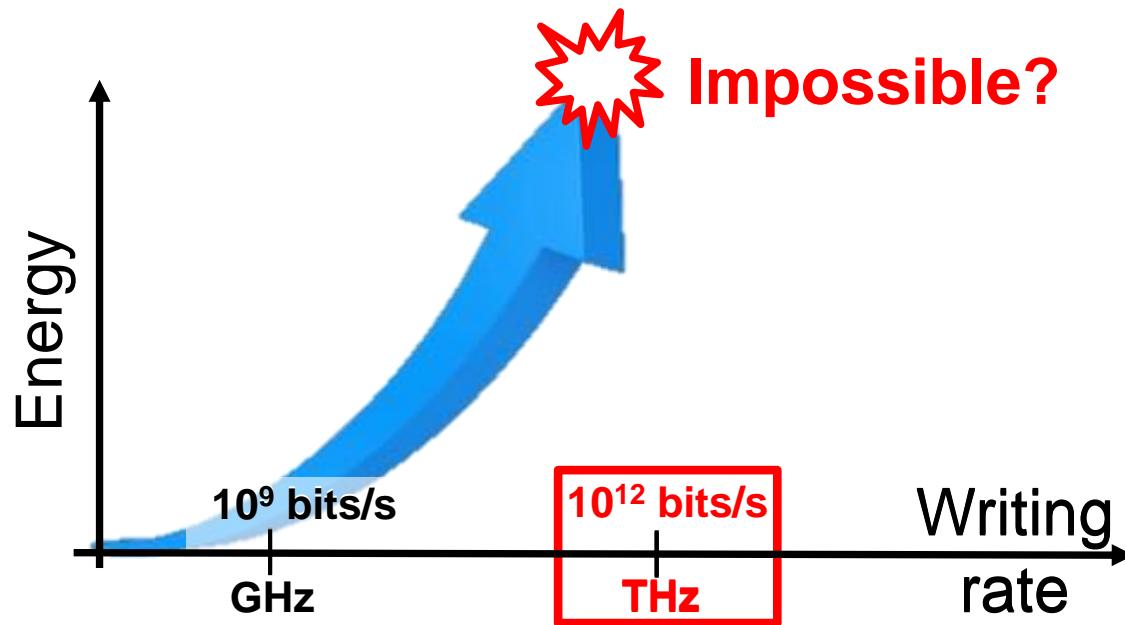


Magnetization reversal: *from fundamental principles* *to fundamental limits*

Alexey V. Kimel

*Ultrafast Spectroscopy of Correlated Materials,
Radboud University, Nijmegen, The Netherlands*

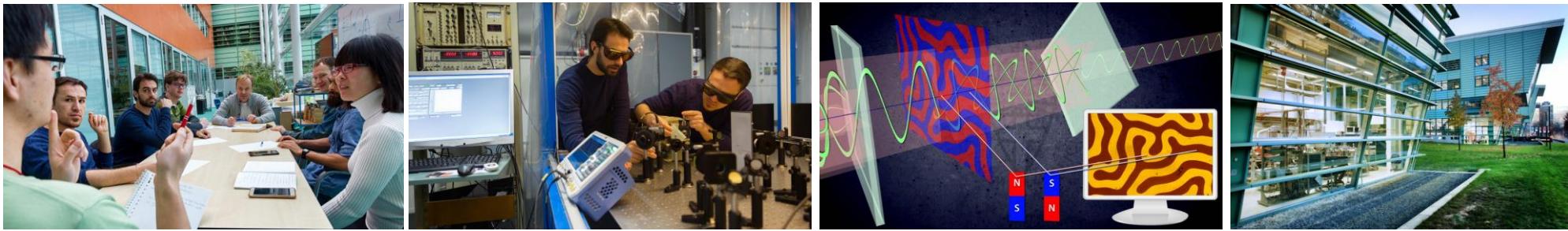
Fundamental Dilemma: fast or energy efficient?



Challenge: fast and energy efficient

Mission:

To discover new fundamental principles
for the fastest possible writing of magnetic bits
accompanied by minimal loss of energy



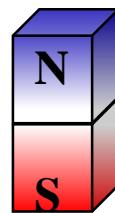
Magnetic recording: experiment, theory, practice

Practice



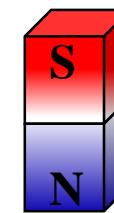
ns
 $0.1 \text{ nJ} = 10^{-10} \text{ J}$

Experiment

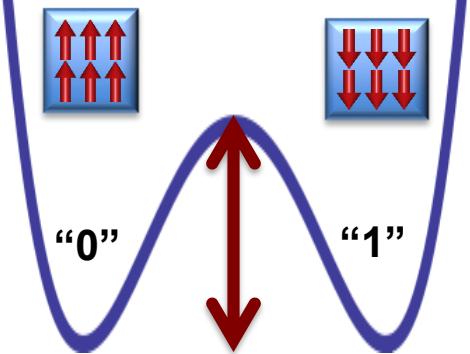


*physics?
ultrafast?*

*high density?
non-dissipative?*



Theory

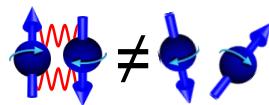


$60 k_B T \sim 10^{-19} \text{ J}$

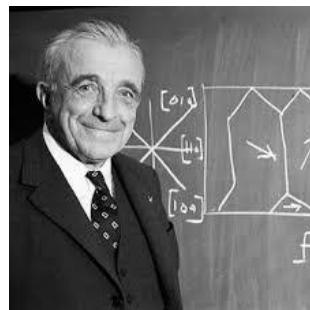
Magnetism – the strongest quantum mechanical phenomenon



$$S_z = \pm \hbar/2$$



$$E_{ex} = -JS_i S_j$$



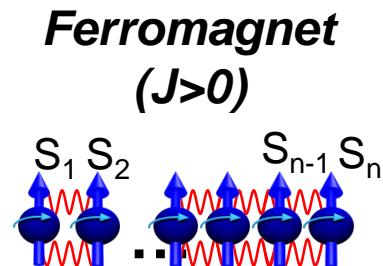
L. Néel

Spin
(1925)

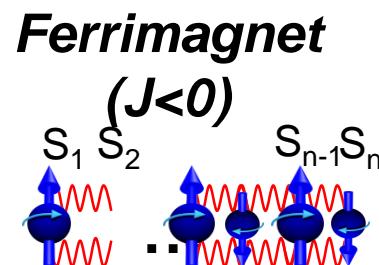
Exchange interaction
(1926)

Antiferromagnetism
(1930)

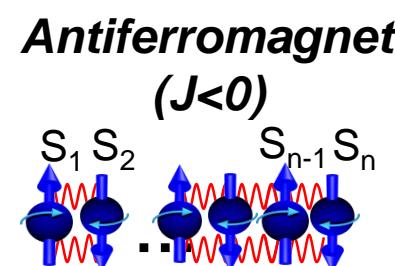
Ferrimagnetism
(1948)



$$\mathbf{M} = -\gamma \frac{\sum S_i}{V} \neq 0$$



$$\mathbf{M} \neq 0$$



$$\mathbf{M} = 0$$

Macrospin approximation: *intuitive (classical) view of quantum (counter-intuitive) phenomenon*

$$dU = dW + dQ$$

$$dW = \mu_0 \mathbf{H} d\mathbf{M}$$

$$\mu_0 \mathbf{H}_{eff} = \left(\frac{\partial U}{\partial \mathbf{M}} \right)_{\sigma}$$

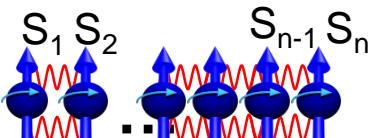
U - internal energy
 W - work
 Q - heat
 σ - entropy

$$\mathbf{M} = -\gamma \frac{\sum S_i}{V} \neq 0$$

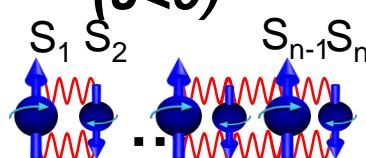
$$\mathbf{M} = -\gamma \frac{\sum S_i}{V} \neq 0$$

$$\mathbf{L} = -\gamma \frac{\sum (S^{\uparrow} - S^{\downarrow})}{V}$$

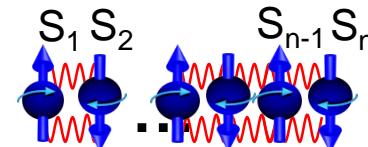
Ferromagnet
 $(J>0)$



Ferrimagnet
 $(J<0)$



Antiferromagnet
 $(J<0)$



Macrospin approximation: *intuitive (classical) view of quantum (counter-intuitive) phenomenon*

$$dU = dW + dQ$$

$$dW = \mu_0 \mathbf{H} d\mathbf{M}$$

$$\left. \begin{aligned} \mu_0 \mathbf{H}_{eff} &= - \left(\frac{\partial U}{\partial \mathbf{M}} \right)_{\sigma} \end{aligned} \right\}$$



P. Curie (1894):

"the symmetries of the causes are to be found in the effects".

