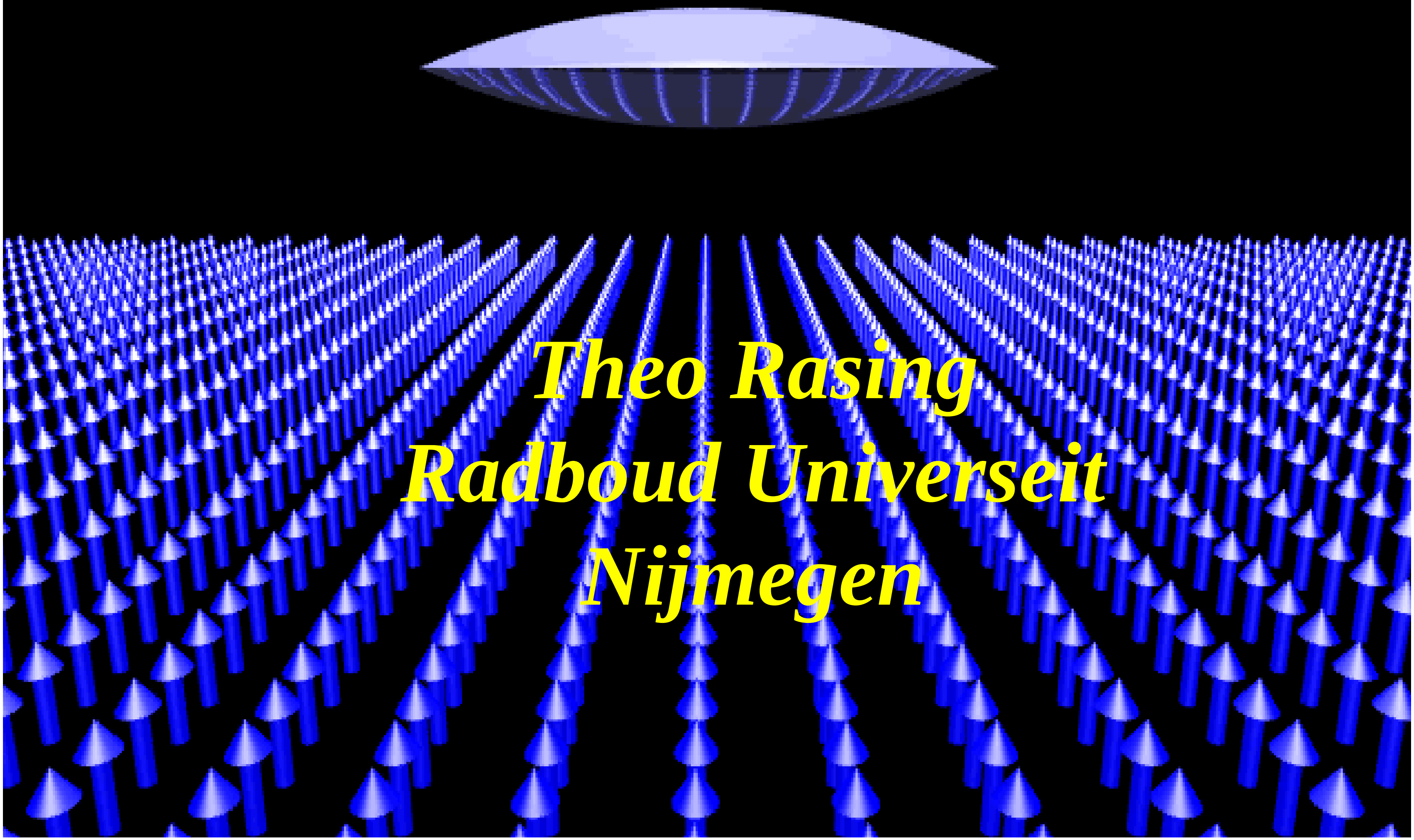
# *Emerging Magnetic Order*

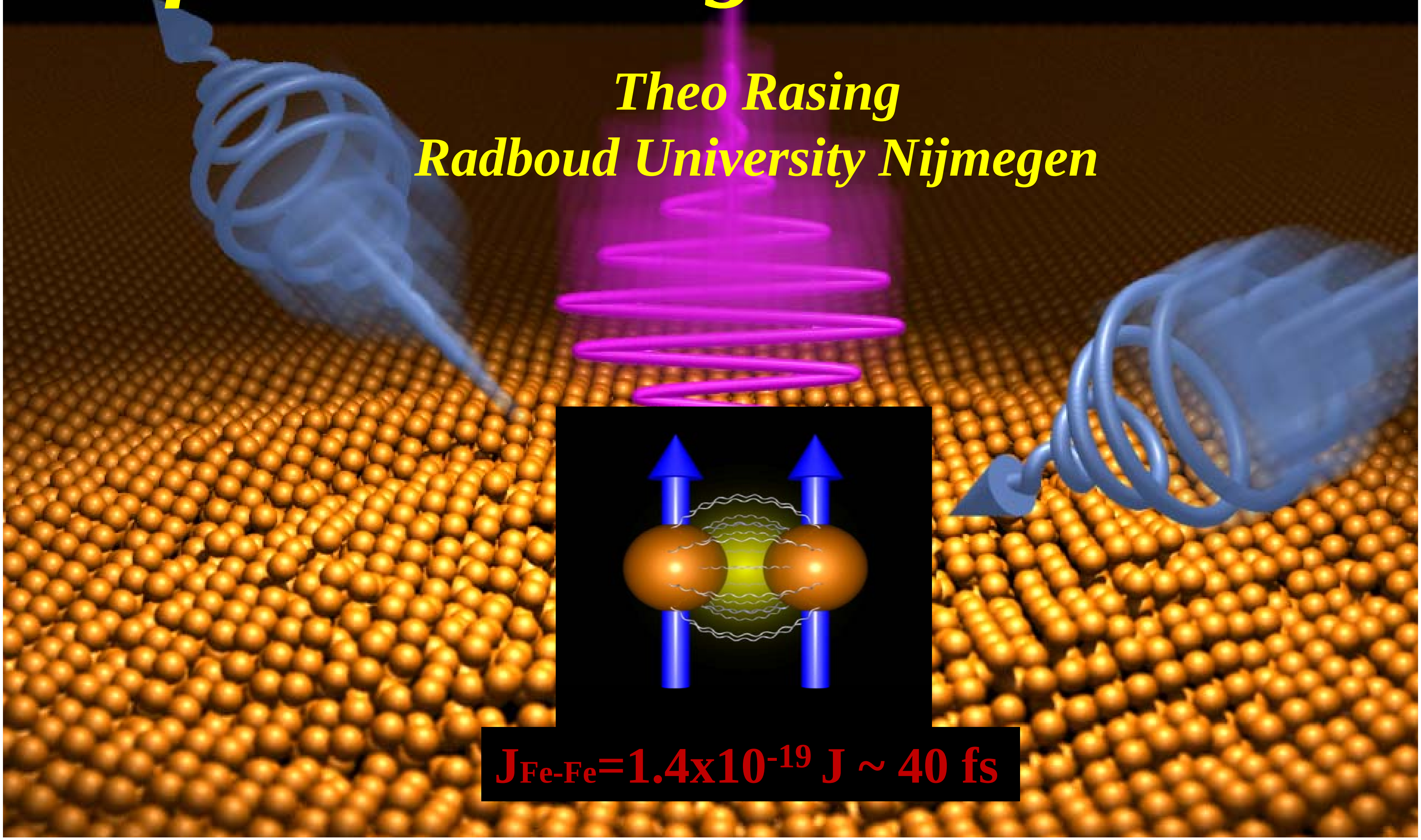


*Theo Rasing*
*Radboud Universeit*
*Nijmegen*

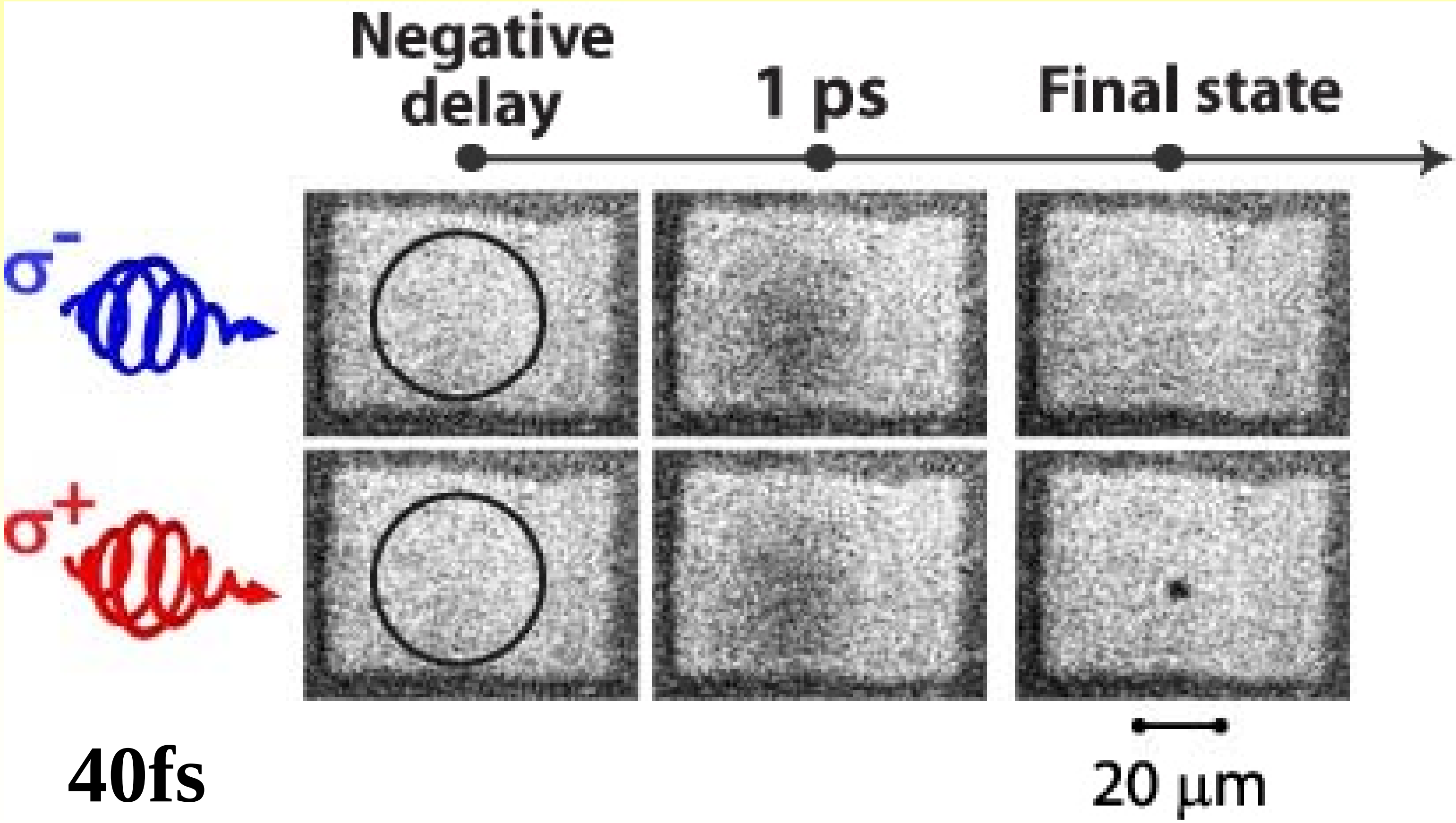
# *Magnetism on the timescale of the exchange interaction*

