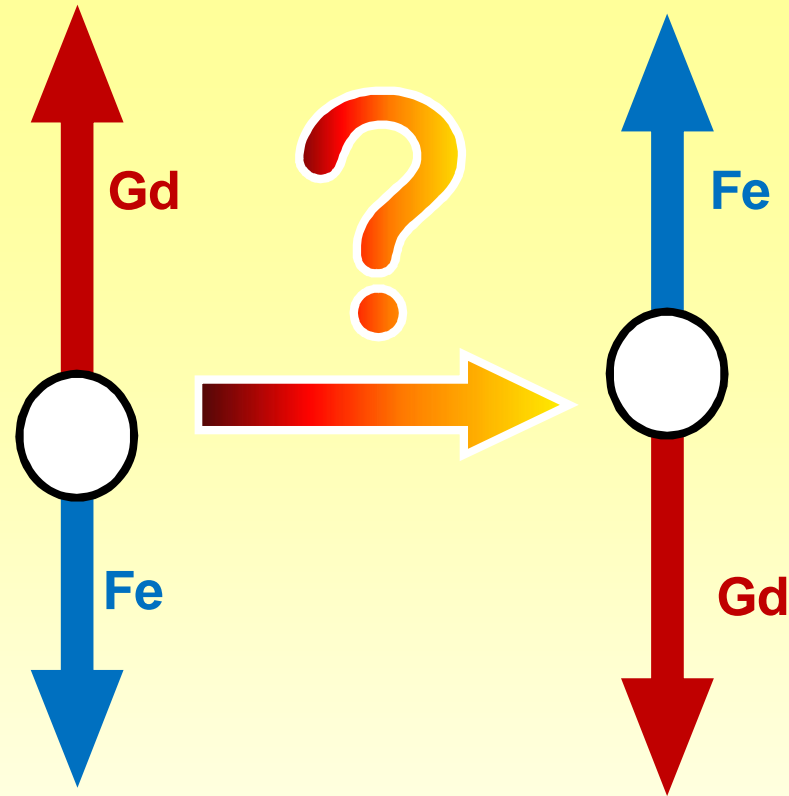
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# How to understand?