Cause	Effect	Energy (U)
\mathbf{F} mechanical force	$d\mathbf{x}$ displacement	Fdx
\mathbf{E} electric field	$d\mathbf{P}$ polarization	$Ed\mathbf{P}$
\mathbf{H} magnetic field	$d\mathbf{M}$ magnetization	$Hd\mathbf{M}$



$$\mathbf{F} = - \frac{\partial U}{\partial \mathbf{x}}$$

Macrospin approximation: *intuitive (classical) view of quantum (counter-intuitive) phenomenon*

$$\left. \begin{array}{l} dU = dW + dQ \\ dW = \mu_0 \mathbf{H} d\mathbf{M} \end{array} \right\} \quad \mu_0 \mathbf{H}_{eff} = \left(\frac{\partial U}{\partial \mathbf{M}} \right)_\sigma$$

U - internal energy
 W - work
 Q - heat
 σ - entropy
 \mathbf{T} - torque

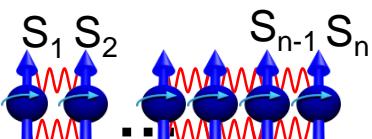
$$\frac{d\mathbf{S}}{dt} = \mathbf{T} \quad \frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{H}_{eff} - \frac{\lambda}{M^2} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{eff})$$

Landau-Lifshitz
equation
(1935)

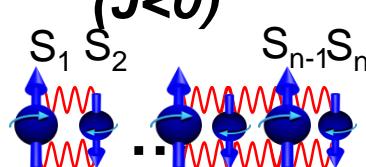
$$\mathbf{M} = -\gamma \frac{\sum S_i}{V} \neq 0$$

$$\mathbf{M} = -\gamma \frac{\sum S_i}{V} \neq 0$$

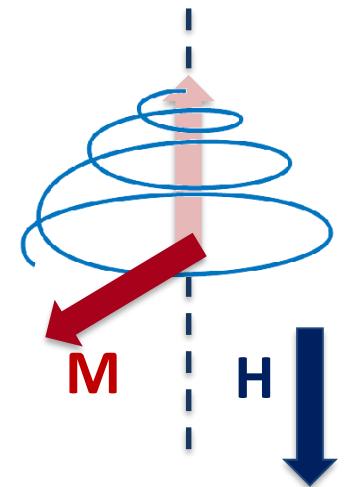
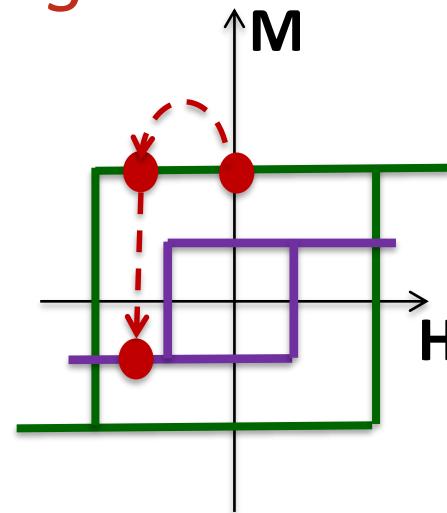
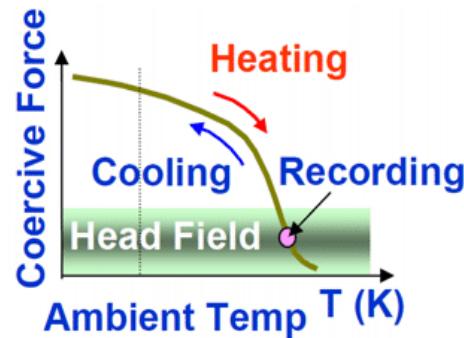
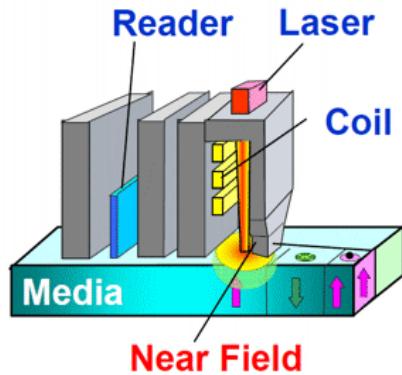
Ferromagnet
 $(J > 0)$



Ferrimagnet
 $(J < 0)$



State-of-the-art in data storage: *heat assisted magnetic recording*

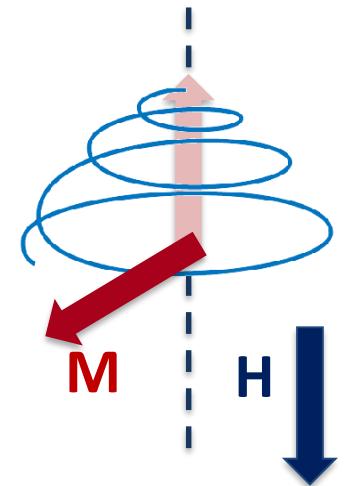
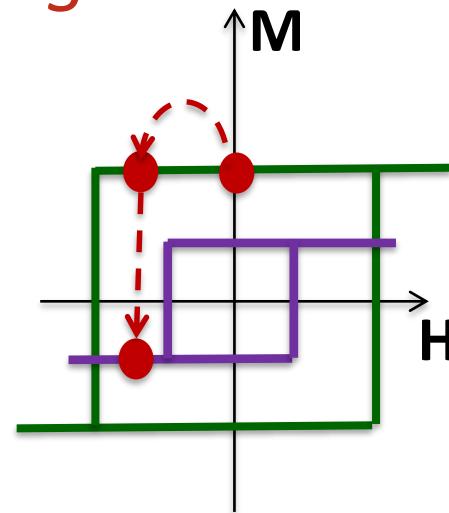
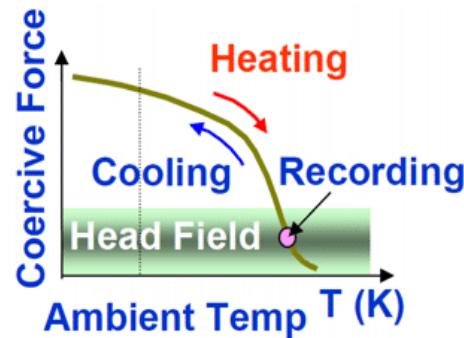
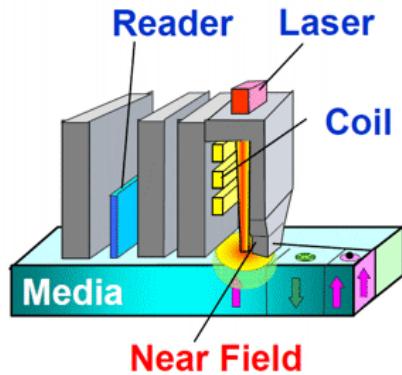


$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{H}_{eff} - \frac{\lambda}{M^2} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{eff})$$

Magnetization reversal

- with a magnetic field in a “wrong” direction;
- without any magnetic field and solely with heat;
- with no heat, no field .