# *Challenges and Opportunities*

*Theo Rasing*
*Radboud University Nijmegen*

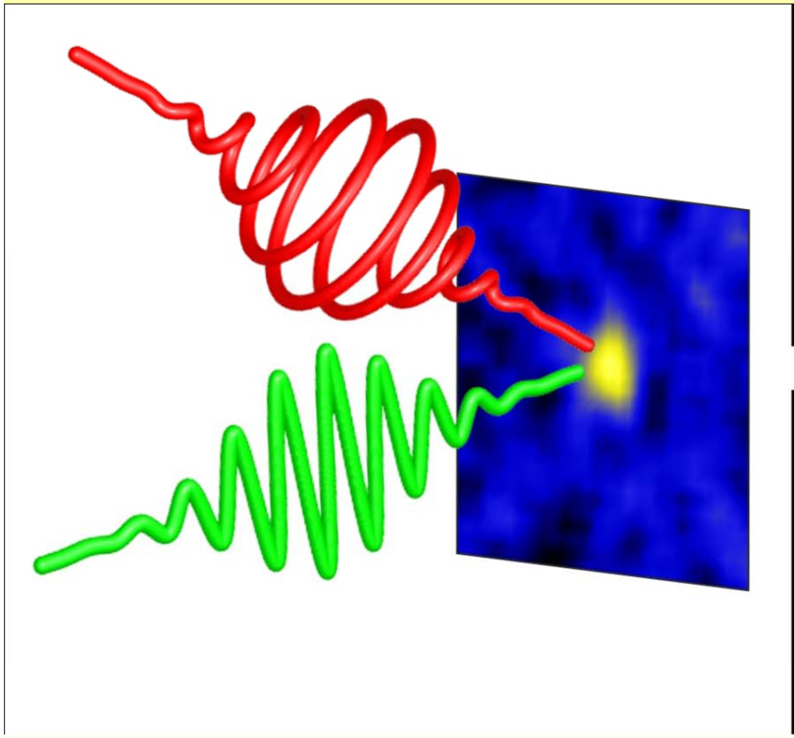


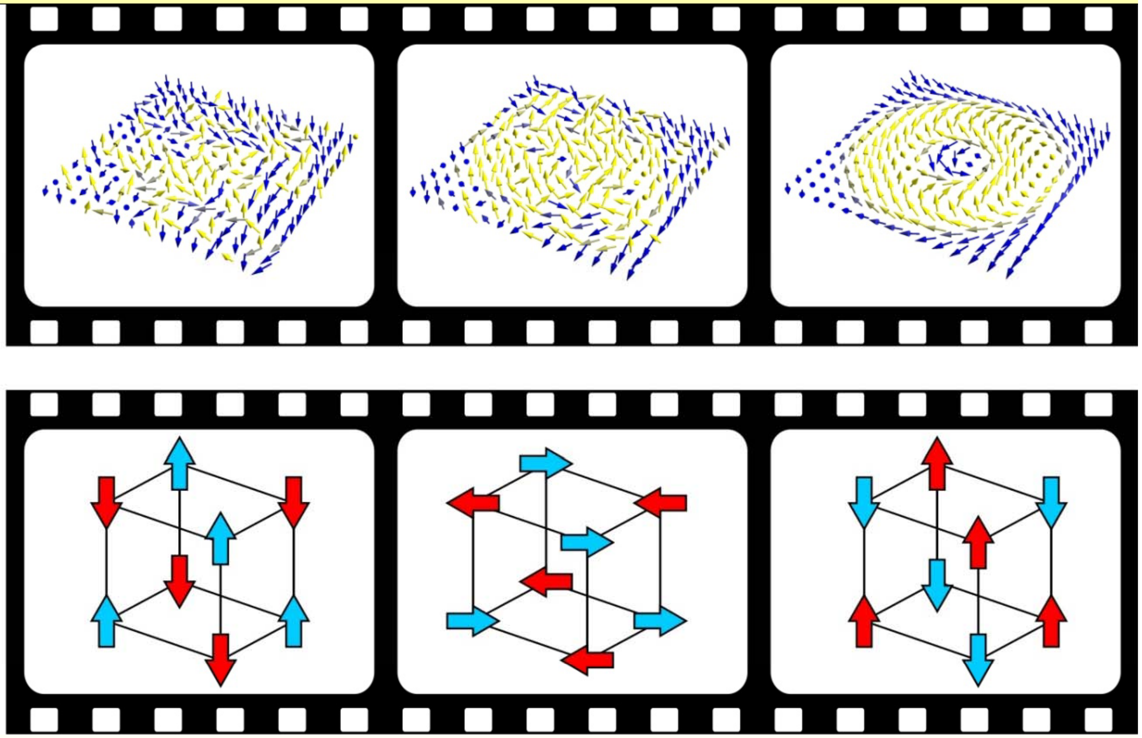
# *Emerging Magnetic Order*



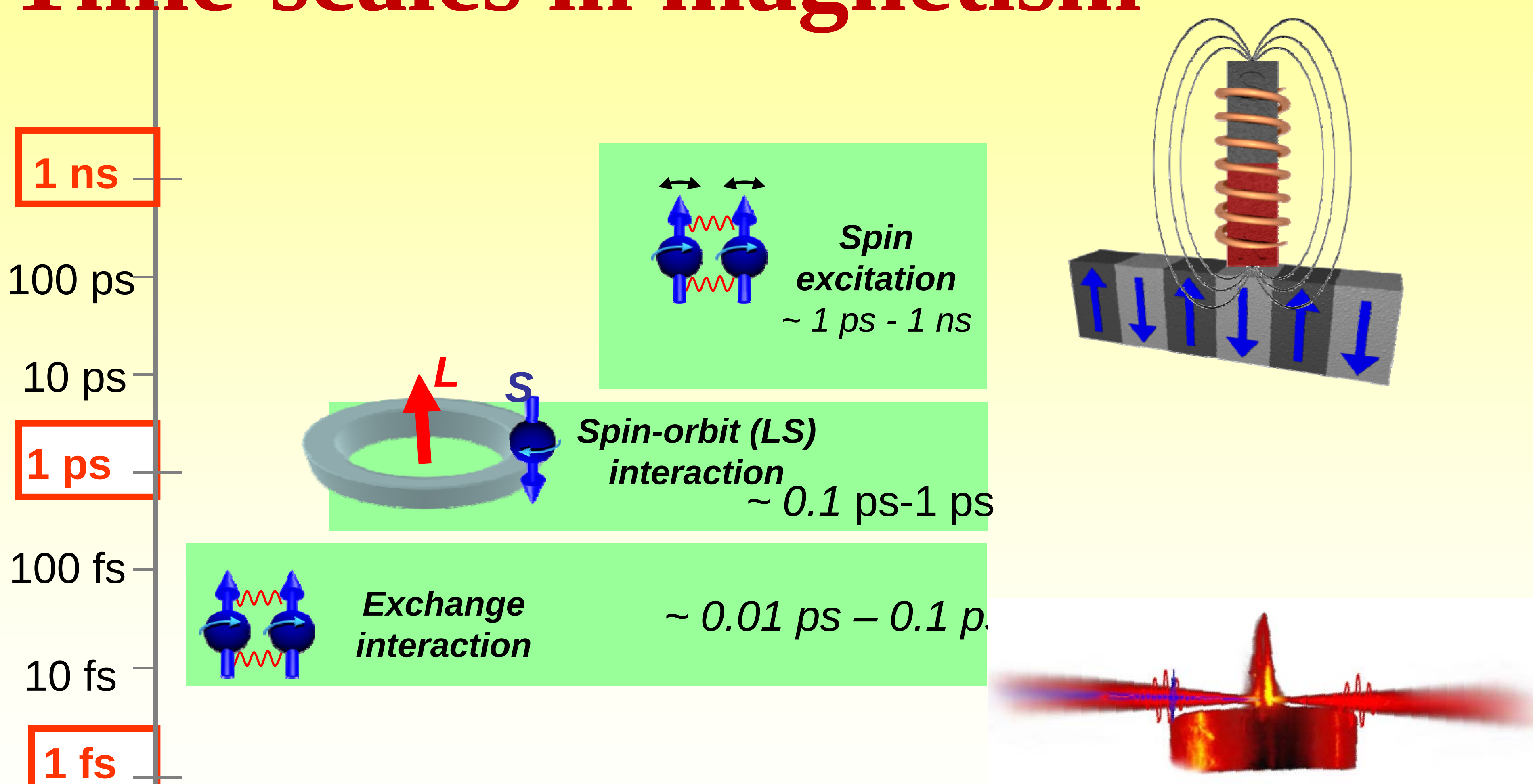

**40fs**

# *Emerging Magnetic Order*





# Time-scales in magnetism




**10-100 femtosecond!**

# Length-scales in magnetism

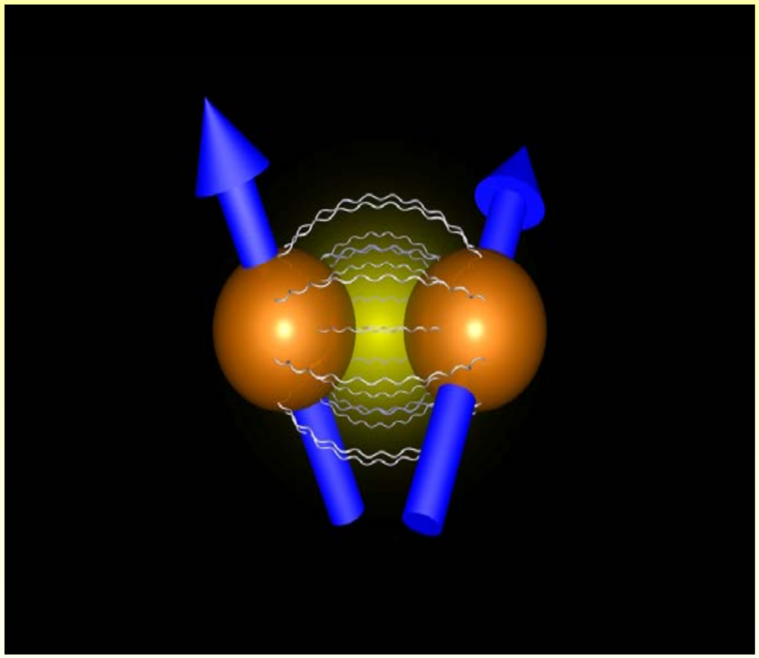
$$l = \sqrt{J/K} \sim nm$$

**Tb+THz**

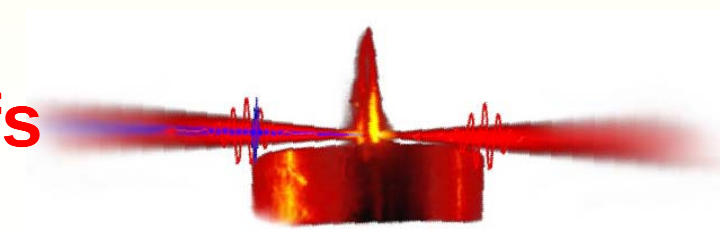
**nm+fs!**

# New Mechanisms?

$J_{Fe-Fe} = 1.4x10^{-19}$ J → 40 fs

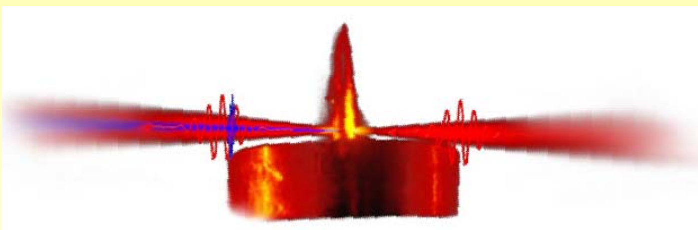


**1-2eV,10-100fs**

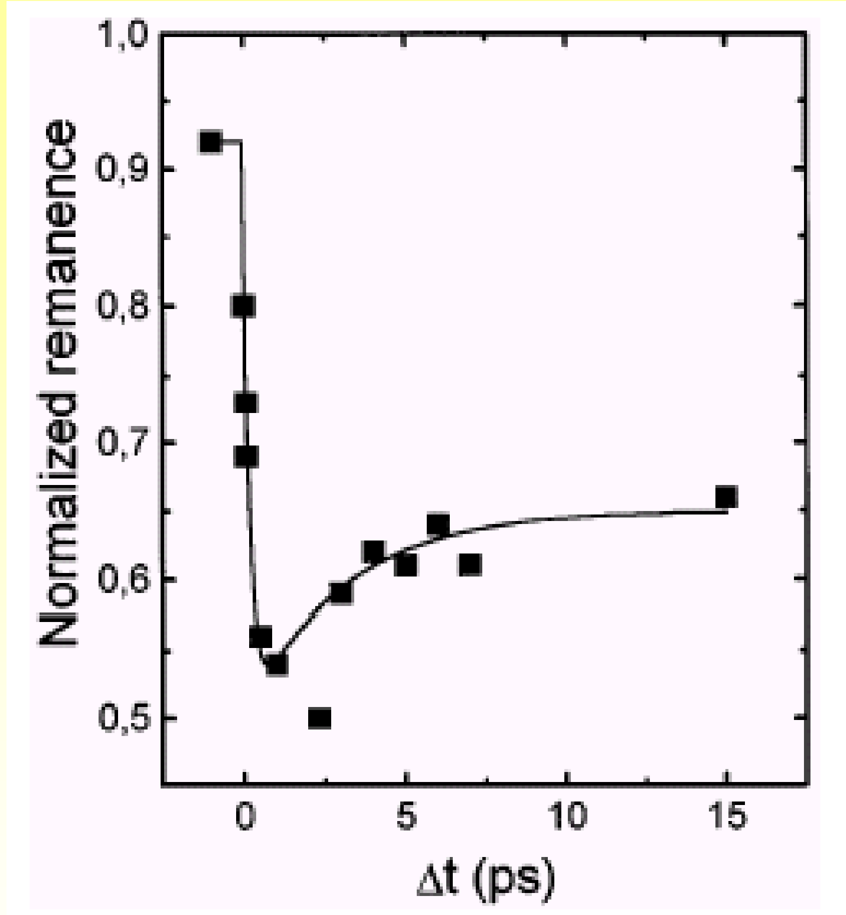


?