# can it be a ferrimagnetic material?

## Femtosecond Magnetic Recording in GdFeCo!





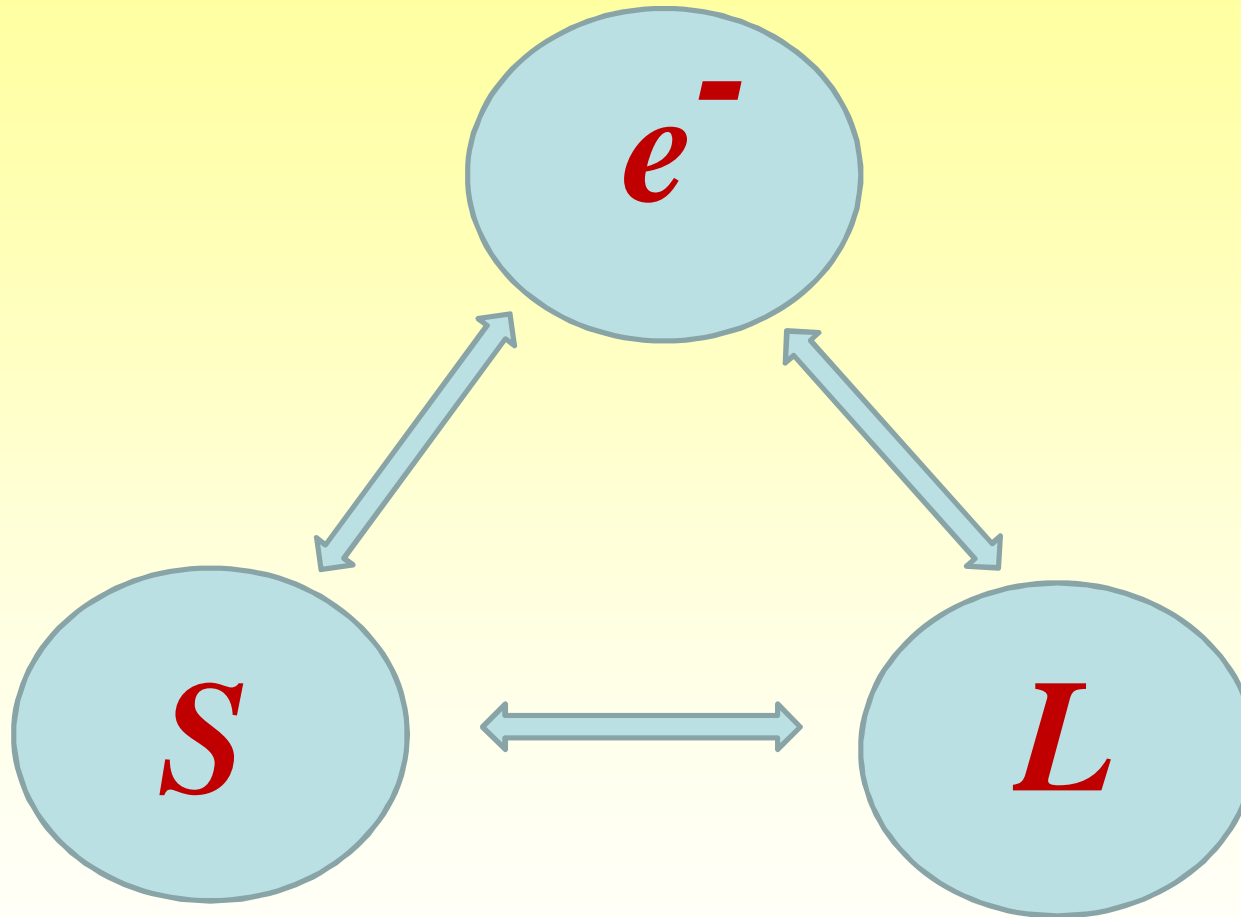
**Angular momentum transfer?**

**Role of temperature?**

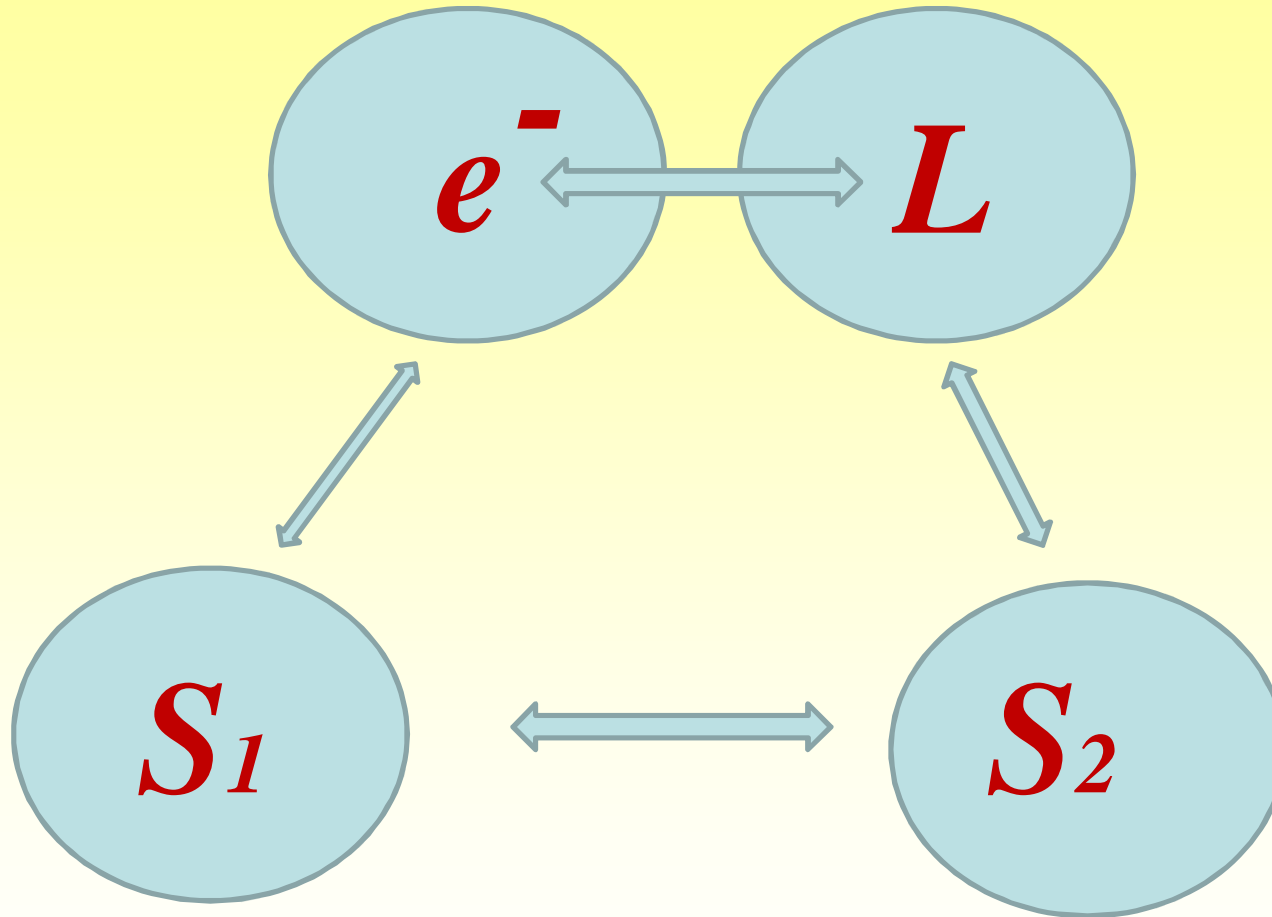
**Role of sublattices?**

**í ..**

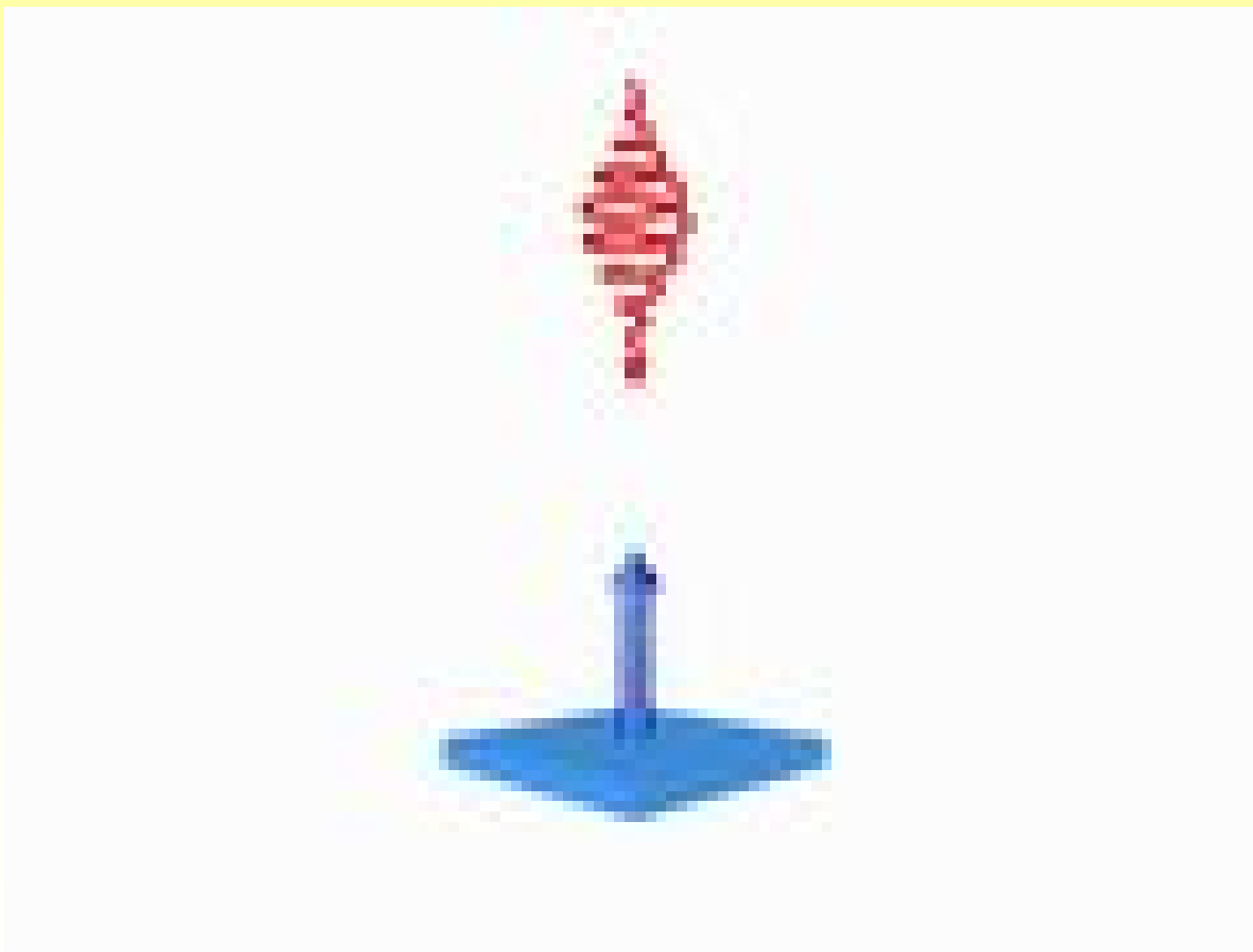
# 3-T model



# Multi-sublattice



# *Lunged at line v dysablics !!*



# Landau-Lifshitz dynamics

ON THE THEORY OF THE PRESSION OF MAGNETIC FLUIDS IN FERROMAGNETIC BODIES.  
 by L. LANDAU and E. LIFSHITZ,  
 Second Edition, 1959

magnetic moment. Therefore in the presence of the field the magnetic moment would act as a free moment, i.e. would precess around  $\mathbf{H}$  and we should have for a (Larmor) differential equation by time) the equation

$$\dot{\mathbf{m}} = \gamma \mathbf{H} \times \mathbf{m} + \lambda \left( \mathbf{m} - \frac{\mathbf{m} \cdot \mathbf{H}}{H^2} \mathbf{H} \right) \quad (100)$$