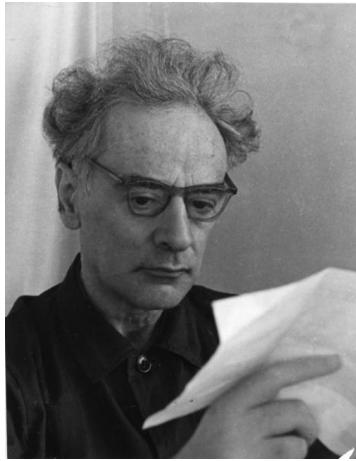
State-of-the-art in data storage: *heat assisted magnetic recording*



$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{H}_{eff} - \frac{\lambda}{M^2} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{eff})$$

Magnetization reversal

- with a magnetic field in a “wrong” direction;
- without any magnetic field and solely with heat;
- with no heat, no field.



L. D. Landau
(1908-1968)

**“Most important part of doing physics
is the knowledge of approximations.”**

Macrospin approximation: *intuitive (classical) view of quantum (counter-intuitive) phenomenon*

Nonequilibrium state



Macrospin
(classical)

$$-\mu_0 H_{eff}^{(j)} = \frac{\partial E_{ex}}{\partial S_i} \sim 1 - 1000 \text{ T}$$

$$\frac{d\mathbf{M}}{dt} = -\gamma \mathbf{M} \times \mathbf{H}_{eff} \quad \frac{\gamma}{2\pi} = 28 \text{ GHz/T}$$

$\tau_{ex} = 30 \text{ fs} - 30 \text{ ps}$

$t \sim \tau_{ex}$

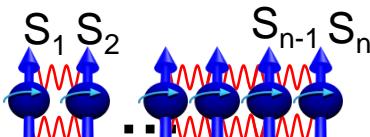
$t \gg \tau_{ex}$

$$\mathbf{M} = -\gamma \frac{\sum S_i}{V} \neq 0$$

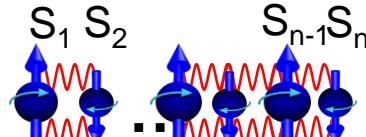
$$\mathbf{M} = -\gamma \frac{\sum S_i}{V} \neq 0$$

$$\mathbf{L} = -\gamma \frac{\sum (S_{2i-1} - S_{2i})}{V}$$

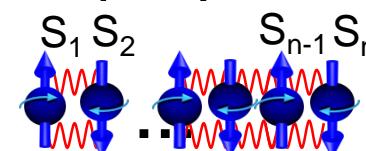
Ferromagnet
($J > 0$)



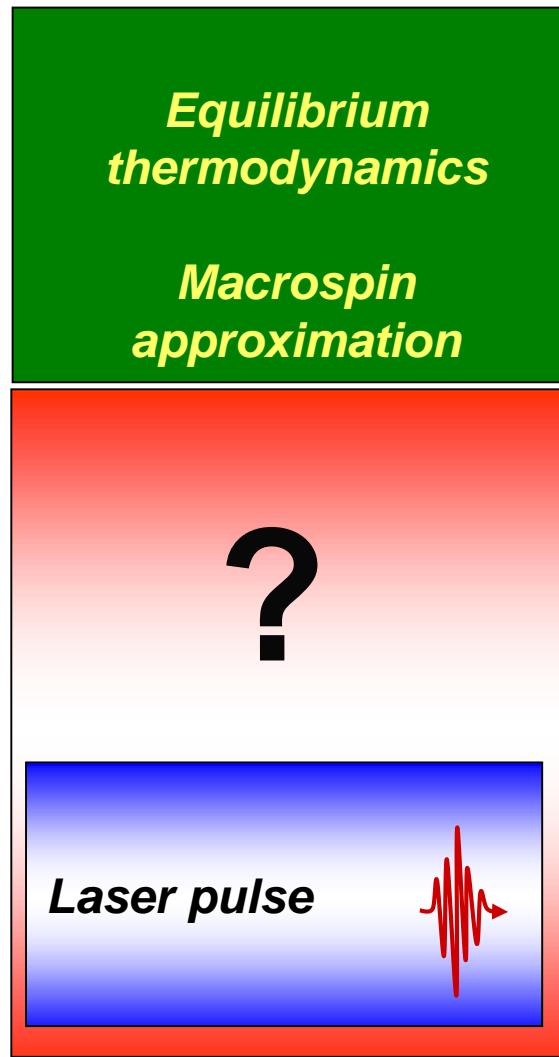
Ferrimagnet
($J < 0$)



Antiferromagnet
($J < 0$)

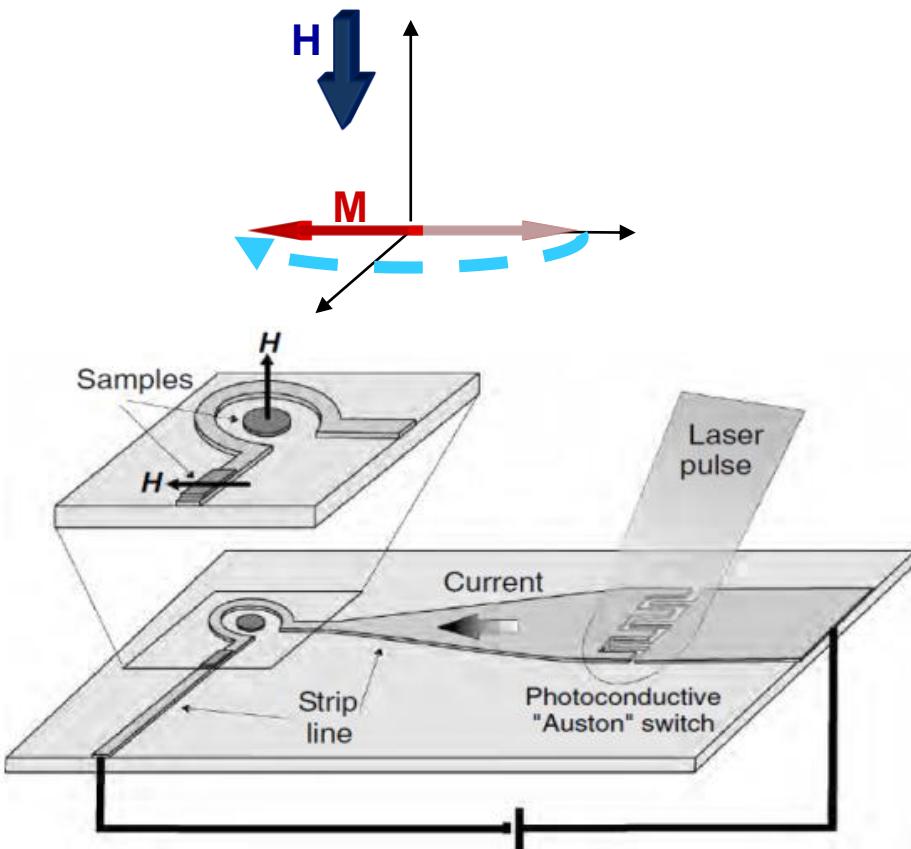


Ultrafast magnetism: *terra incognita* of modern science



Curie's principle
fails!

Precessional switching

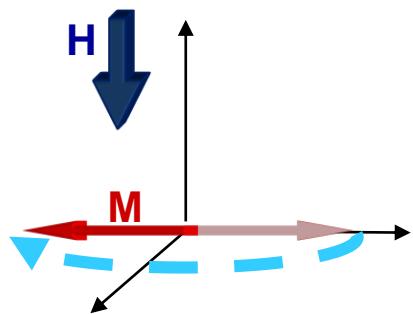


$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{H}_{eff} - \frac{\lambda}{M^2} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{eff})$$

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

The shortest time achieved is 100 ps!