# 1996 Beaurepaire et al:

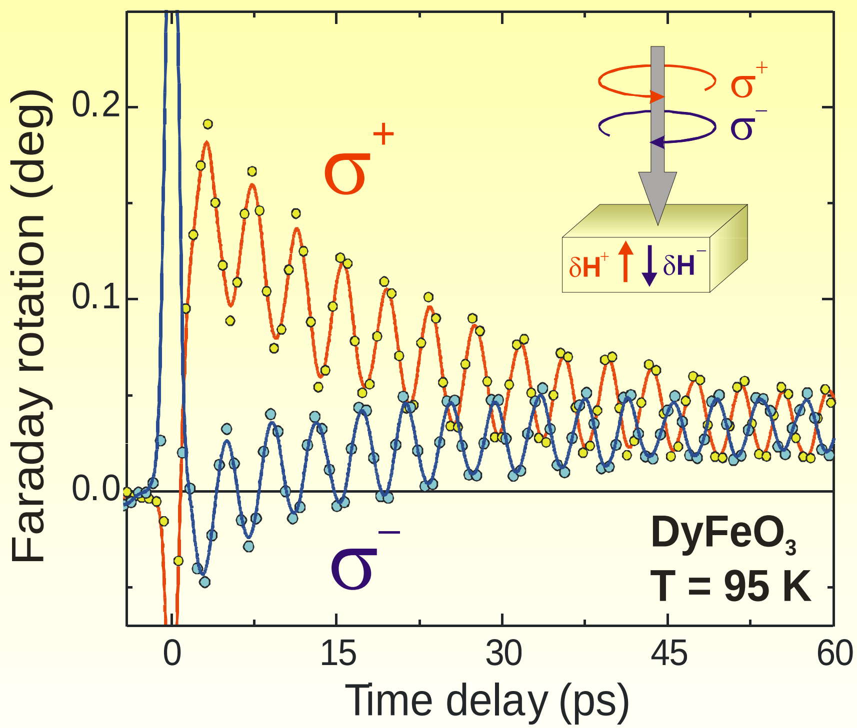


60fs




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

# Ultrafast excitation of spins in $DyFeO_3$ via the Inverse Faraday effect

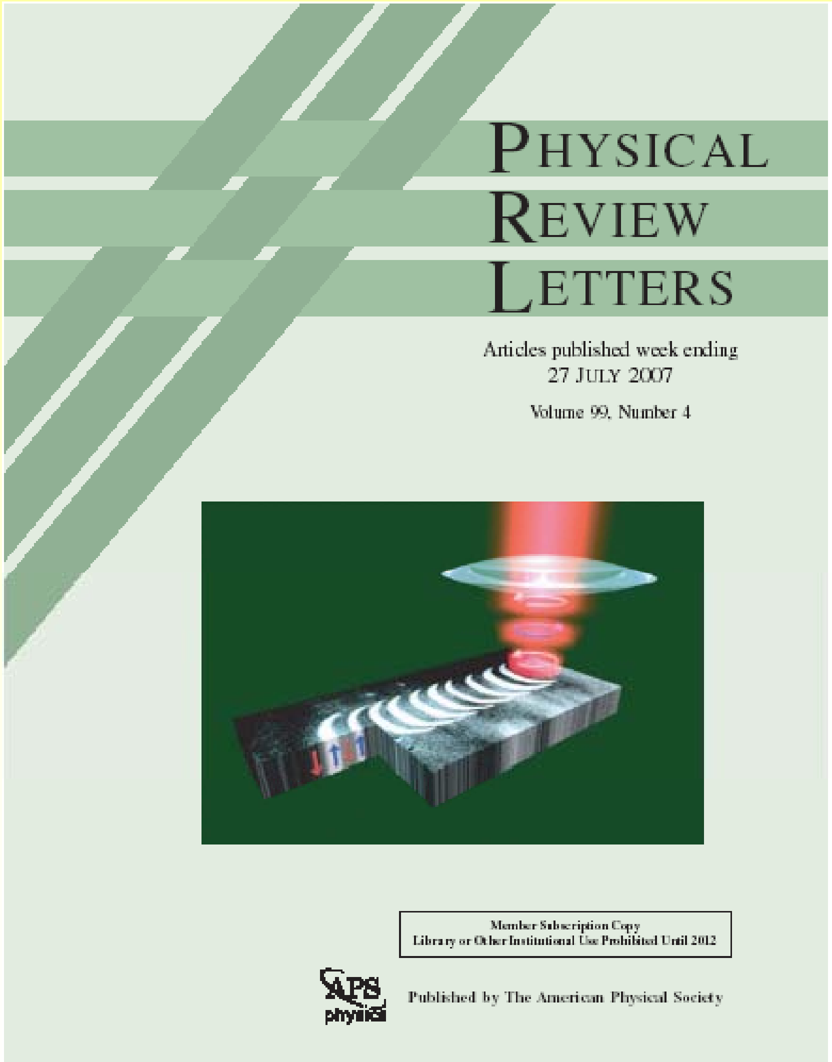



A. Kimel et al, *Nature* **435** 655 (2005).

## Referee A:

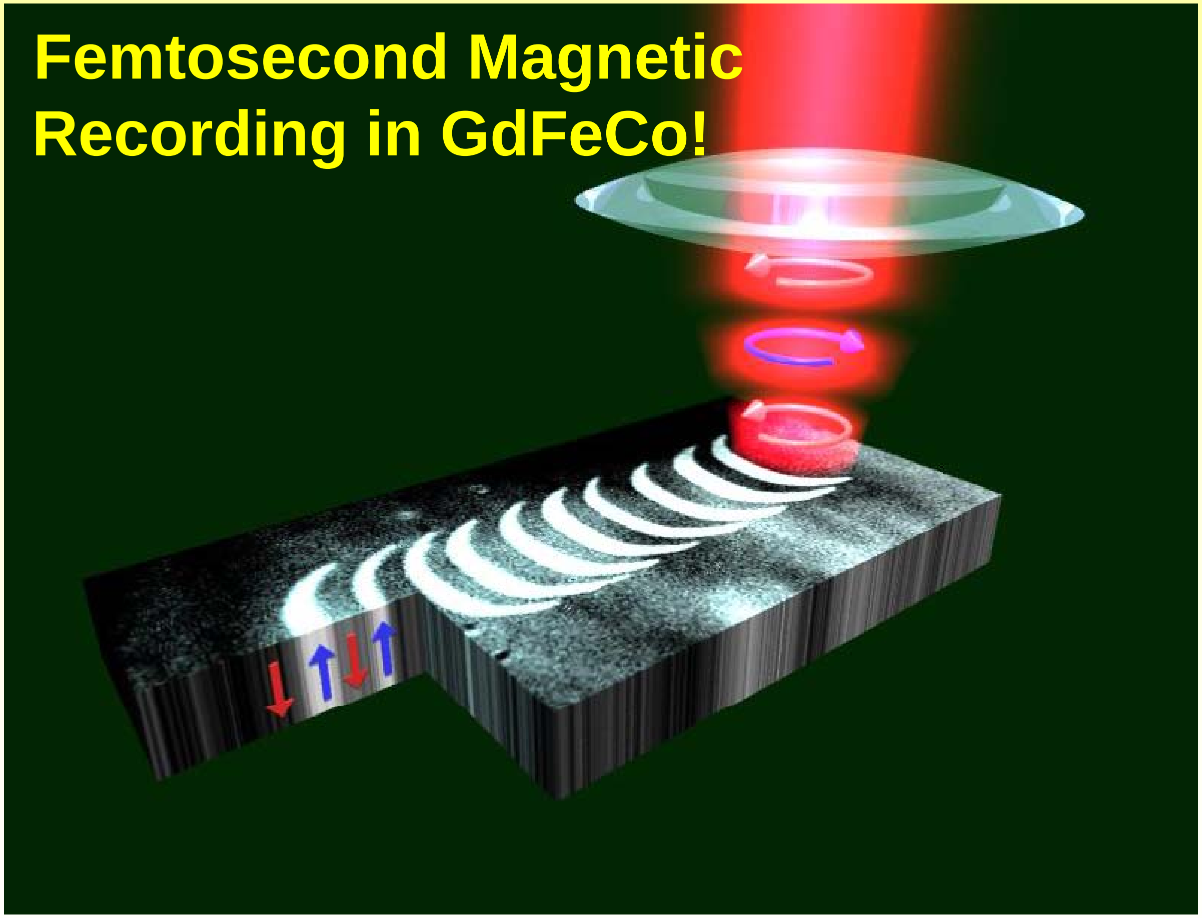
***"But, unfortunately, the observed signal is so small that it seems impractical to utilize the inverse Faraday effect for the purpose of ultrafast control of magnetization in metallic materials."***

# Ultrafast all optical recording

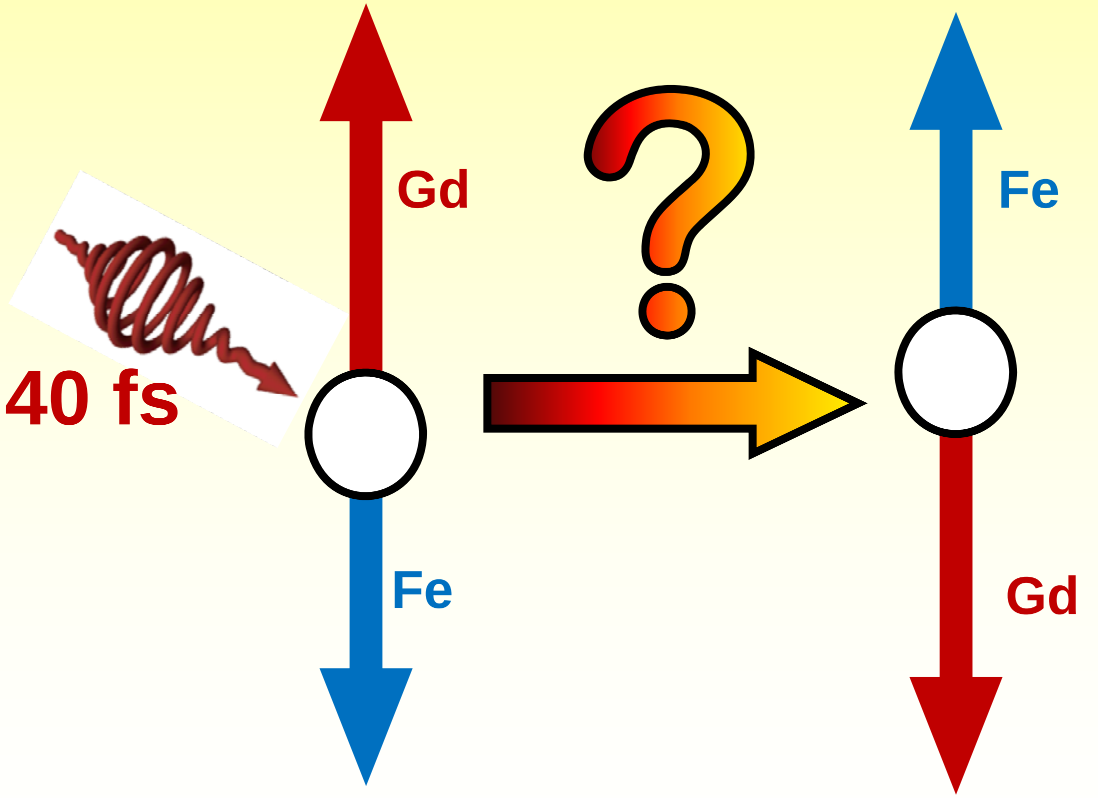


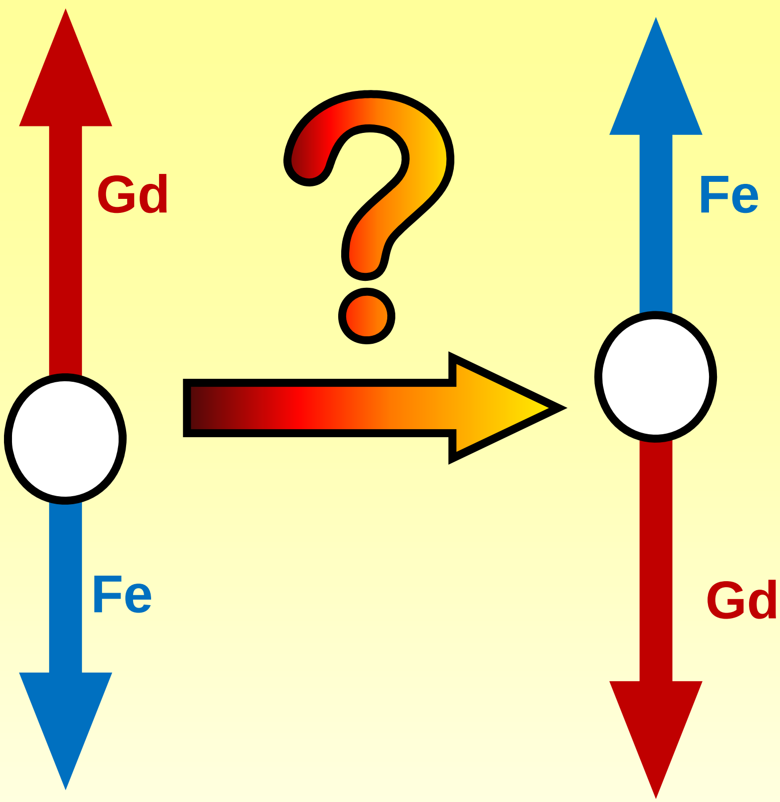

C.D. Stanciu et al., *patent* #P77323PC00, PRL **99**, 047601 (2007)

# GdFeCo: multi-sublattice ferrimagnet!



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






**Angular momentum transfer?**

**Role of temperature?**

**Role of sublattices?**

**.....**

# Beyond Landau-Lifshitz dynamics

ON THE THEORY OF THE DISPERSION OF MAGNETIC PERMEABILITY IN FERROMAGNETIC BODIES.