The second term here is a vector directed from  $\mathbf{m}$  to  $\mathbf{H}$ . The constant  $\lambda$  is  $\sim 10^9$  in accordance with the fact that the relativistic interaction is weak. We disregard here altogether the variation of the absolute value of  $\mathbf{m}$ .

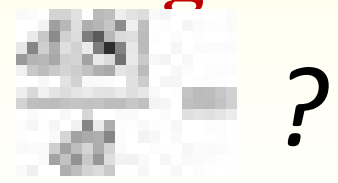
$$\frac{d\mathbf{S}}{dt} = -\gamma \mathbf{S} \times \mathbf{H}$$

$$\gamma = g \frac{e}{2m} = 0.28 \text{ GHz/T}$$

Typical laboratory fields  $\sim 1\text{T}$   
 → Reversal time  $\sim 1 \text{ ns!}$

## Magnetism on the timescale of exchange:

→ *LONGITUDINAL spin dynamics*





# Phenomenological theory\*

“ **Onsager** relations for spin dynamics

Generalized force  $F_i = H_i(\mathbf{r}, t)$  =effective magnetic field

Generalized flux  $J_i = dS_i(\mathbf{r}, t)/dt$

“ Equation of motion from symmetry relations

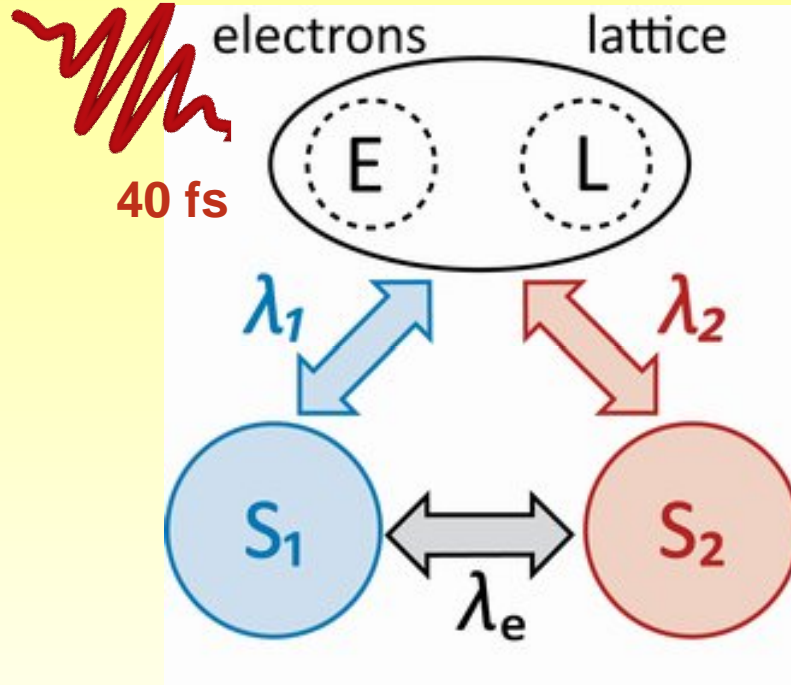
$$d\mathbf{S}/dt = -\gamma \mathbf{S} \times \mathbf{H} + \lambda \mathbf{H} + \dots$$