Precessional switching

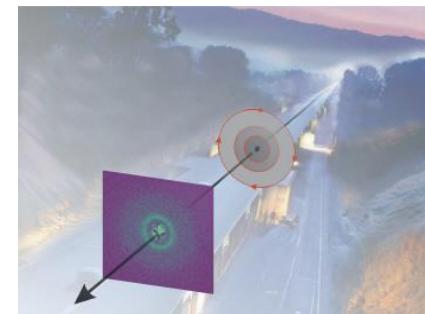
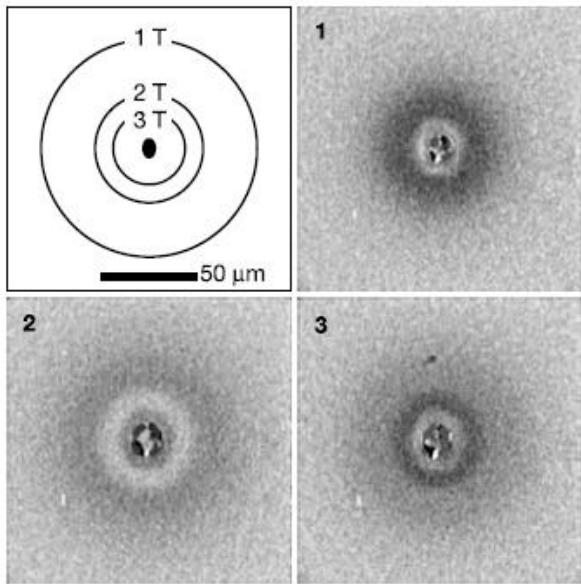


$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{H}_{eff} - \frac{\lambda}{M^2} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{eff})$$

letters to nature

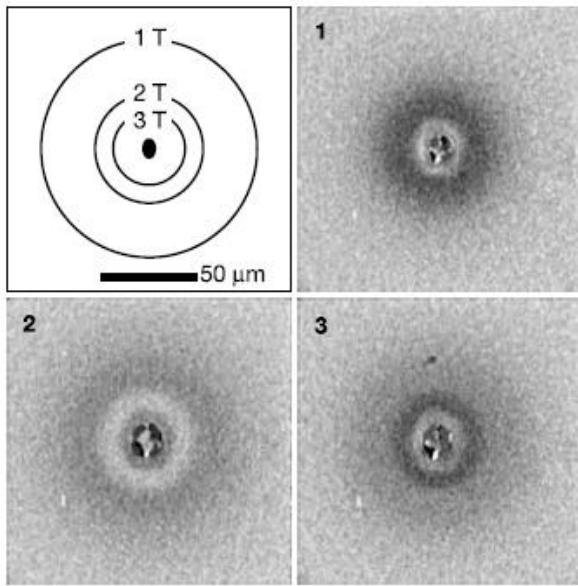
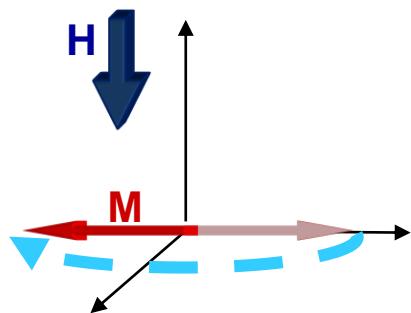
The ultimate speed of magnetic switching in granular recording media

I. Tudosa¹, C. Stamm¹, A. B. Kashuba², F. King³, H. C. Siegmann¹, J. Stöhr¹, G. Ju⁴, B. Lu⁴ & D. Weller⁴



2.3 ps,
several T pulses

Precessional switching



$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{H}_{eff} - \frac{\lambda}{M^2} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{eff})$$

letters to nature

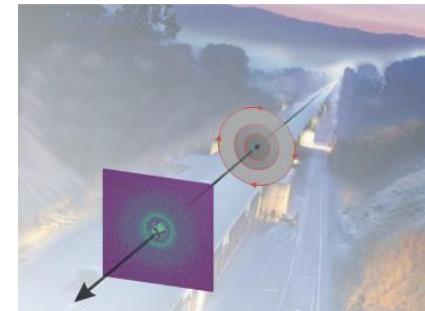
The ultimate speed of magnetic switching in granular recording media

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Applied physics

Speed limit ahead

C. H. Back and D. Pescia



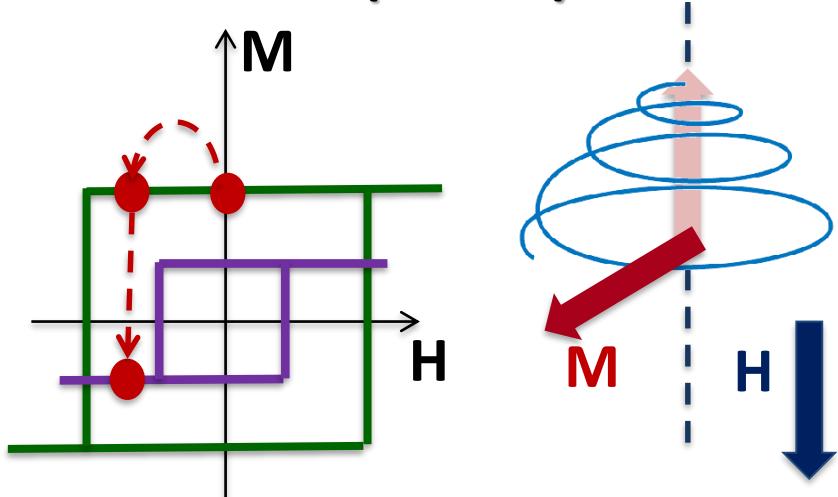
2.3 ps,

several T pulses

“No matter how short and strong the magnetic-field pulse, magnetic recording cannot be made ever faster.”

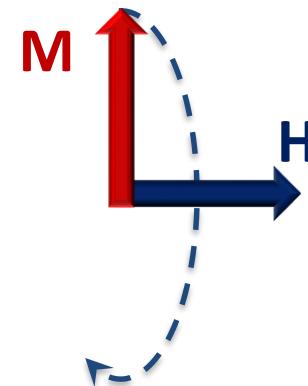
New route for heat-assisted magnetic recording

Heat Assisted Magnetic Recording (HAMR)



best spatial resolution

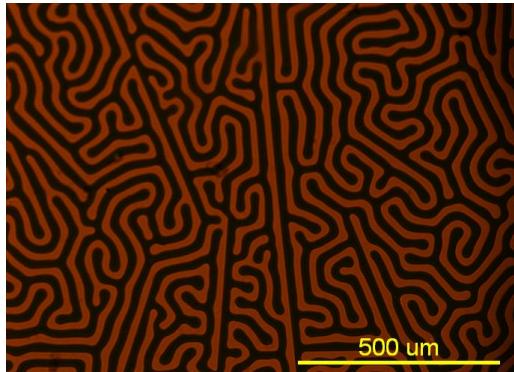
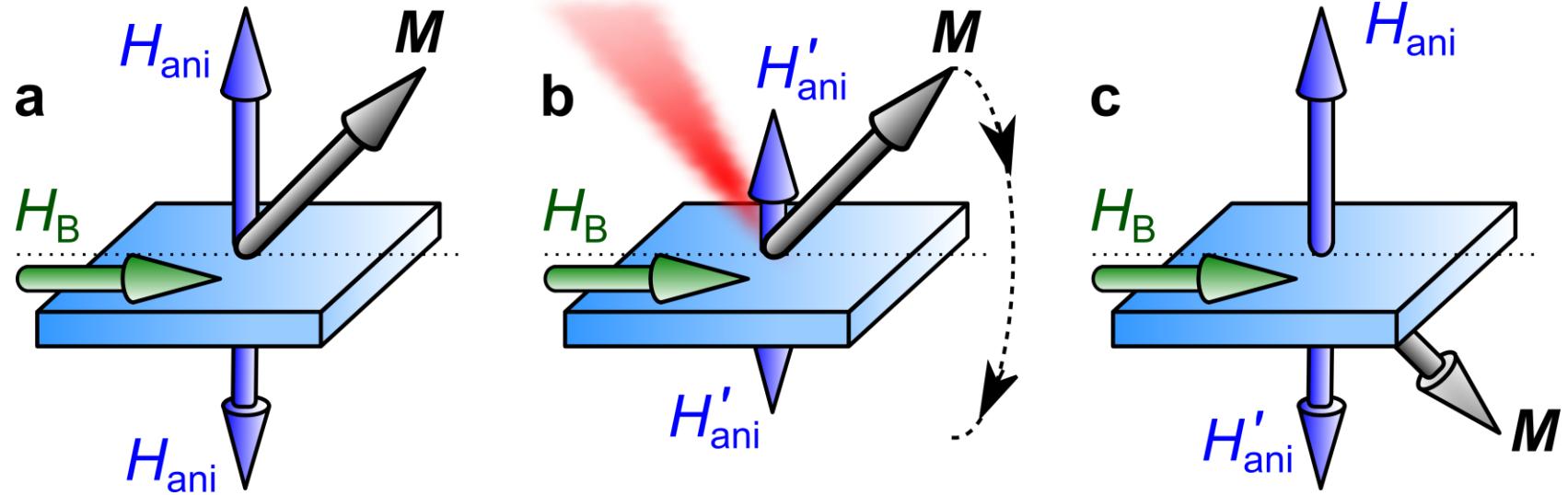
Precessional Switching