By L. Landau and E. Lifshitz.

(Received June 3, 1935).

magnetic moment. Therefore in the presence of the field the magnetic moment would act as a free moment, i. e. would rotate around **f** and we should have for **s** (˙ denotes differentiation by time) the equation

$$\dot{\mathbf{s}}/\mu_0 = [\mathbf{fs}] + \lambda\left(\mathbf{f} - \frac{(\mathbf{fs})\mathbf{s}}{s^2}\right). \qquad (21)$$

• The second term here is a vector directed from **s** to **f**. The constant $\lambda$ is $\lambda \ll s$ in accordance with the fact that the relativistic interaction is weak. We disregard here altogether the variation of the absolute value of **s**.

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

$\gamma = g\frac{e}{2m}$ **=0.28 Ghz/T**

**Typical laboratory fields ~ 1T**

**→ Reversal time ~ 1 ns!**

## Magnetism on the timescale of exchange:

→ ***LONGITUDINAL spin dynamics*** $\frac{d|\mathbf{S}|}{dt} =$ ?

# Onsager's relations for spin dynamics

Baryakhtar, JETP 1984

**Magnetic free energy**

$$F = \int \{f(S^2) - \mathbf{S}\cdot\mathbf{H}_0 - K_u S_z^2 + \ldots\}\, dV$$

$$\frac{dF}{dt} = -\int \frac{d\mathbf{S}(\mathbf{r},\mathbf{t})}{dt}\cdot\mathbf{H}(\mathbf{r},t)\, dV$$

Generalized force
$\mathbf{H}(\mathbf{r},t) = -\delta F/\delta\mathbf{S}(\mathbf{r},t)$

Generalized position
$\mathbf{S}(\mathbf{r},t)$ *angular momentum*

# Symmetry of kinetic coefficients

$$dS_i(\mathbf{r}',t)/dt = \int \lambda_{ik}(\mathbf{r}'-\mathbf{r},\mathbf{S}(\mathbf{r}))H_k(\mathbf{r}',t)dV$$

$$= \lambda_{ik}(\mathbf{S})H_k(\mathbf{r},t) + \lambda_{ik,lm}\partial^2 H_k/\partial r_l\partial r_m + \ldots$$

$$\lambda_{ik}(\mathbf{S}) = \lambda_{ki}(-\mathbf{S})$$

$$\lambda^s_{ik}(\mathbf{S}) = (\lambda_{ik}+\lambda_{ki})/2$$

$$\lambda^a_{ik}(\mathbf{S}) = (\lambda_{ik}-\lambda_{ki})/2$$

$$d\mathbf{S}/dt = \mathbf{S}\times\mathbf{H} + \lambda^s\mathbf{H} + \ldots$$

Anti-symmetric precession — Symmetric longitudinal

# More general: multiple sublattices

## Rate of change free energy

$$\frac{dF}{dt} = -\int \left\{ \frac{d\mathbf{S}_1}{dt} \cdot \mathbf{H}_1 + \frac{d\mathbf{S}_2}{dt} \cdot \mathbf{H}_2 \right\} dV$$

## Longitudinal dynamics to lowest order

$$\frac{d}{dt}\begin{pmatrix} S_1 \\ S_2 \end{pmatrix} = \begin{pmatrix} \lambda^s_{11} & \lambda^s_{12} \\ \lambda^s_{21} & \lambda^s_{22} \end{pmatrix} \begin{pmatrix} H_1 \\ H_2 \end{pmatrix}$$

$$\begin{pmatrix} \lambda_1 & 0 \\ 0 & \lambda_2 \end{pmatrix} + \begin{pmatrix} \lambda_e & -\lambda_e \\ -\lambda_e & \lambda_e \end{pmatrix}$$

relativistic exchange

$$dS_1/dt + dS_2/dt = 0$$

**J.H. Mentink *et al.*, *Phys. Rev. Lett.* (2012)**

# More general: multiple sublattices

$$dS_1/dt = \lambda_e(H_1 - H_2) + \lambda_1 H_1$$
$$dS_2/dt = -\lambda_e(H_1 - H_2) + \lambda_2 H_2$$

$$F = \int dV \, \{f_1(S_1^2) + f_2(S_2^2) - J_{12}(S_1 \cdot S_2)\}$$

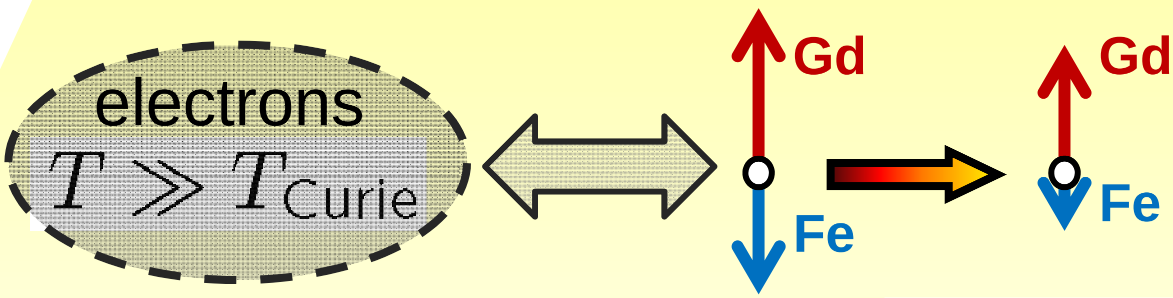
formation sublattice spin moment

mutual alignment sublattices

**Temperature dominated: T>>$T_C$ , t~100fs**

# Temperature dominated regime

**Relativistic relaxation dominates**




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

**Distinct dynamics Gd and Fe** $\tau_{\text{Fe}} < \tau_{\text{Gd}}$

Bloch relaxation

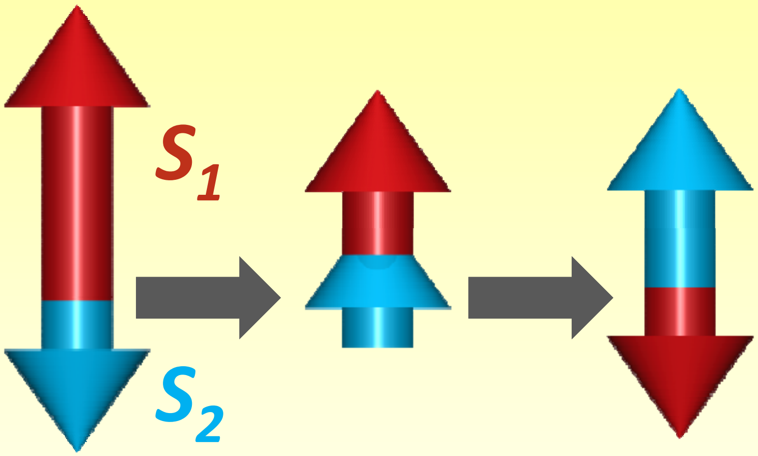
$$\frac{dM_i}{dt} = -\frac{M_i - M_i^{(0)}}{\tau_i}$$

$$\tau_i = \frac{\mu_i}{2\alpha\gamma k_{\text{B}} T}$$

*Brown Phys. Rev. 1963*
*Kubo et al. Prog. Theor. Phys. Suppl. 1970*

# Exchange dominated: $T<T_C$ , t~1ps

## Conservation total angular momentum




$$dS_1/dt = \lambda_e(H_1 - H_2) + \lambda_1 H_1$$
$$dS_2/dt = -\lambda_e(H_1 - H_2) + \lambda_2 H_2$$

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

What if $S_2$=0?

$$dS_2/dt|_{S_2=0} > 0 \Leftrightarrow$$

$$(S_1^2 - \overline{S}_1^2)/\chi_1 > |J_{12}|(1 + \lambda_2/\lambda_e) > 0$$

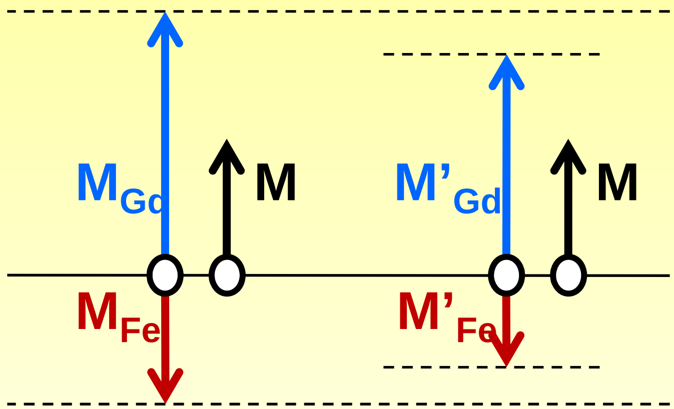
$$f_i(S_i^2) = (S_i^2 - \overline{S}_i^2)^2/4\chi_i$$