**Precession**  
| slow+

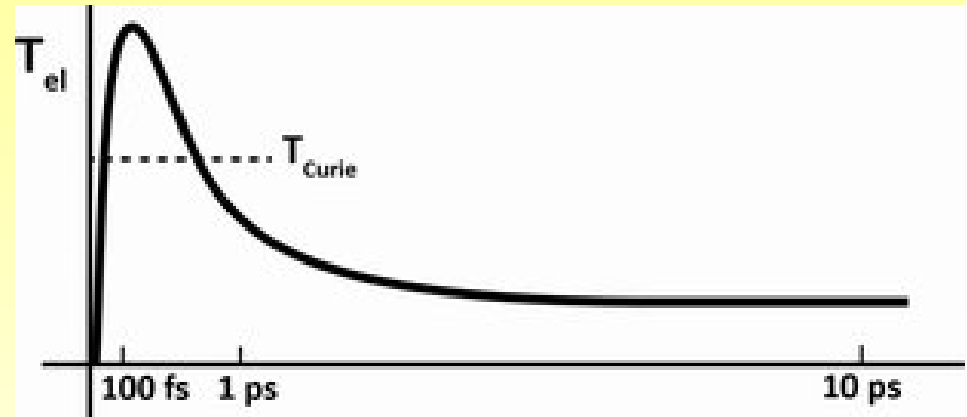
**Longitudinal change |M|**  
**FAST**

\*Baryakhtar JETP 1984

# second laser excitation



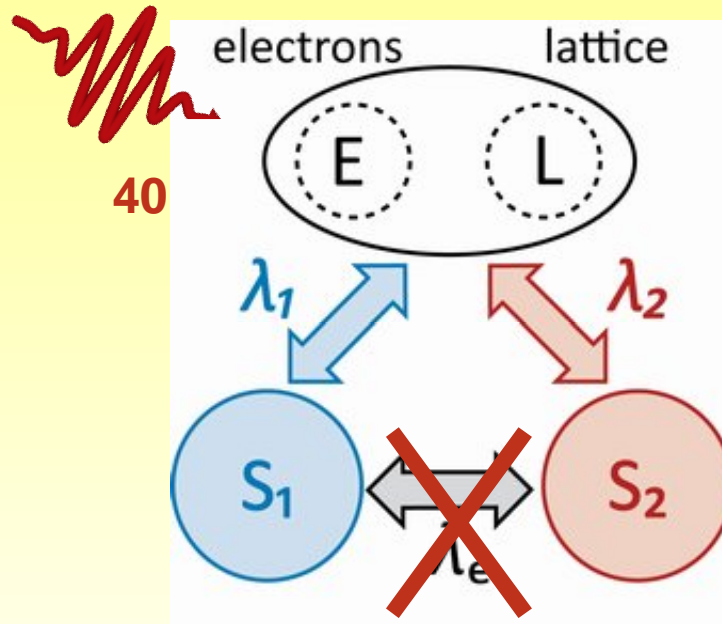
## Ultrafast heating electrons



# Temperature dominated:

$$T \gg T_{Curie}$$

$t \sim 100$  fs



## Bloch relaxation

$$dS_i/dt = -S_i/\tau_i$$

$$\tau_i = \mu_i / (2\alpha_i \gamma k_B T)$$

Dynamics scales with magnetic moment

$$\mu_2 < \mu_1 \Rightarrow \tau_2 < \tau_1$$

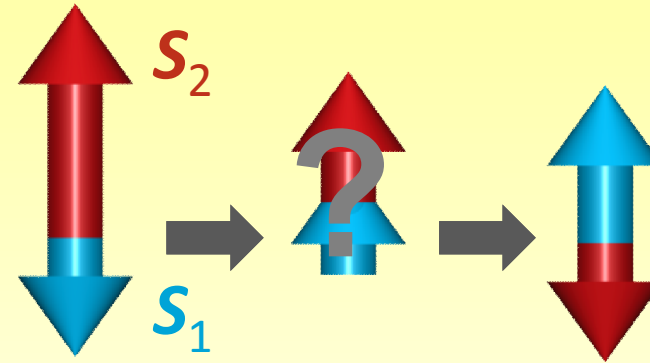
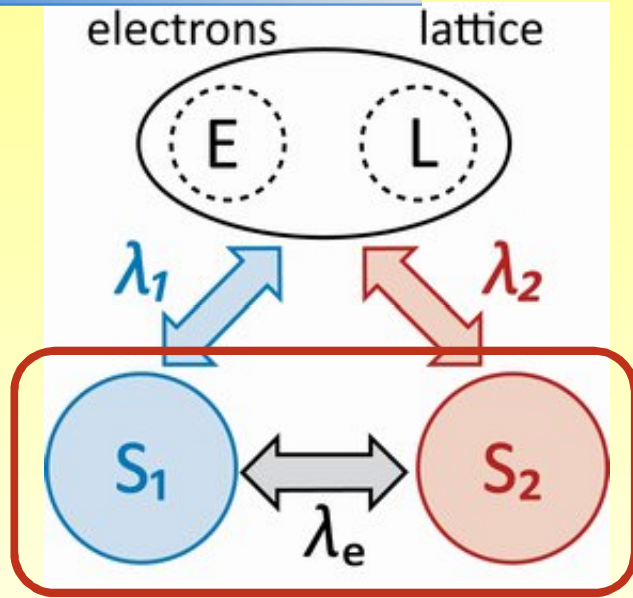
$$\mu_{Fe} < \mu_{Gd}$$