fastest route

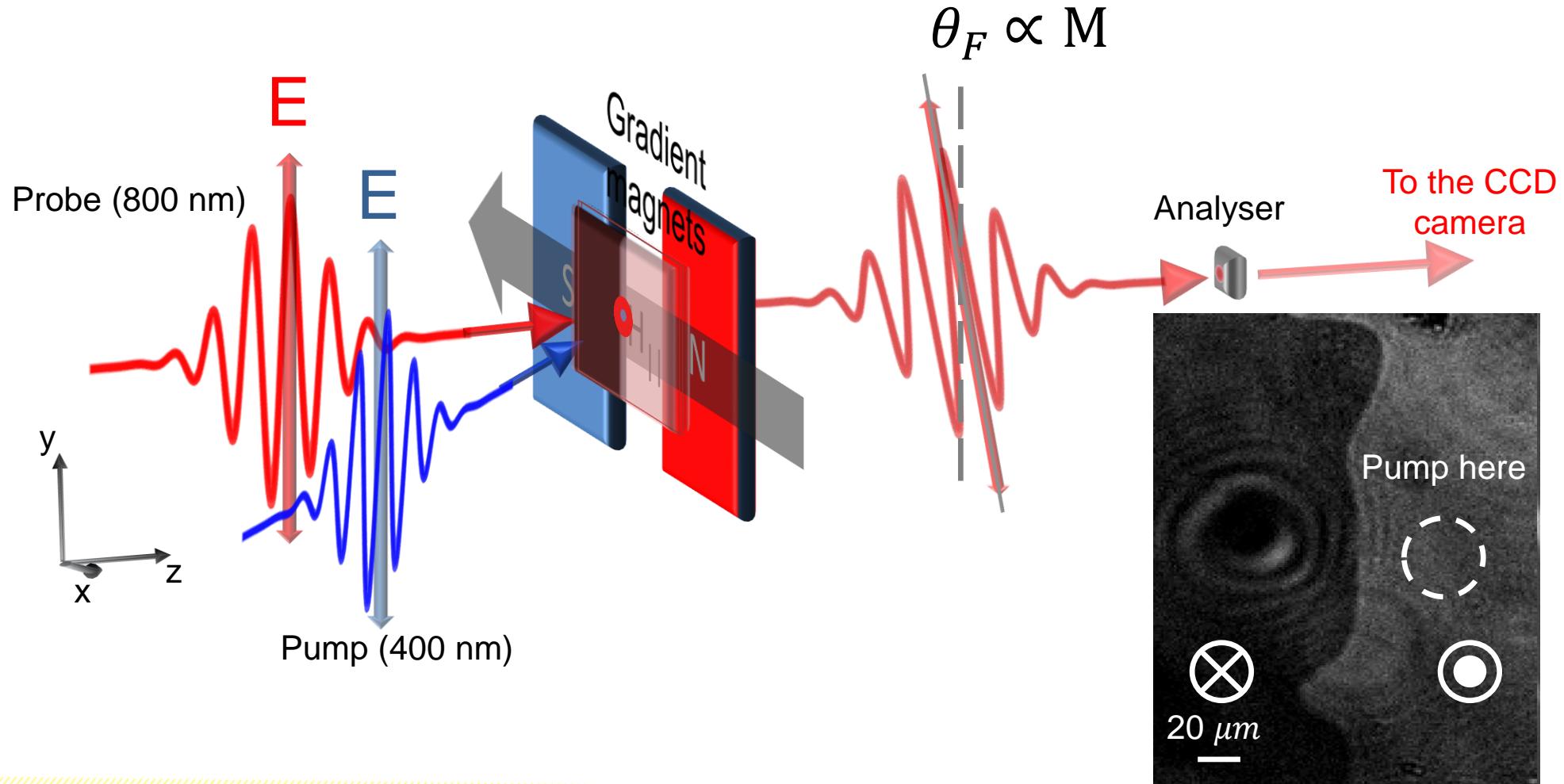
Combine?

Heat-assisted magnetic switching



Bi-substituted Yttrium Iron Garnet (Bi:YIG)
Ferrimagnet with strong out-of-plane anisotropy
Low Gilbert damping

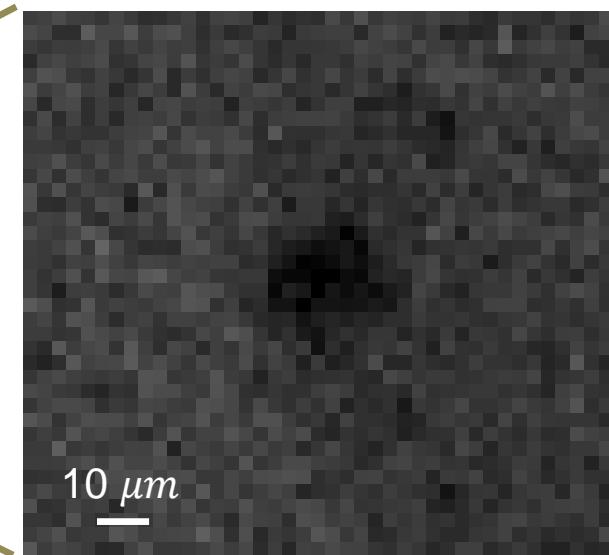
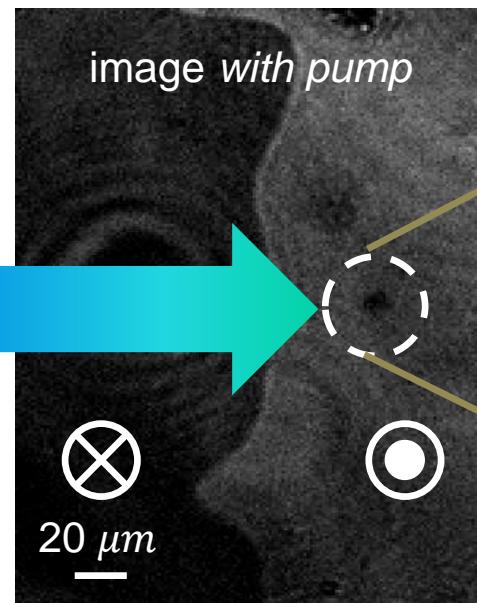
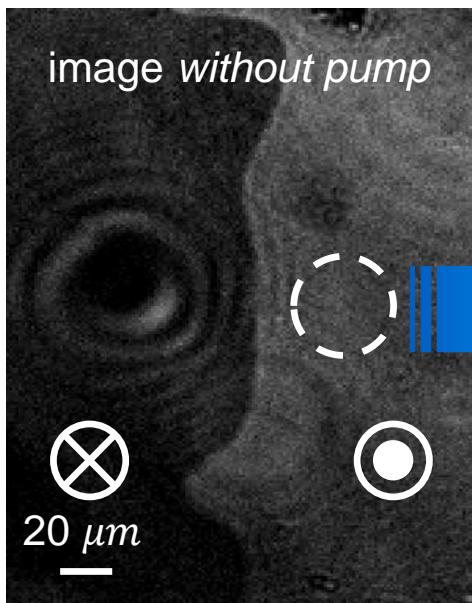
Experimental setup