**Nonequilibrium** $S_1^2 \gg \overline{S}_1^2$

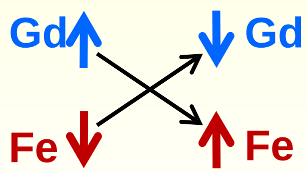
Equilibrium value

# Ground state AFM, transient FM!

# Exchange dominated regime




spin-spin interactions conserve total magnetization **M**



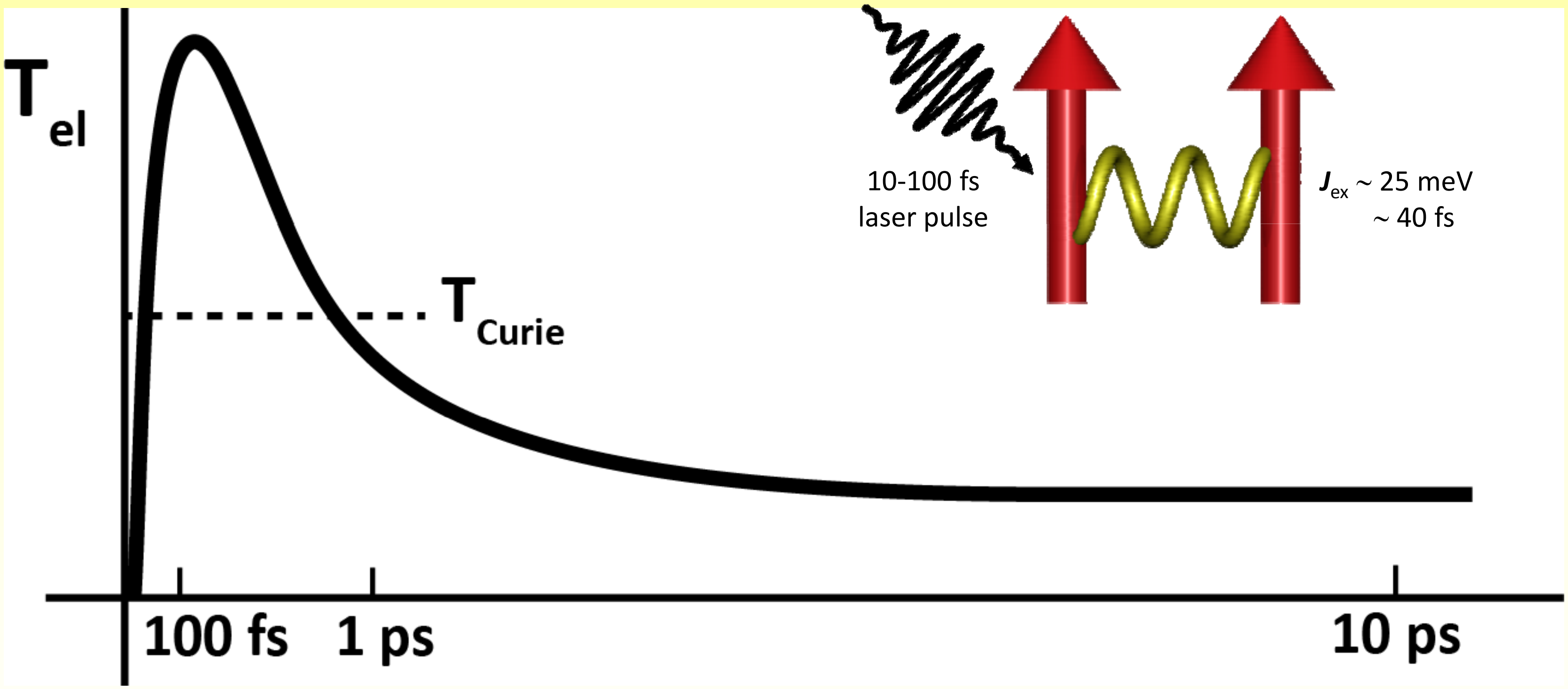

$$\frac{dM_{\mathrm{Fe}}}{dt} = -\frac{dM_{\mathrm{Gd}}}{dt}$$

**Transfer of magnetization between sublattices!**

*Baryakhtar Fiz. Nizk. Temp. 1985*

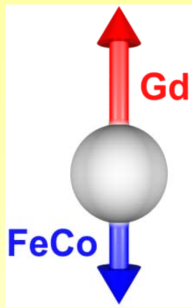
# Laser-induced spin dynamics

**Ultrafast heating of electrons**




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

# Element specific view

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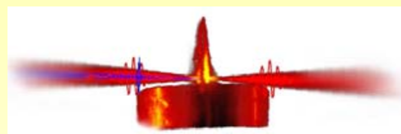
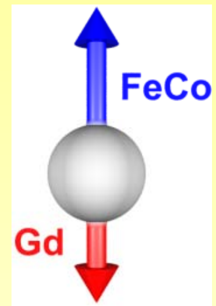
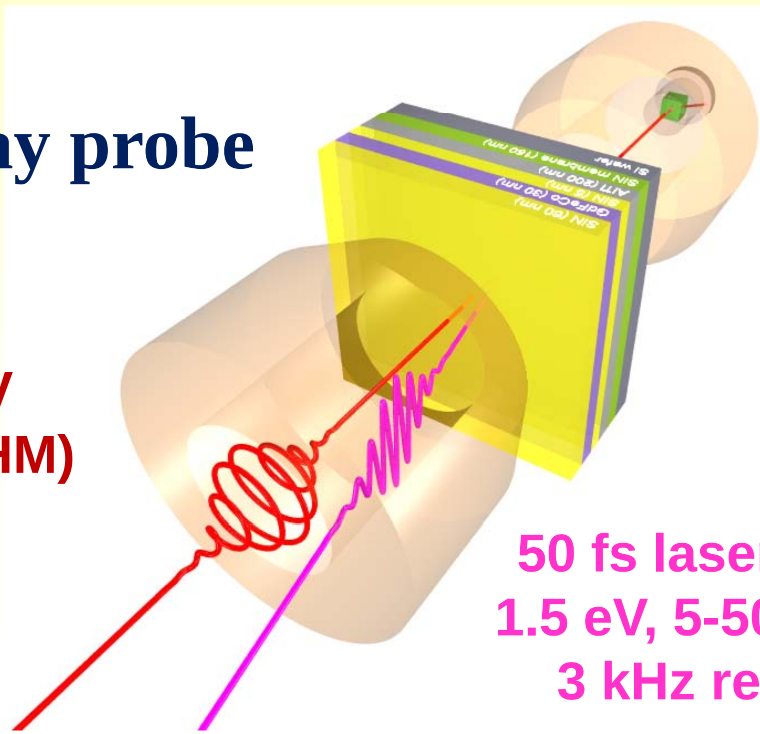





**Laser pump – X-ray probe**

**BESSY**

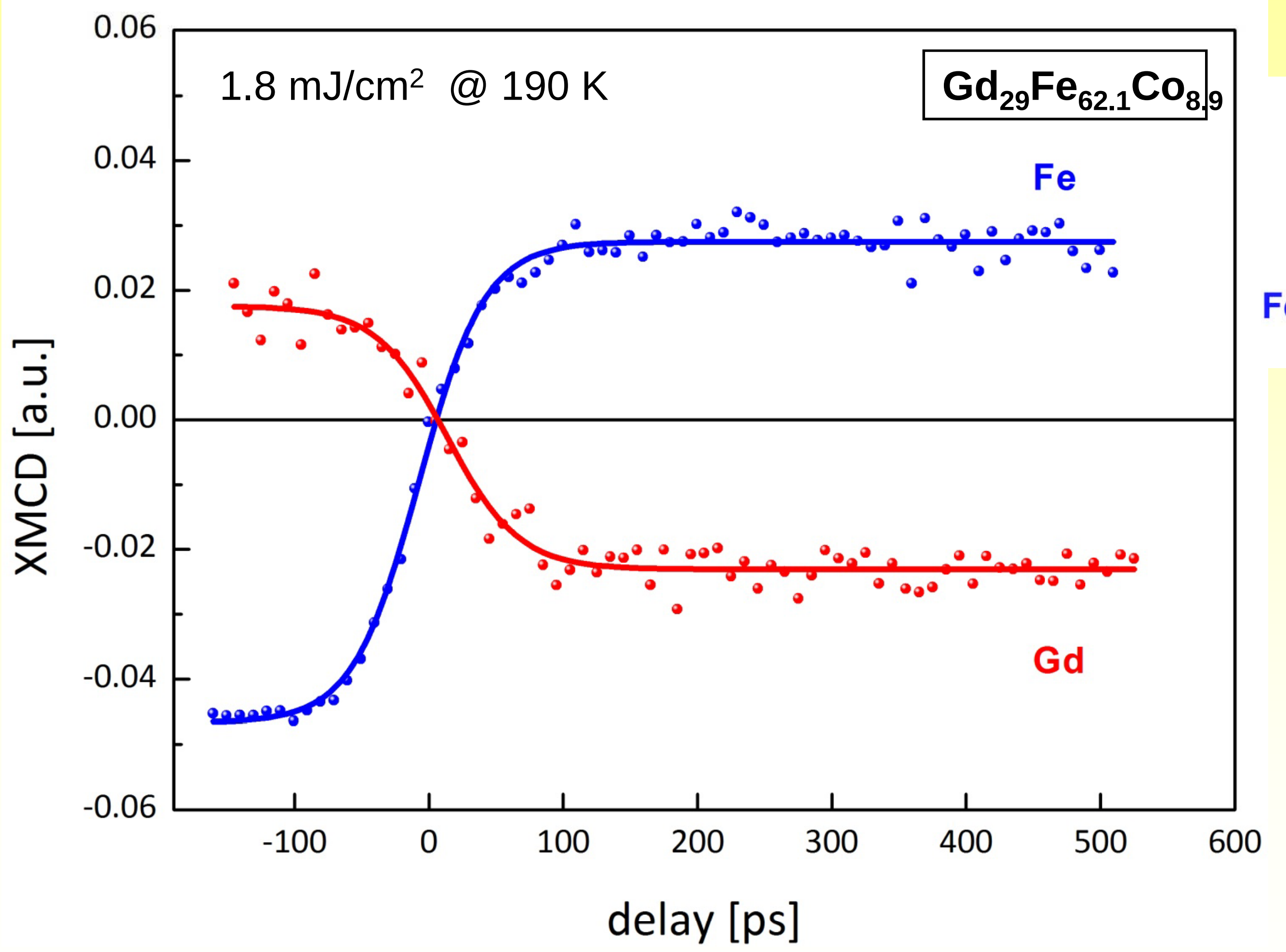


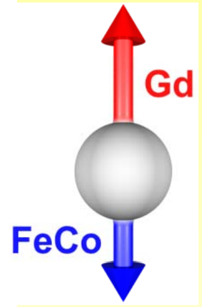
**X-rays**
**400-1400 eV**
**10-50 ps (FWHM)**

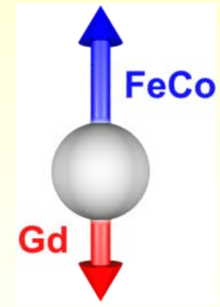
**50 fs laser pulses**
**1.5 eV, 5-50 mJ/cm$^2$**
**3 kHz rep. rate**

# TR-XMCD @BESSY

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



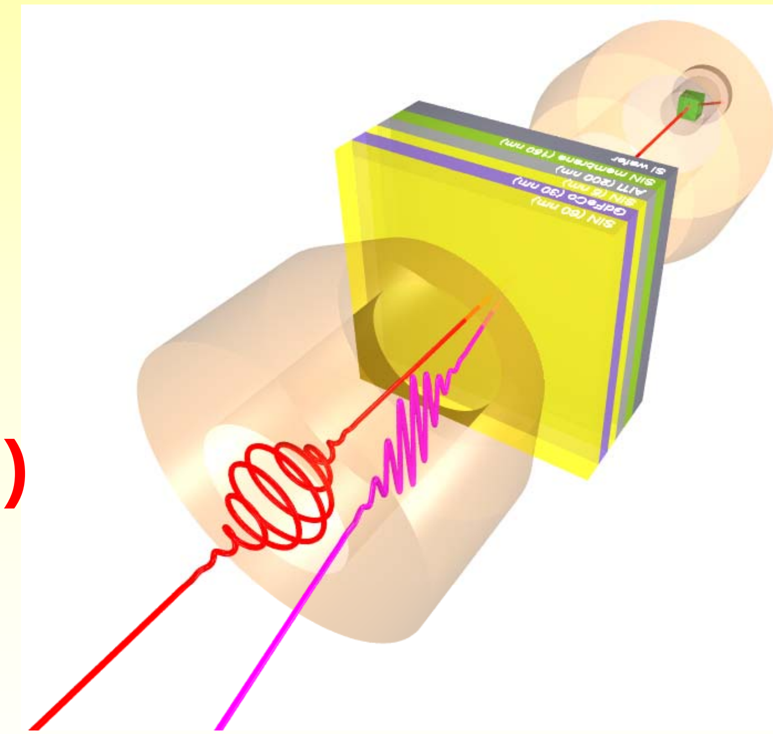



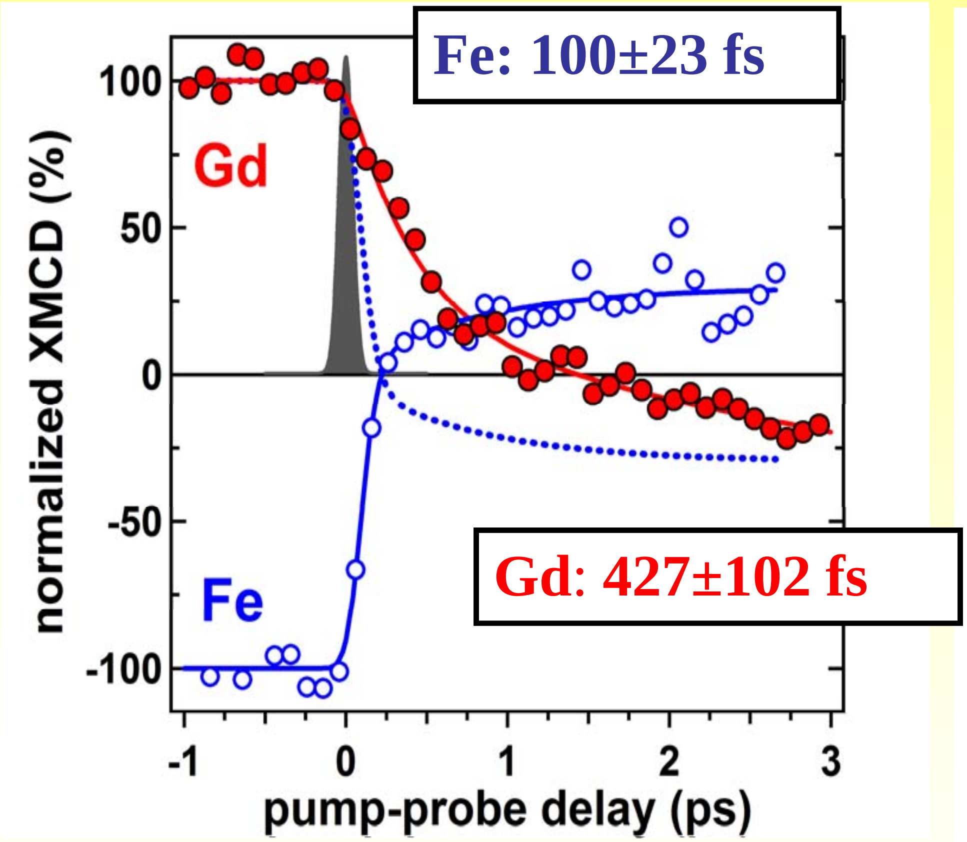
# Can’t we look faster?