$$\tau_{Fe} < \tau_{Gd}$$

**Distinct** dynamics Gd and Fe

dominated:  $T < T_{Curie}$   $t \sim 1$  ps



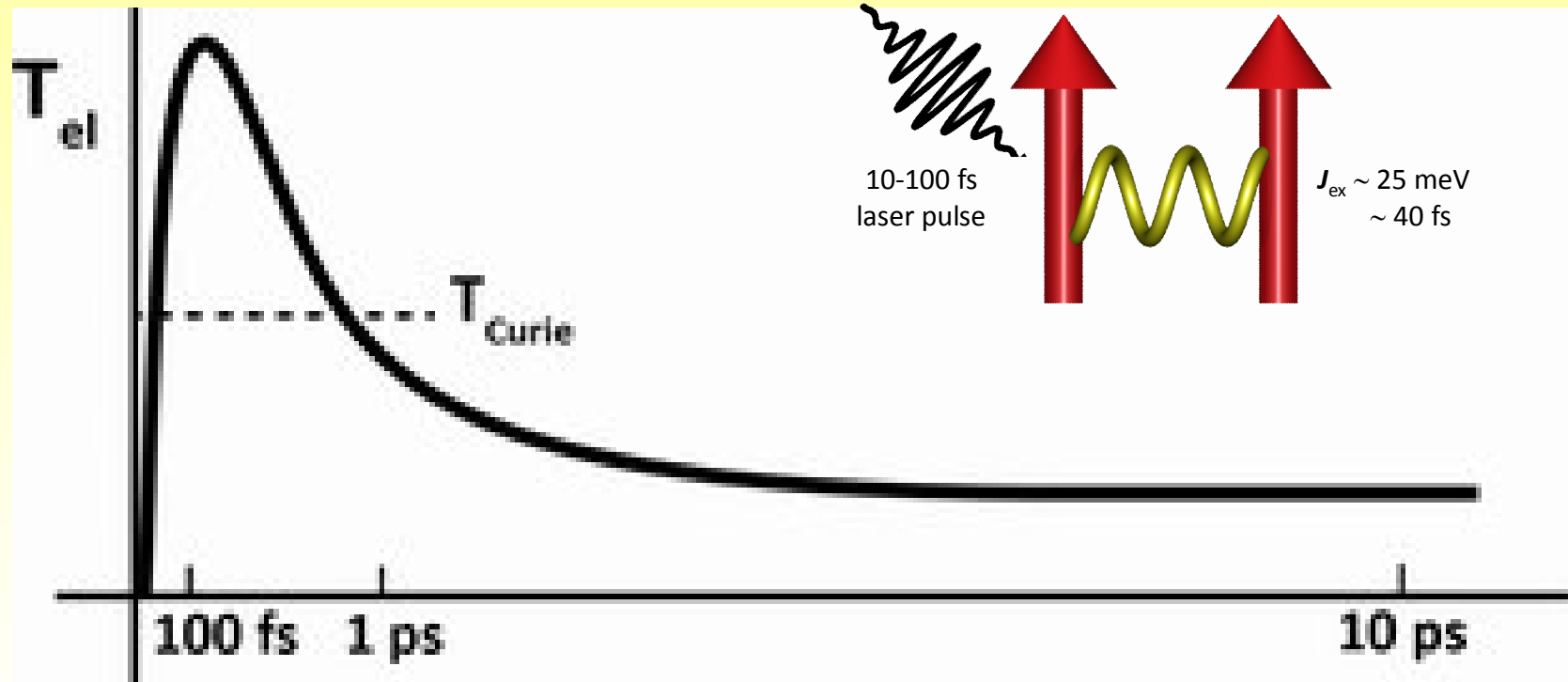
$$dS_1/dt = -dS_2/dt$$

Conservation total angular momentum

Ground state AFM, transient FM!