change perspective

Heat-assisted magnetic switching

$H_x = 5.47 \text{ kOe}$

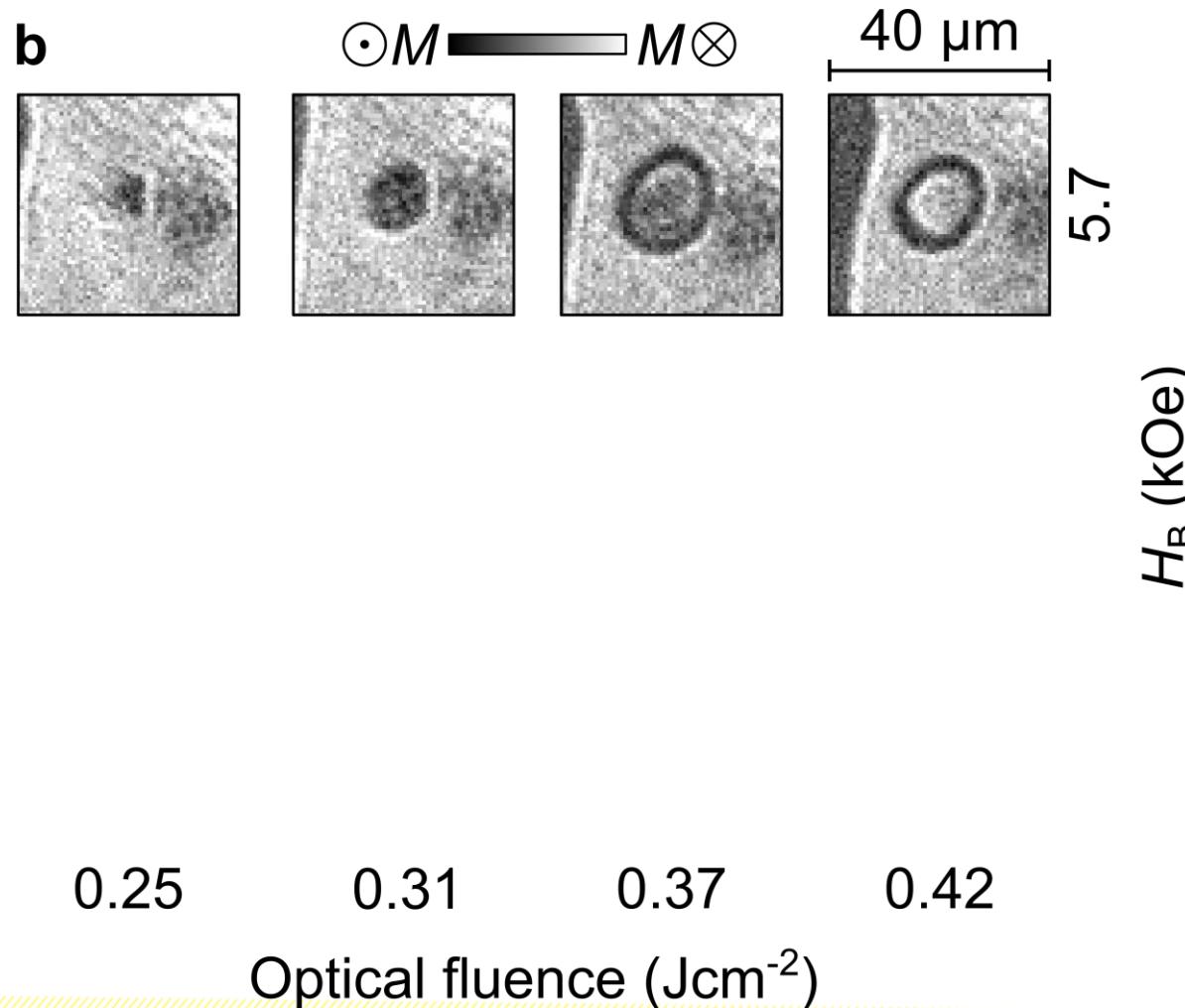


$\tau = 3.45 \text{ ns}$

change perspective

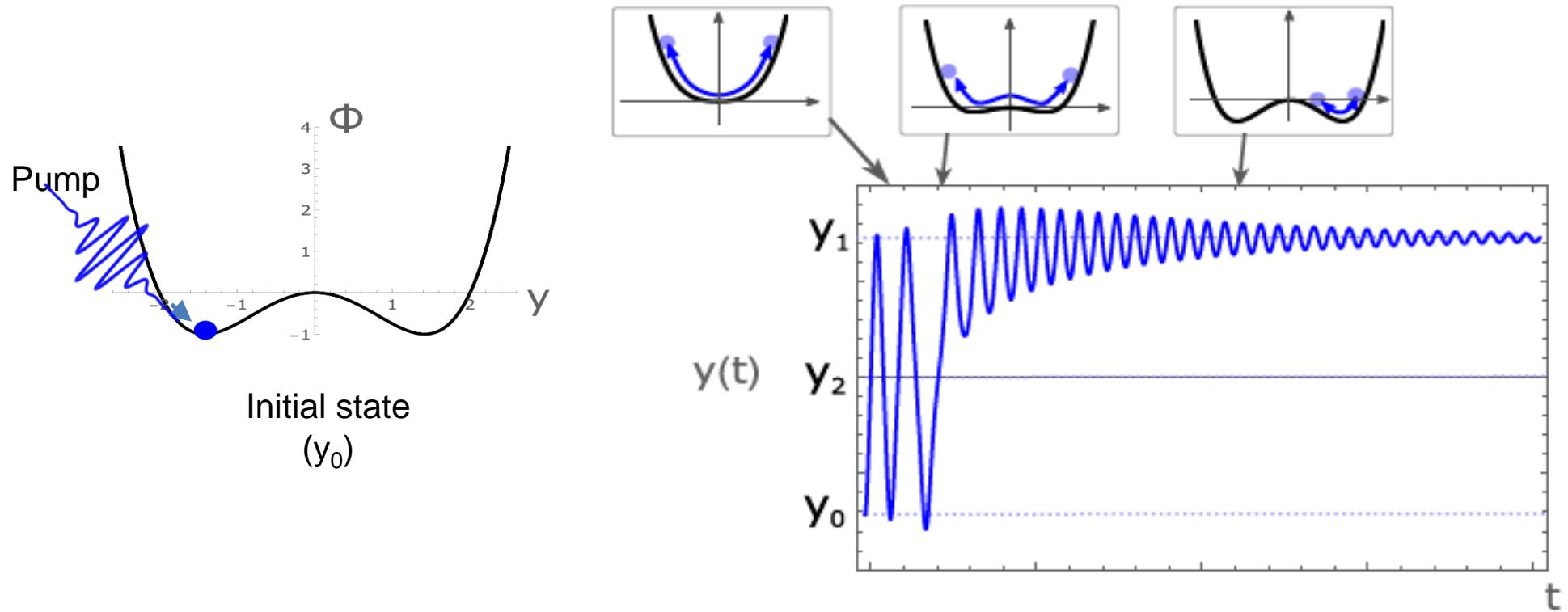
Switching as a function of pump and magnetic field

Inhomogeneous switching!