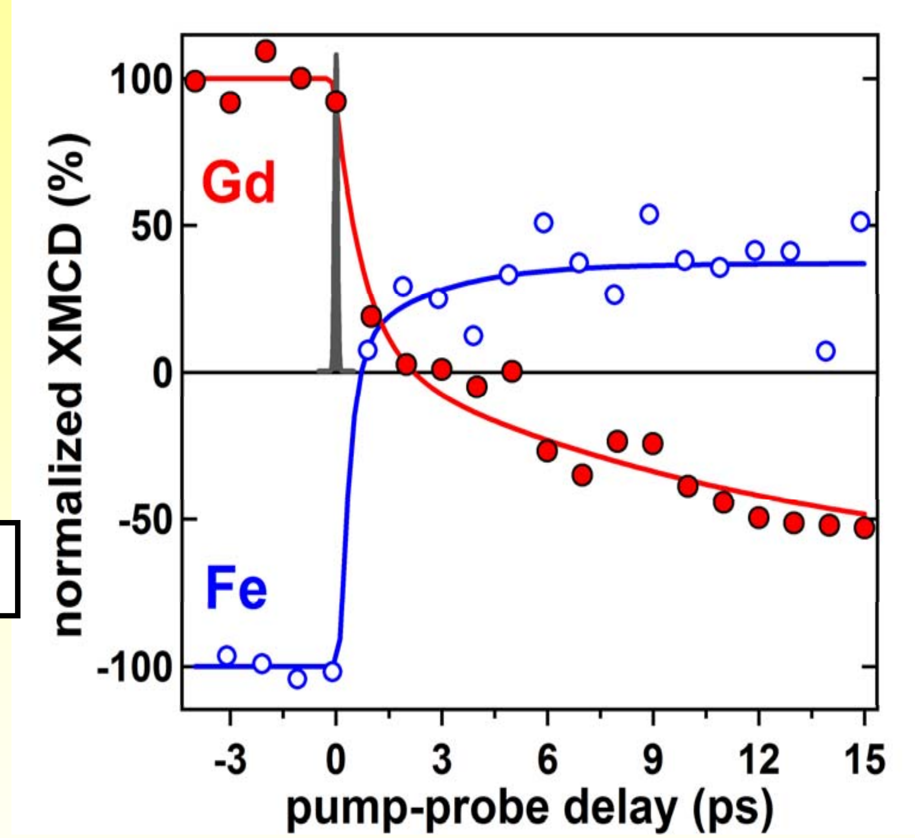
# FEMTO-SLICING!

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



# Element-specific magnetization reversal: 100 fs time resolution

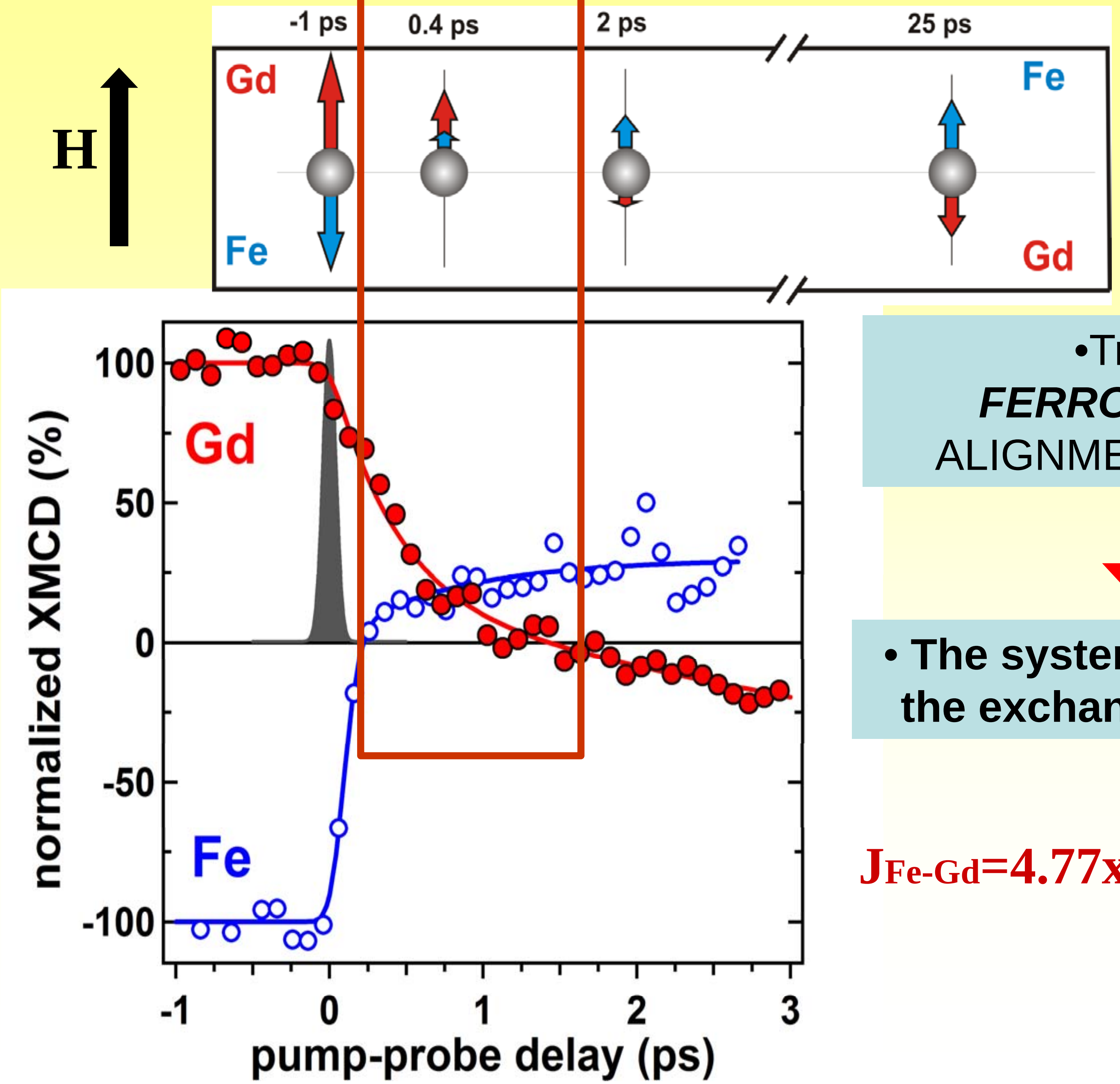



- **Different magnetization switching dynamics at Fe and Gd sites !!!**

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

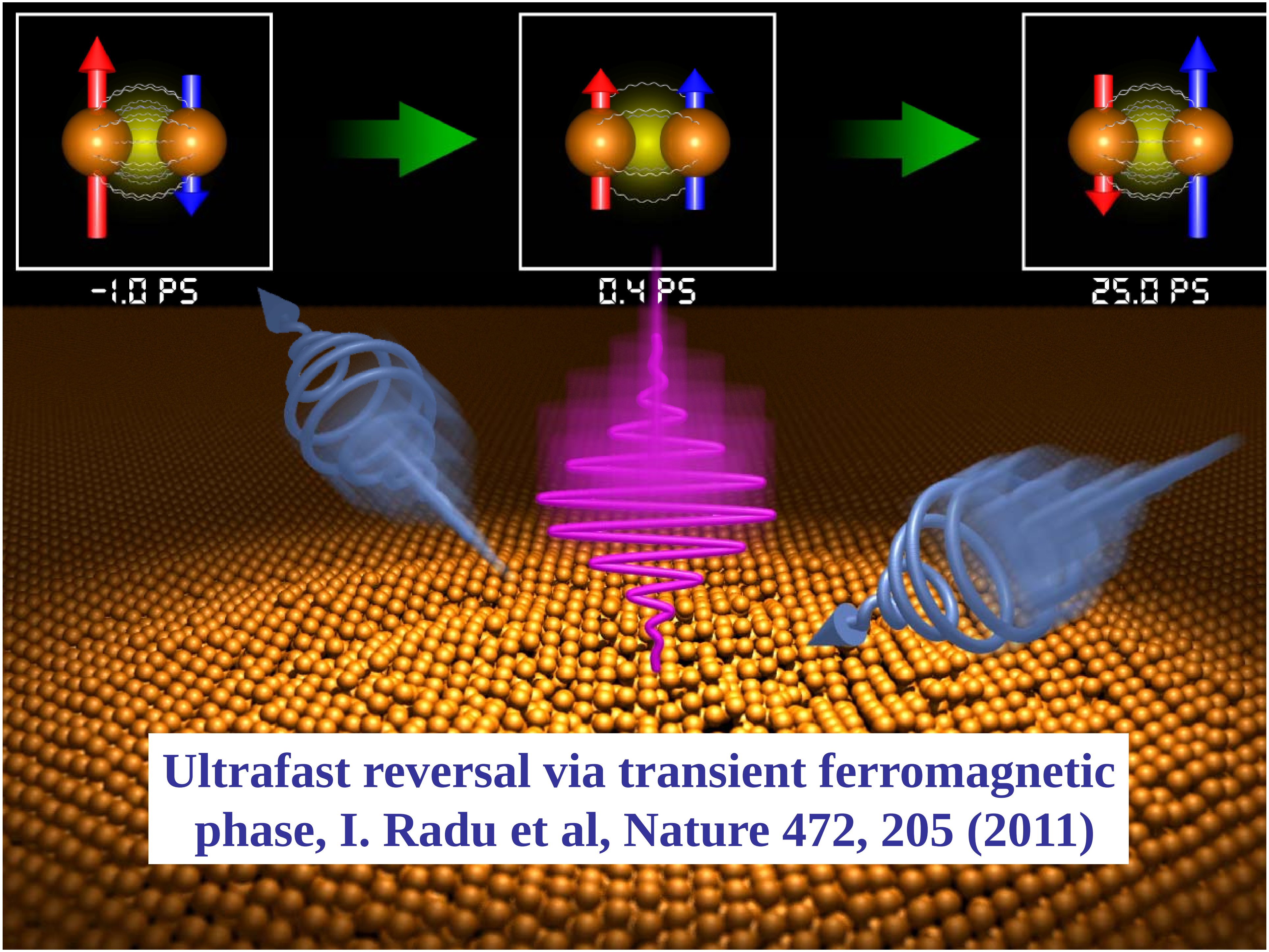
# Switching via a novel non-equilibrium state




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

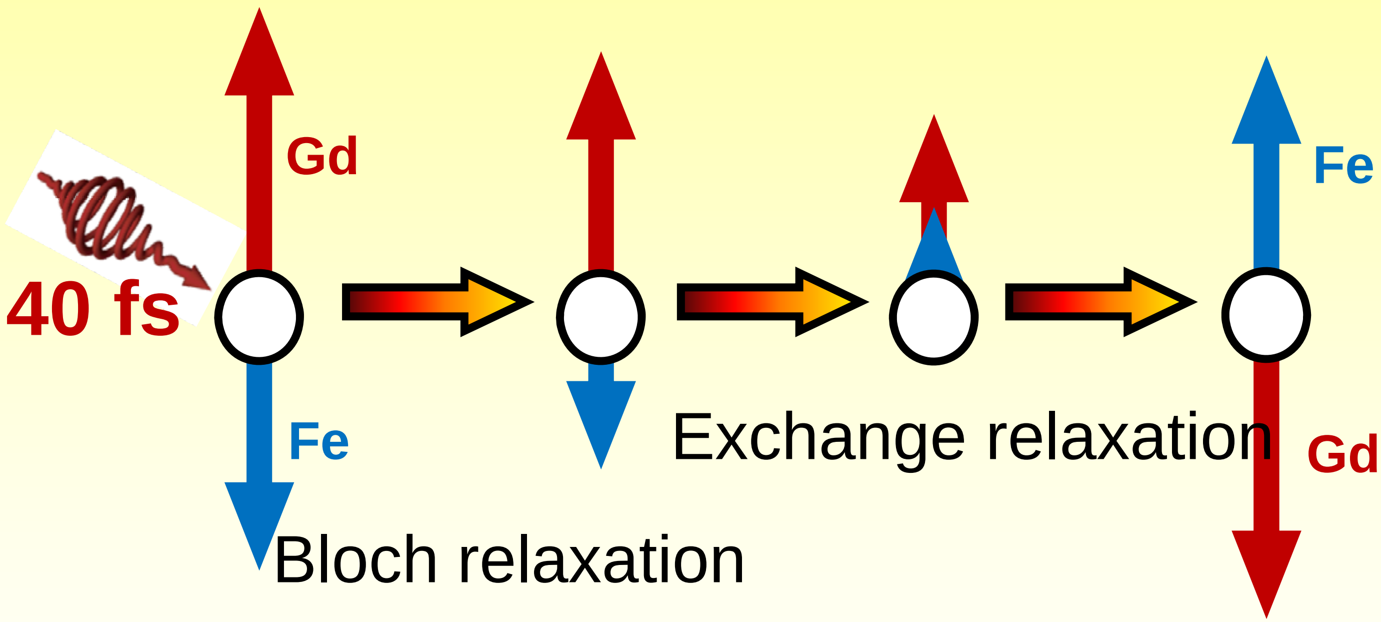
**• The system evolves against the exchange interaction !!!**