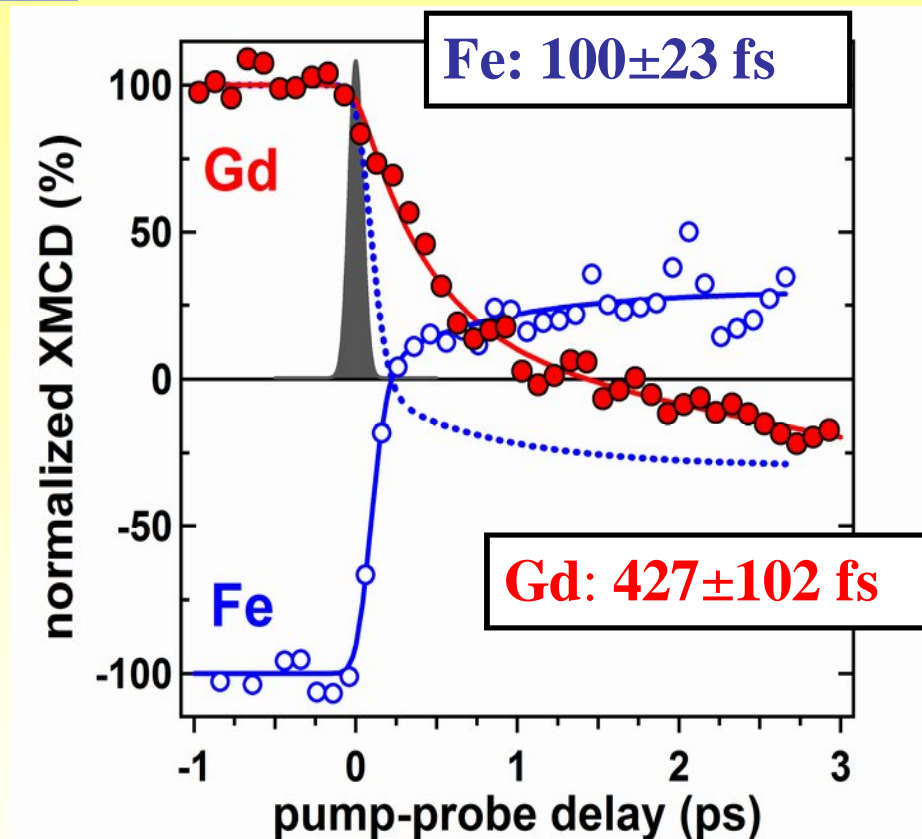
# Laser-induced spin dynamics

## Ultrafast heating of electrons



**Access to temperature and exchange dominated regime!**

# specific magnetization reversal



**Different magnetization switching dynamics Fe and Gd !!!**

**Mentink:** 
$$\tau_i = \frac{\mu_i}{2\alpha\gamma k_B T}$$

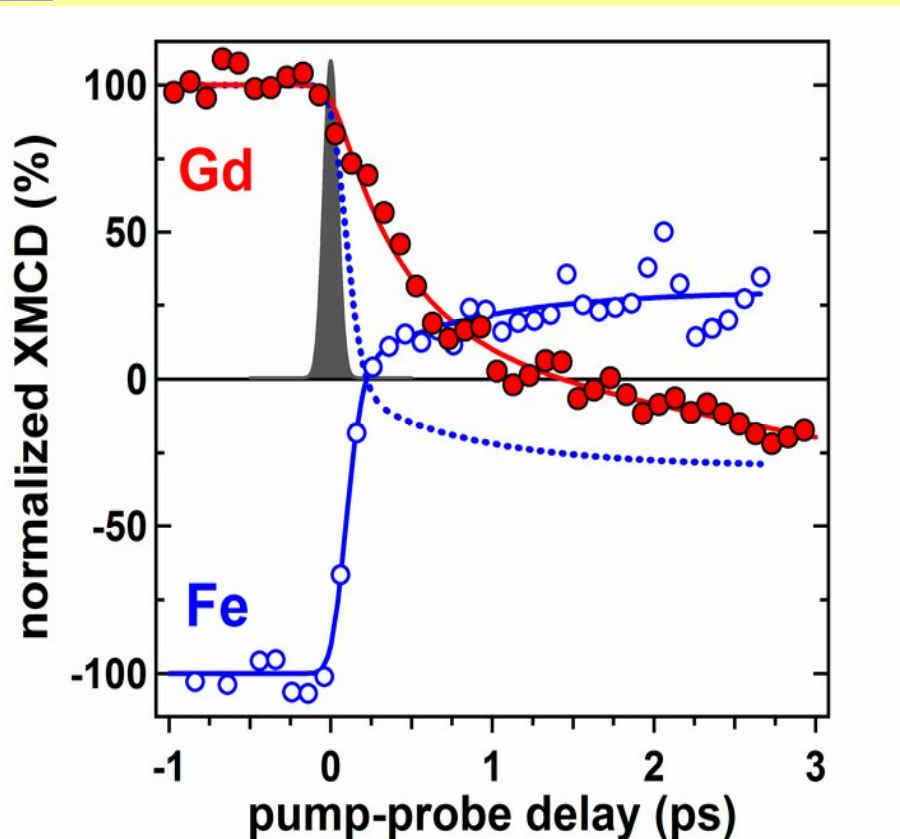


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# *Consequences?*

# duced magnetization reversal!

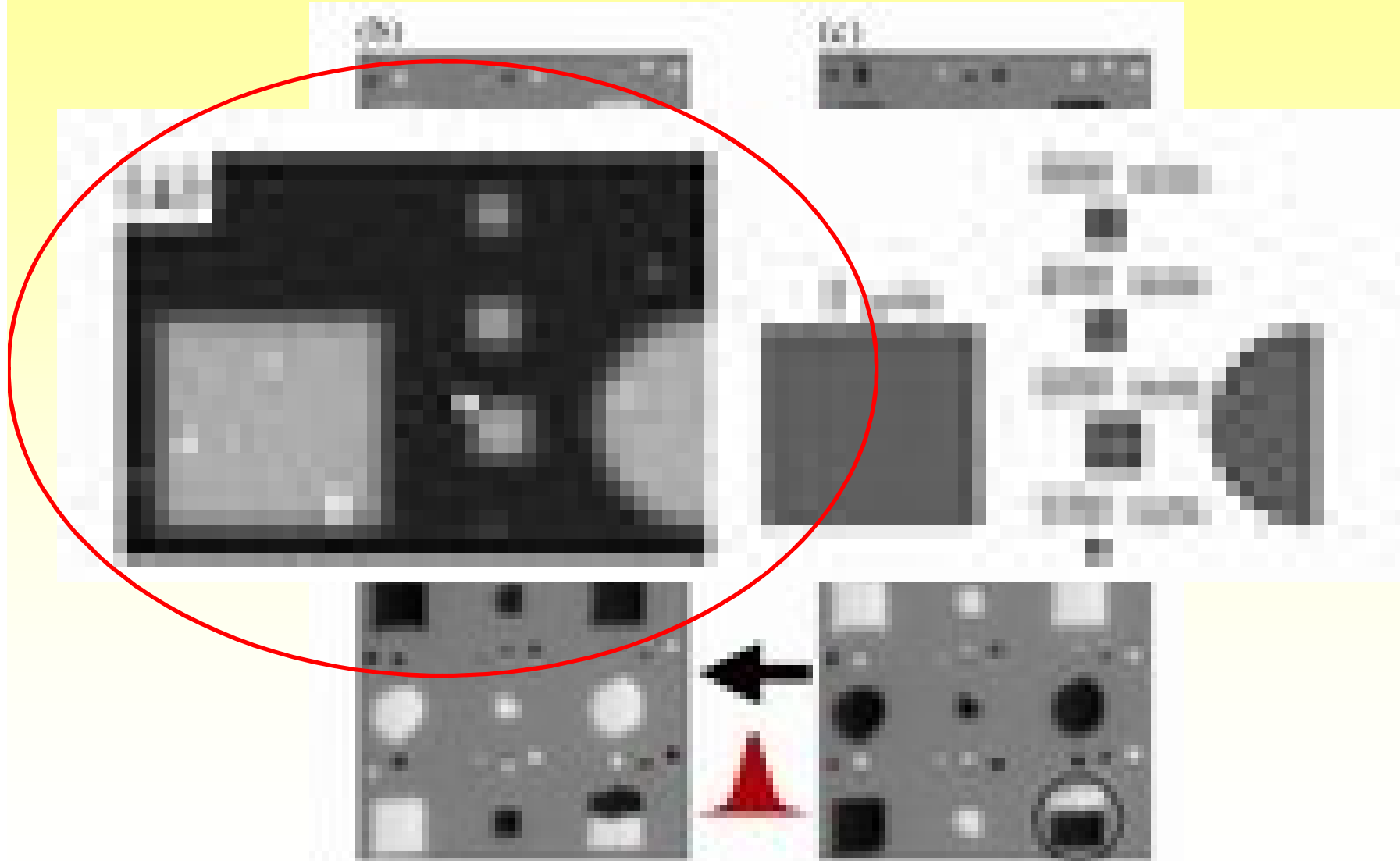


**reversal of magnetization driven by exchange!!!**

**T. Ostler et al, Nature Comm. 3, 666 (2012)**

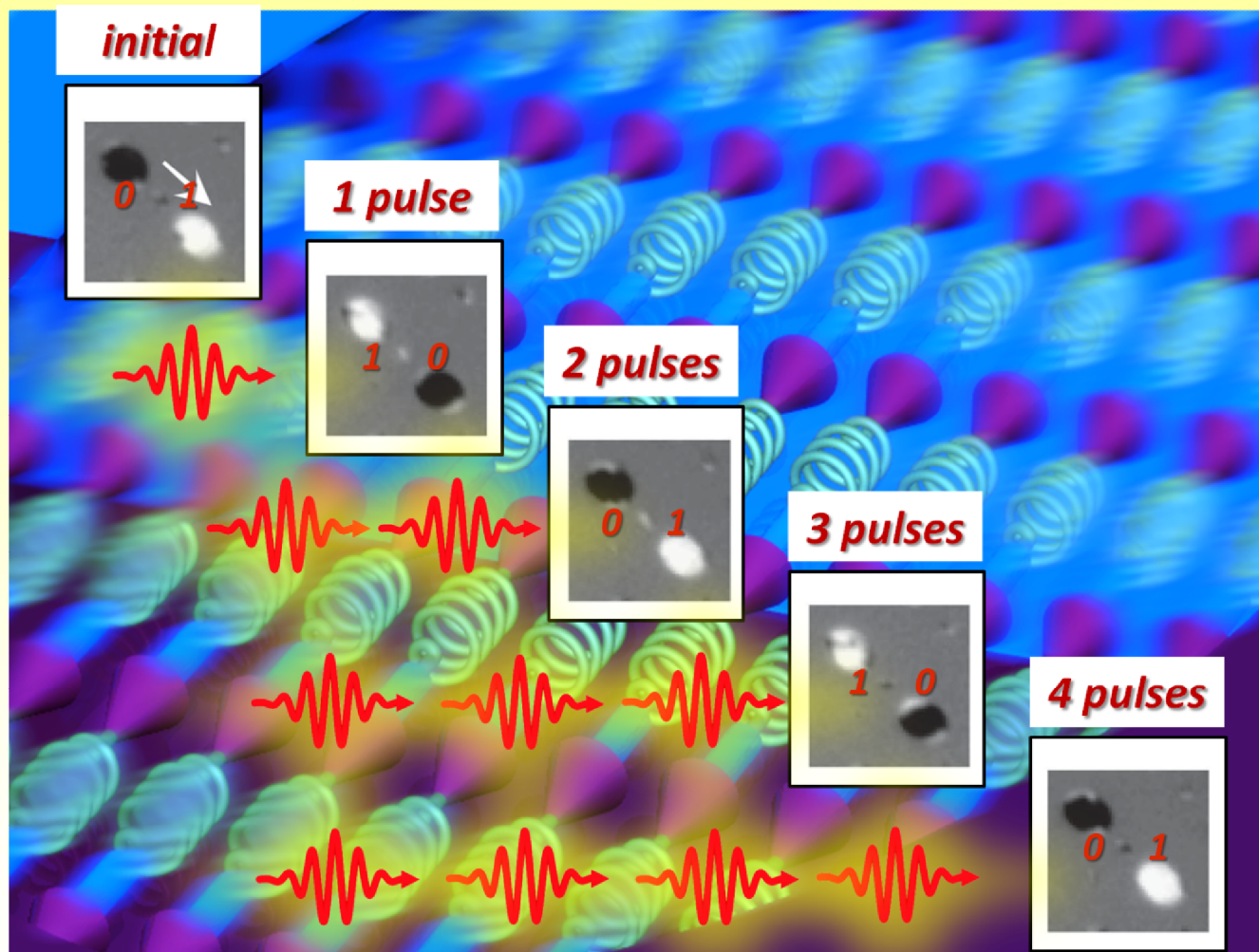


# at induced switching



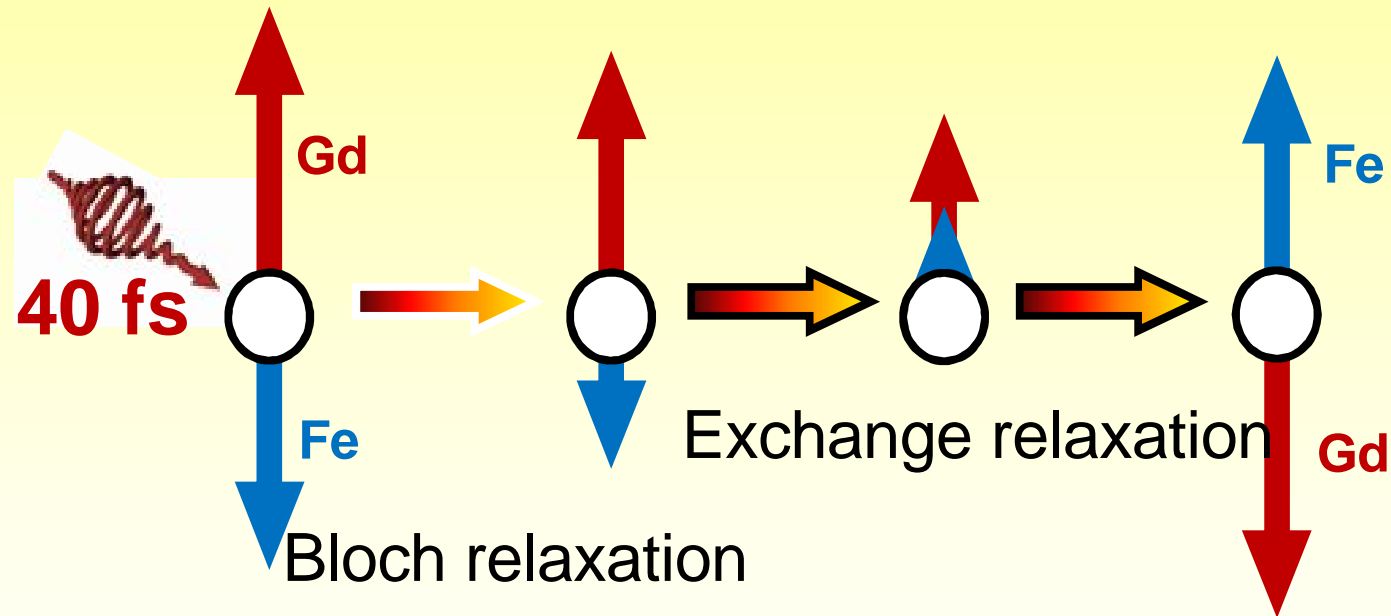
L. Le Guyader et al, APL to appear