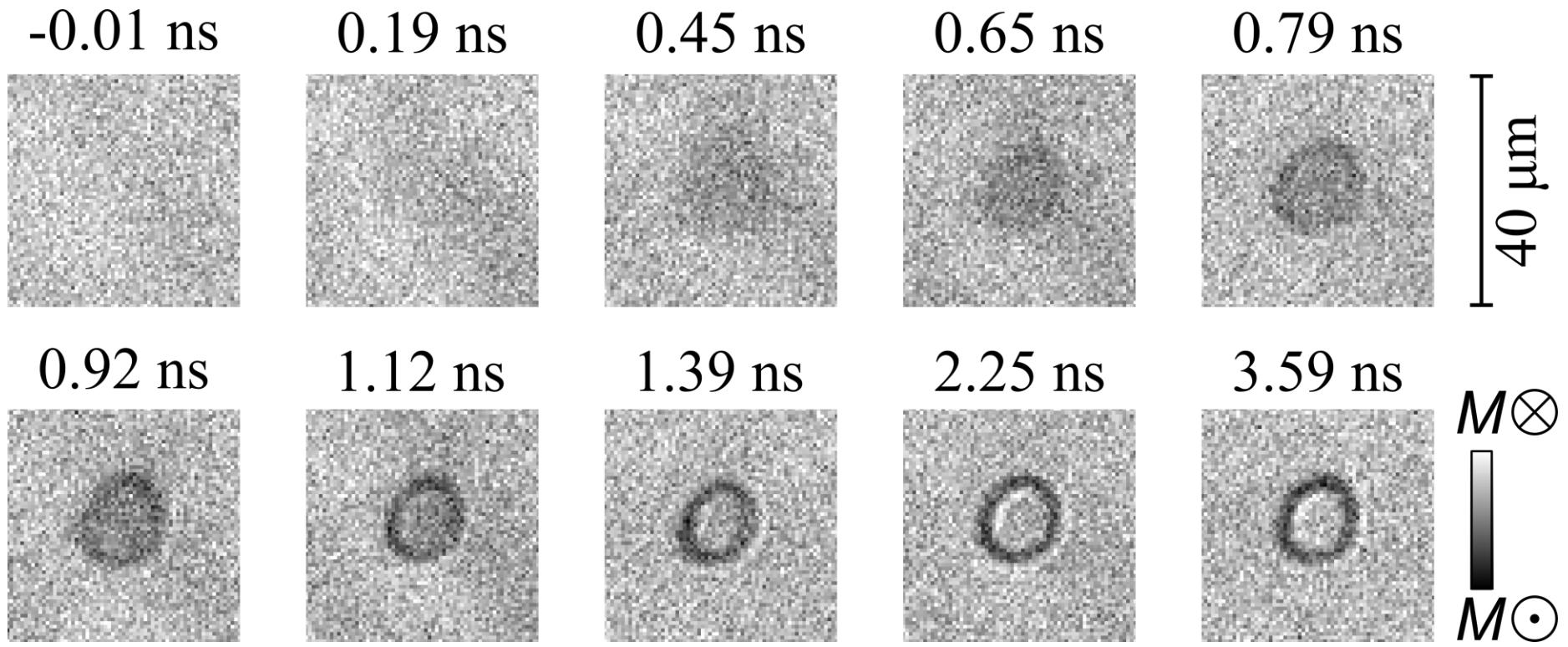


Toy model of the switching

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{H}_{eff} - \frac{\lambda}{M^2} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{eff})$$

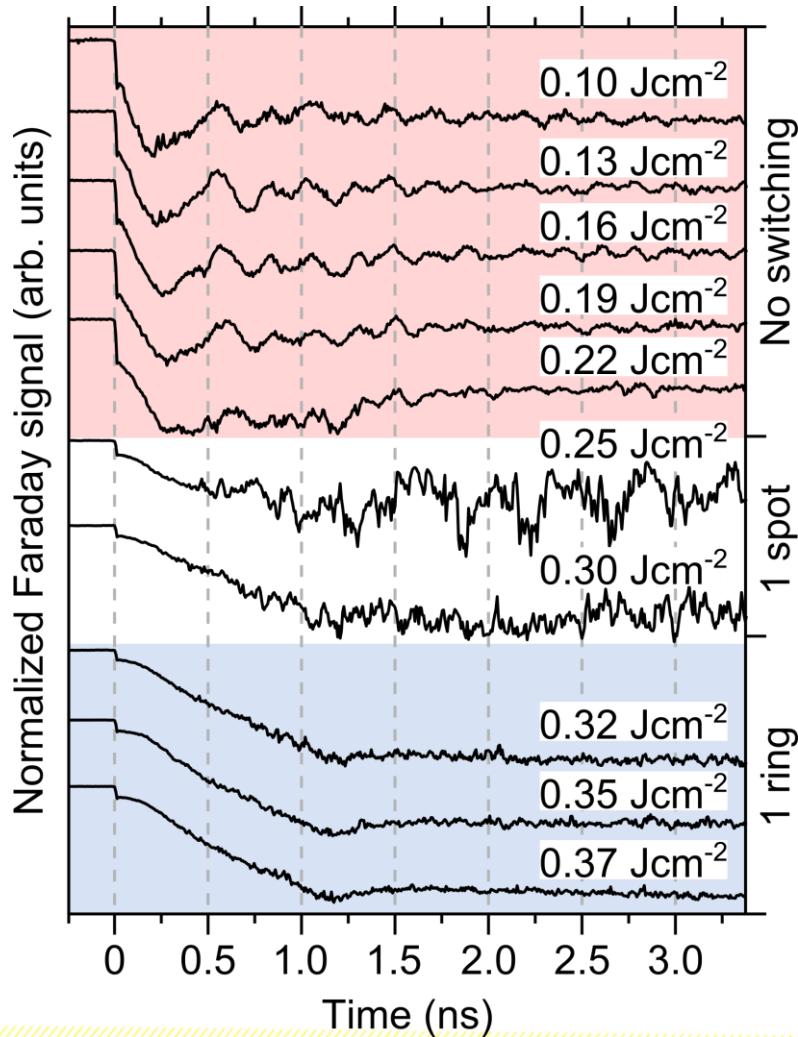


Dynamics of the switching



Huge damping!

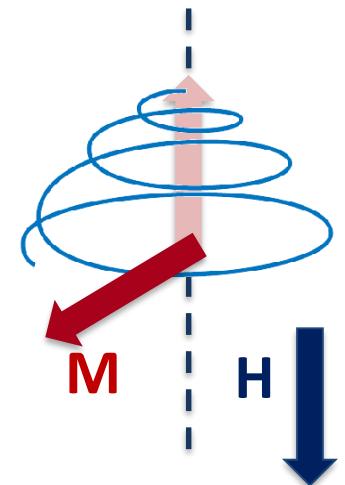
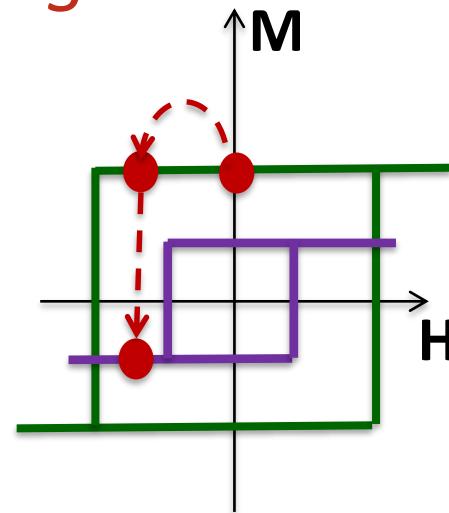
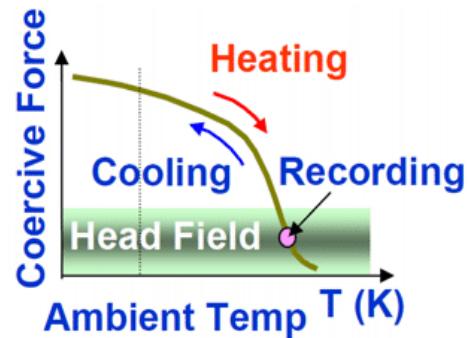
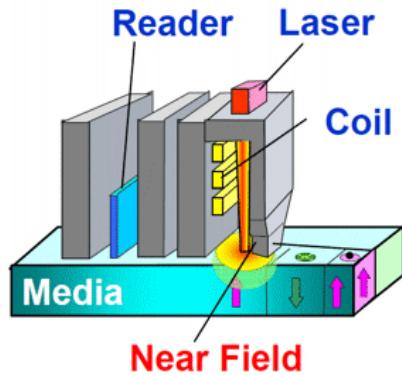
Damping as a function of amplitude



$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{H}_{eff} - \frac{\lambda}{M^2} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{eff})$$

λ is a function of the amplitude!