**$J_{Fe\text{-}Gd}$=4.77x10$^{-20}$ J ~ 140 fs**




**Ultrafast reversal via transient ferromagnetic phase, I. Radu et al, Nature 472, 205 (2011)**

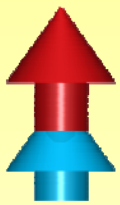
# Ultrafast magnetism in multi-sublattice magnets




**What about multi-sublattice ferromagnets?**

# Prediction: ultrafast demagnetization

## FM coupling

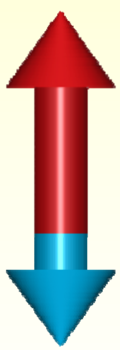


$$dS_1/dt = -\lambda_1 S_1/\chi_1 - \underline{\lambda_e(S_1/\chi_1 - S_2/\chi_2)}$$

positive/negative

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

## AFM coupling



$$dS_1/dt = -\lambda_1 S_1/\chi_1 - \underline{\lambda_e(S_1/\chi_1 + |S_2|/\chi_2)}$$

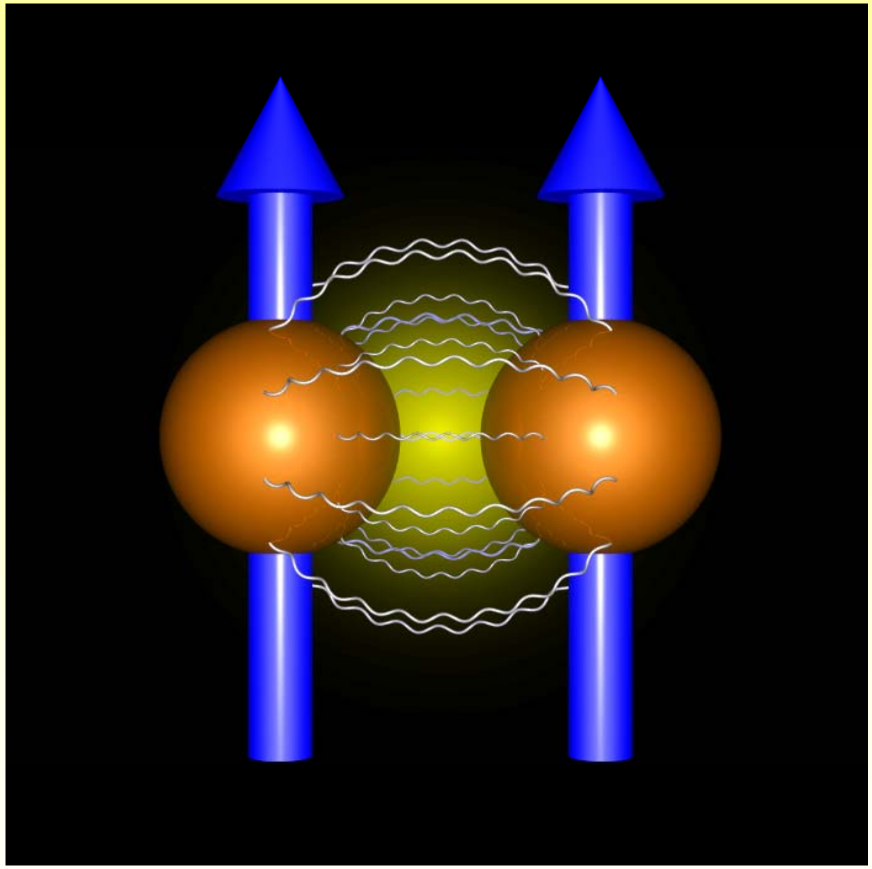
always positive

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

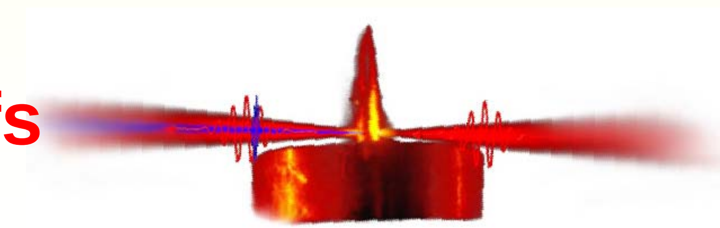
**J.H. Mentink *et al.*, *Phys. Rev. Lett.* (2012)**

# FeNi

$J_{Fe\text{-}Ni}=1.4\text{x}10^{-19}$ J ~ 40 fs

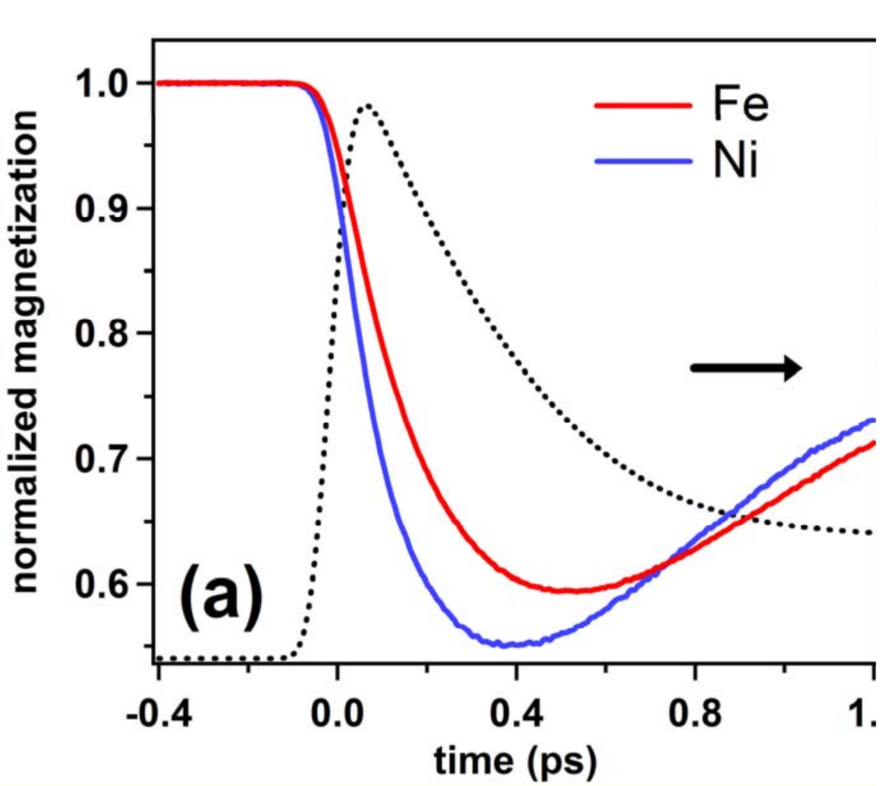


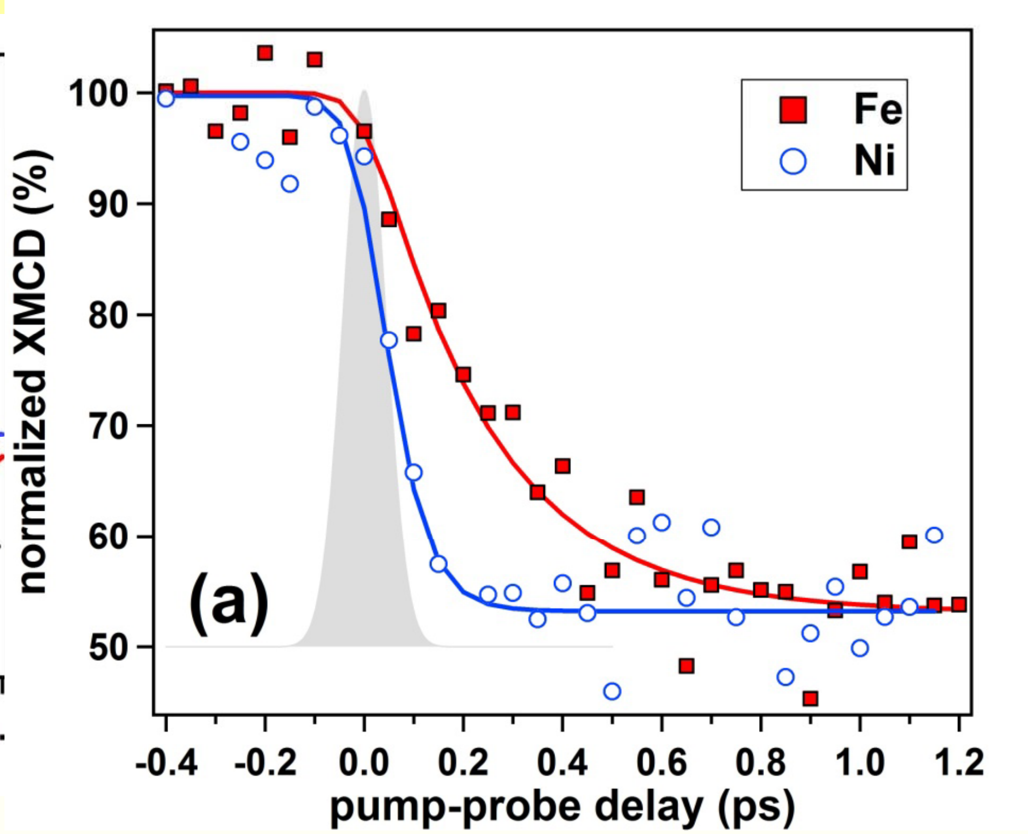
1-2eV,10-100fs



?