# Fast *THERMAL* Switching



\* Ostler et al., Nature Communications 2011  
%Magnetization Reversal, Patent Pending

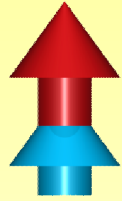
# Ultrafast magnetism in multi-sublattice magnets



What about multi-sublattice ferromagnets?

# : ultrafast demagnetization

## FM coupling

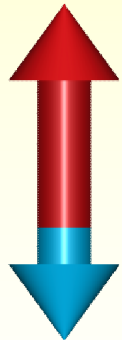


$$dS_1/dt = -\lambda_1 S_1/\chi_1 - \lambda_c(S_1/\chi_1 - S_2/\chi_2)$$

positive/negative

- Coupling makes one sublattice **faster**, other **slower**

## AFM coupling



$$dS_1/dt = -\lambda_1 S_1/\chi_1 - \lambda_c(S_1/\chi_1 + |S_2|/\chi_2)$$

always positive

- Both sublattices **faster** than in uncoupled case!

clude:

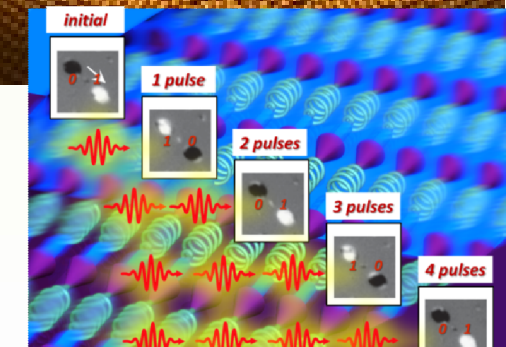
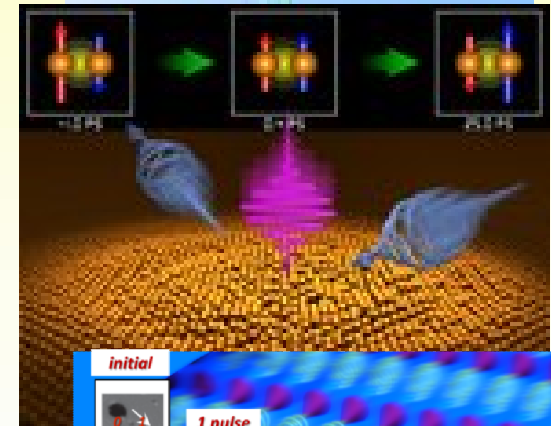
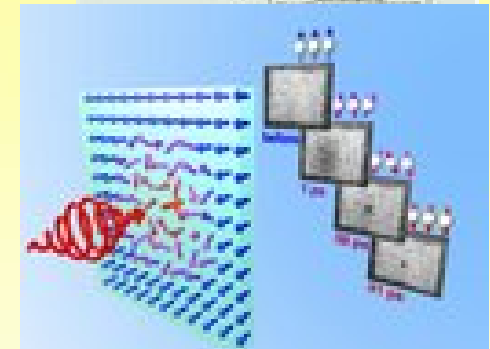