# simulations    experiment

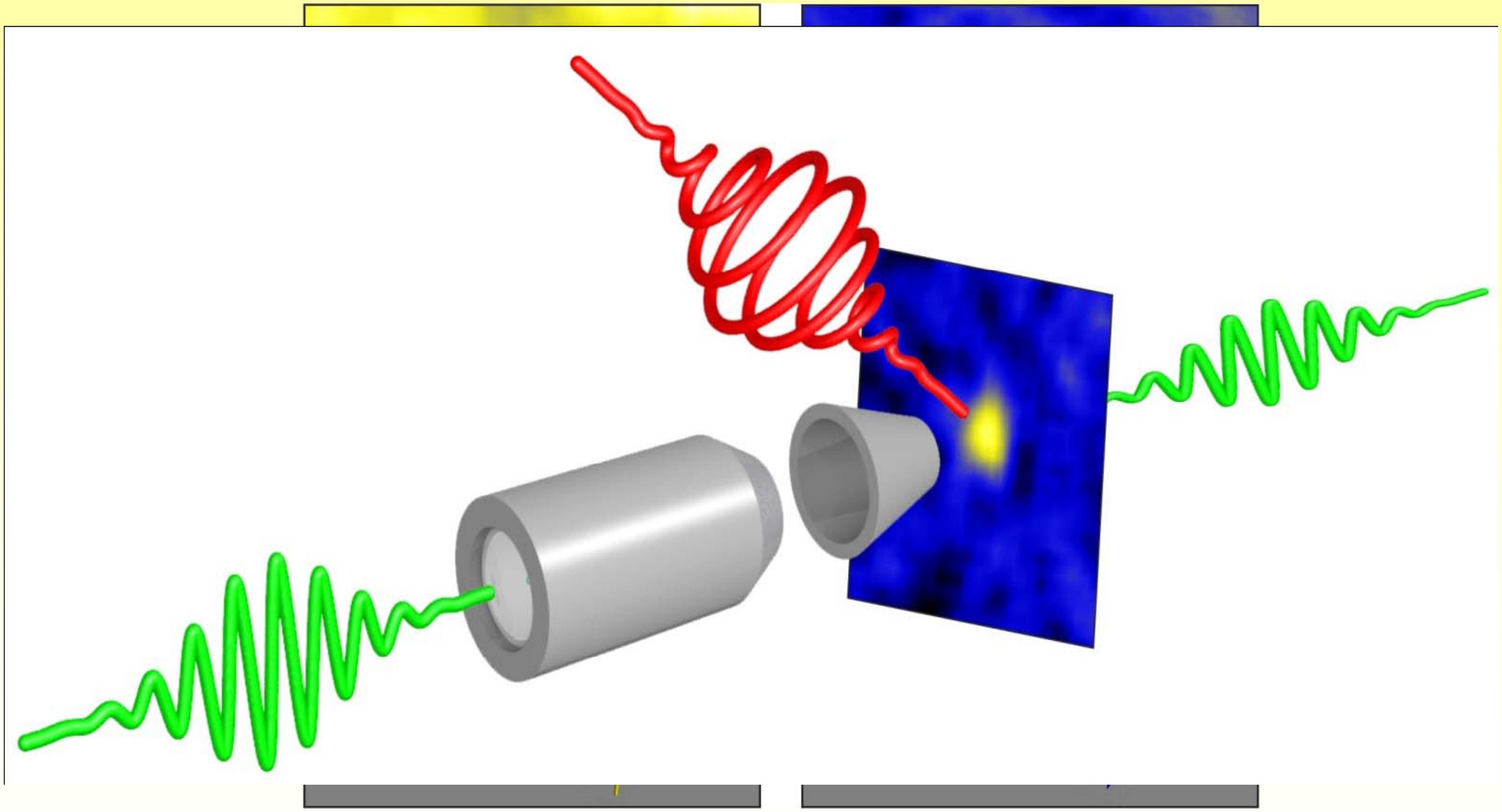



# Fe slower than Ni

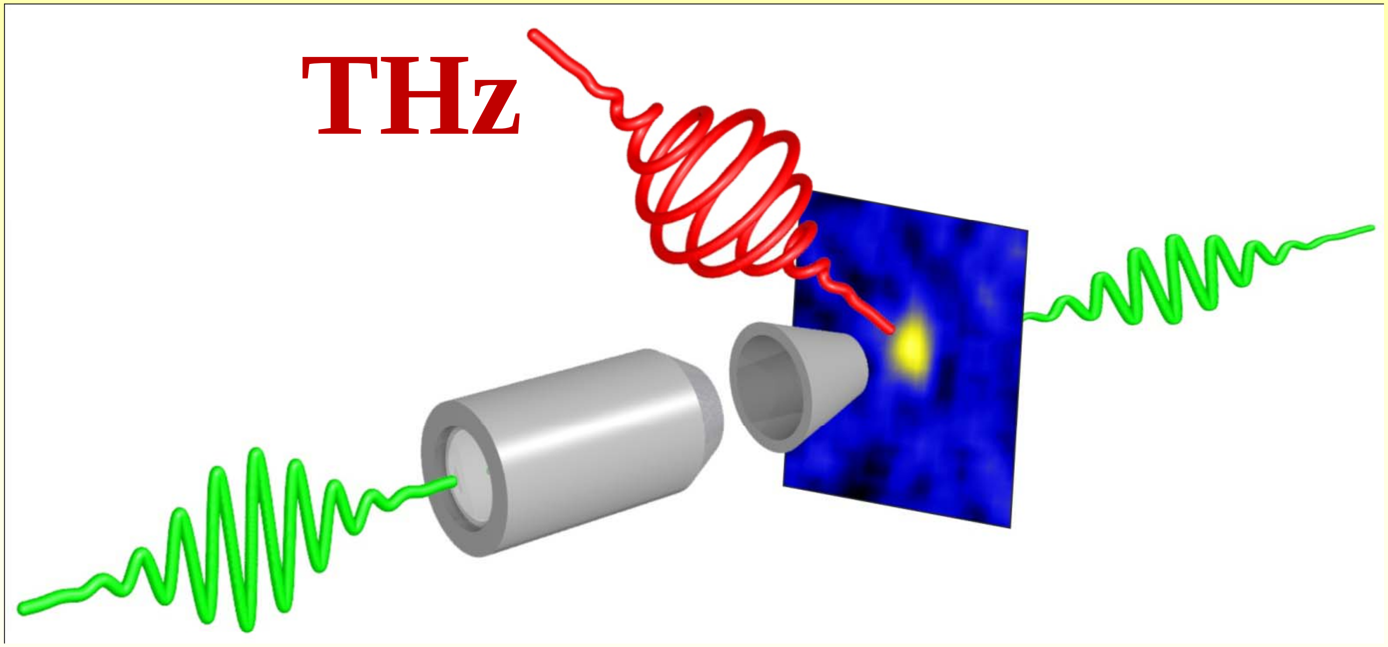
**I. Radu et al, submitted(2012)**

# Perspectives



# Skyrmions?

# Perspectives

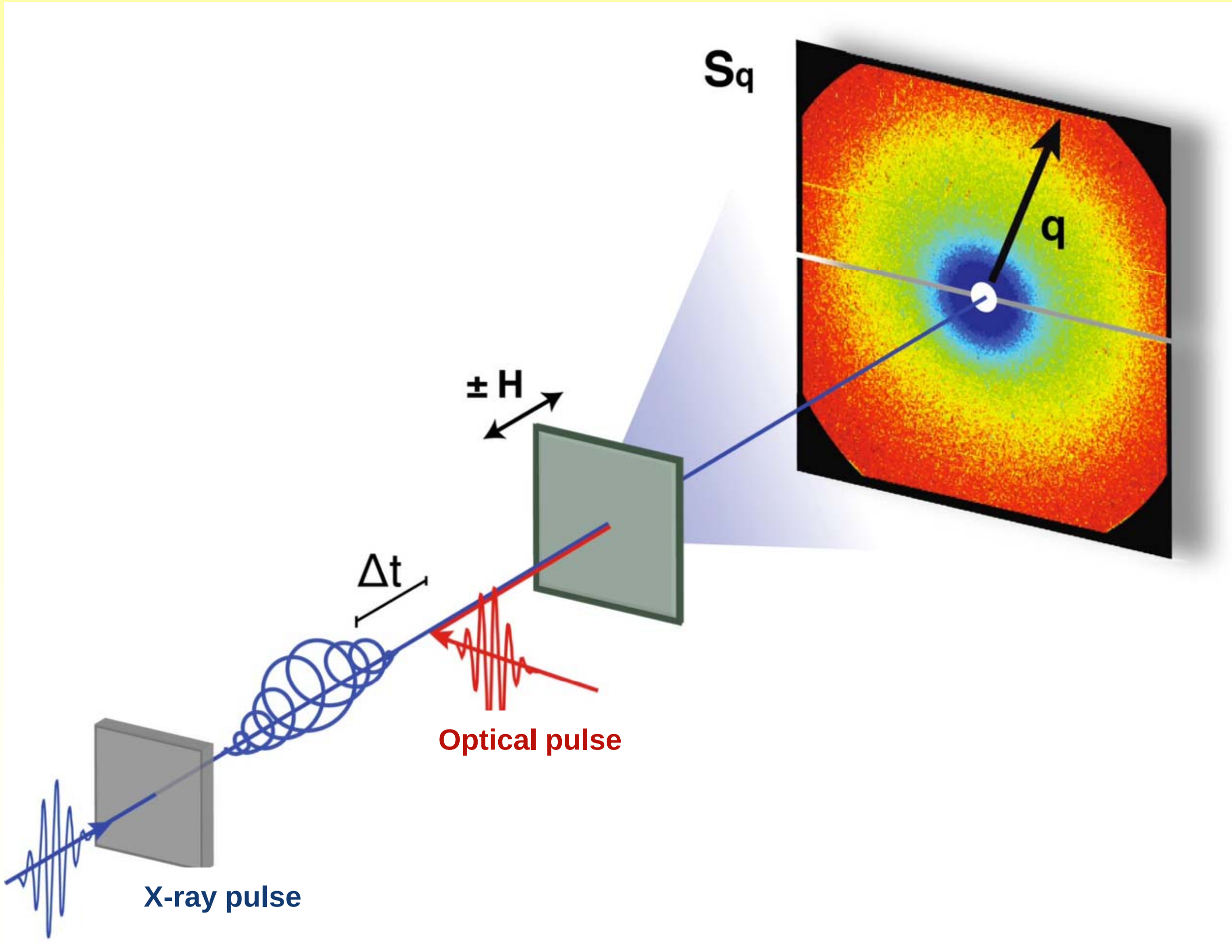


# Perspectives

$J = J(t)$ → $H(t)$ →

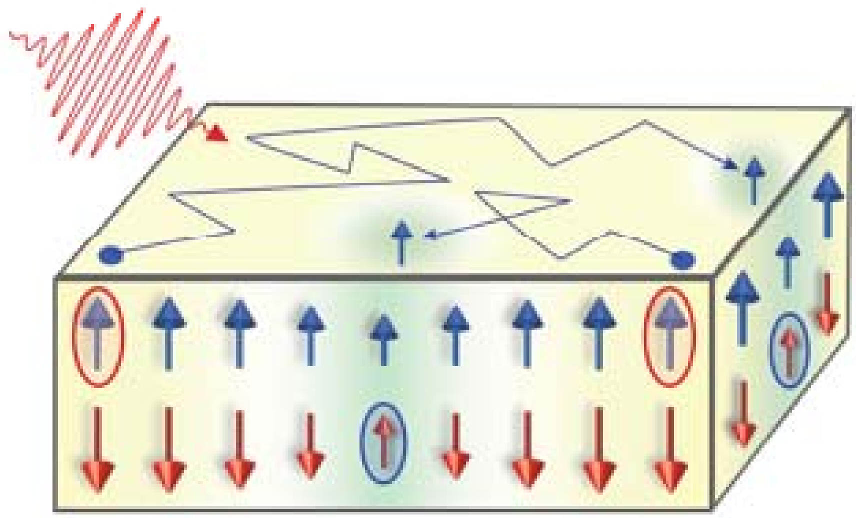
**Xrays+XMCD+nm+fs!**

# First LCLS data



# Supersonic transport of angular momentum!

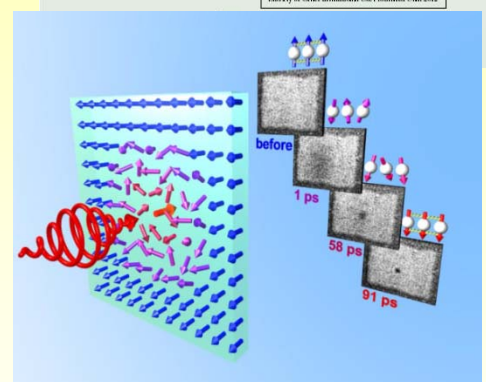
**(~nm/fs)**



# *To conclude:*

# *With light*

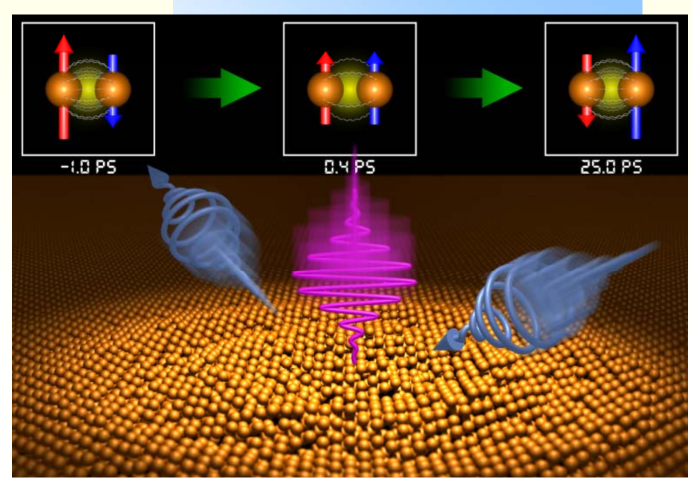
- **Coherent optical control of magnetism !**
- **All-optical ultrafast magnetic recording**
- **Novel (linear) ultrafast reversal path!**
- **Novel transient ferromagnetic state**


## Future challenges

- **Femto magnetism!**
- **Combine chemical, magnetic**

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



# *with many thanks to:*


**IMM**
A.V. Kimel
A.Kirilyuk
F. Hansteen
D.Stanciu
A.v.Etteger
K.Vahaplar
A. Kalashnikova
J. Mentink
**University of York**
R. Chantrell
T.A. Ostler
J.Barker
R.Evans

**Ioffe Institute**
Prof. R.V. Pisarev
P. Usachev
**Nihon University**
Prof. A. Itoh
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**

# *Spectroscopy of Solids and Interfaces*