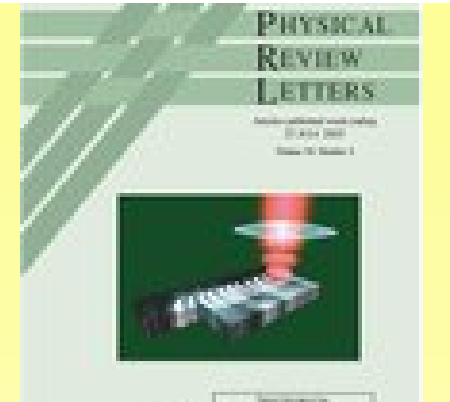
## With light

- Coherent optical control of magnetism !
- All-optical ultrafast magnetic recording
- Novel (linear) ultrafast reversal path!
- Novel transient ferromagnetic state
- Heat driven deterministic switching

## Future challenges

- Femto magnetism!
- Combine chemical, magnetic

spatial and time resolution: **X(Z)FEL!**



# y thanks to:

## **IMM**

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A. Tsukamoto

## **University of Konstanz**

Denise Hinzke

Ulrich Nowak

## **Bessy**

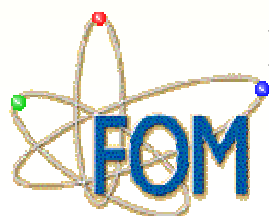
I. Radu

C. Stamm

T.Kachel

N.Pontius

Herman Durr (Stanford)



**STW, NWO, EU, NanoNed, UltraMagnetron, FANTOMAS**

# copy of *Solids and Interfaces*

Several PhD / postdoc positions available!



2011