State-of-the-art in data storage: *heat assisted magnetic recording*

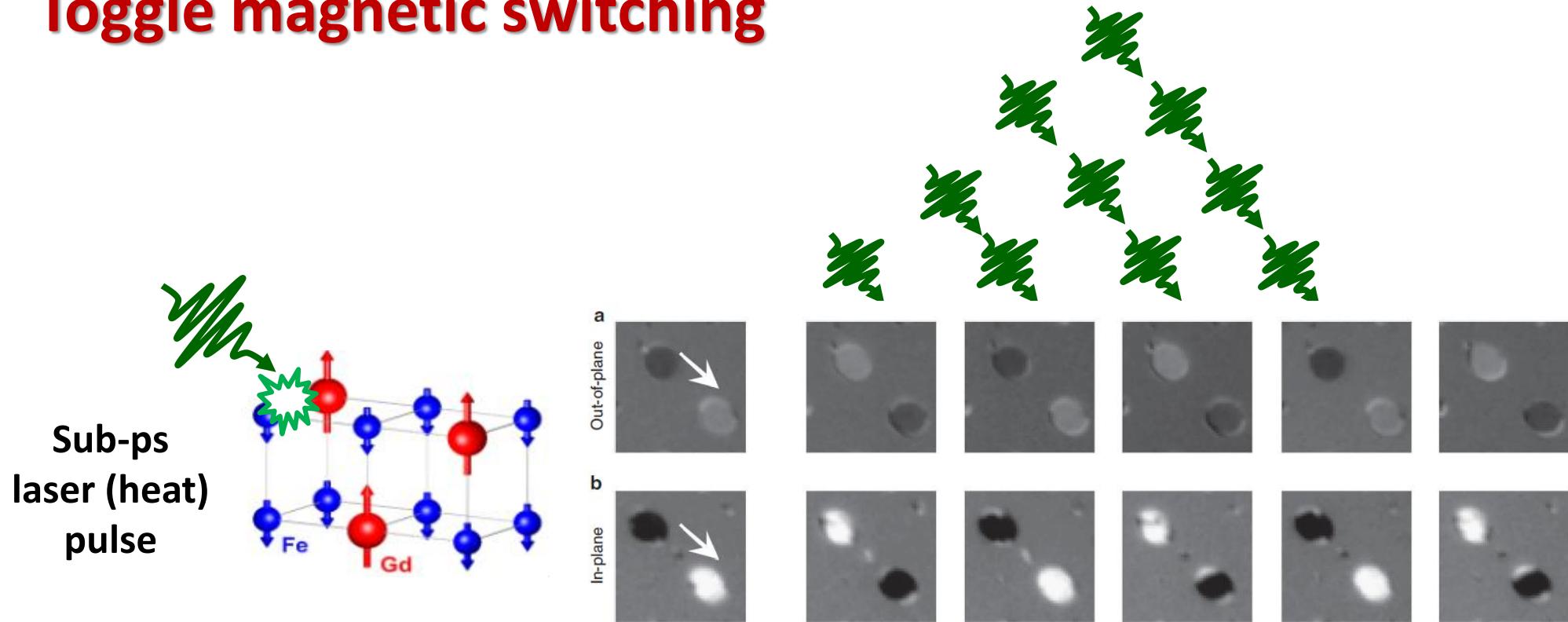


$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{H}_{eff} - \frac{\lambda}{M^2} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{eff})$$

Magnetization reversal

- with a magnetic field in a “wrong” direction;
- without any magnetic field and solely with heat;
- with no heat, no field.

Toggle magnetic switching



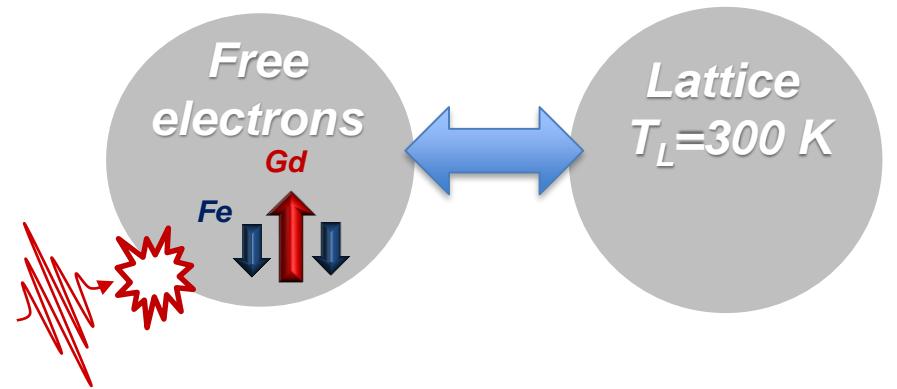
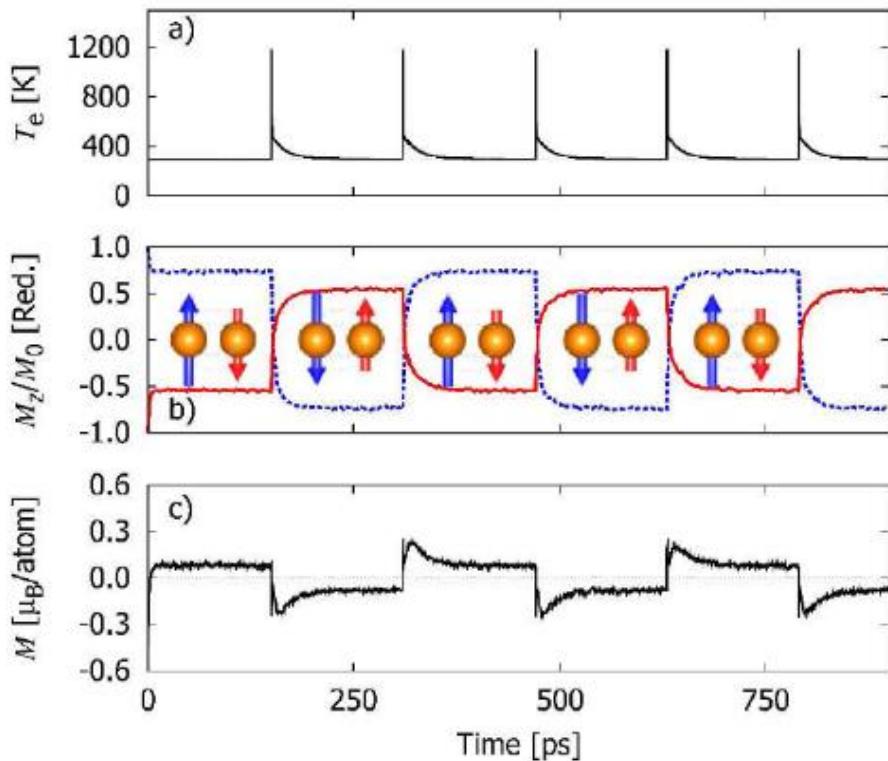
T. Ostler et al *Nature Comm.* **3**, 666 (2012).

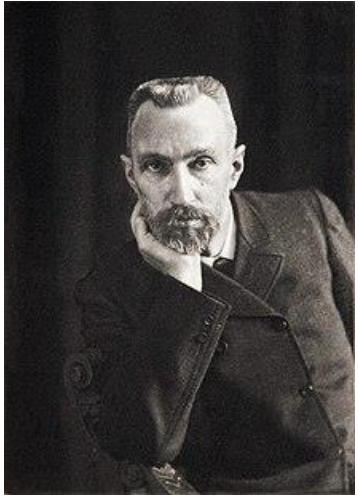
**Ultrafast toggle switching of magnetization
violates the Curie principle!**

Heat as a sufficient stimulus for magnetization reversal

“Two-spin” model. Heat-induced reversal

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



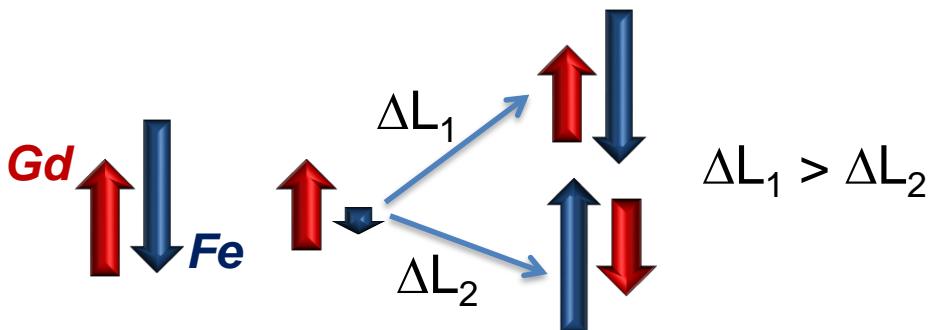


P. Curie (1894):

"the symmetries of the causes are to be found in the effects".

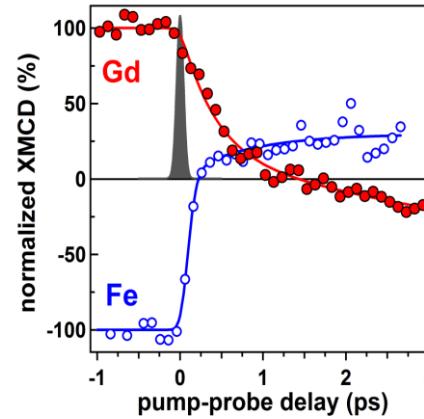
**How can heat-induced magnetization reversal
be possible at all?**

Two-spin model and conservation of angular momentum

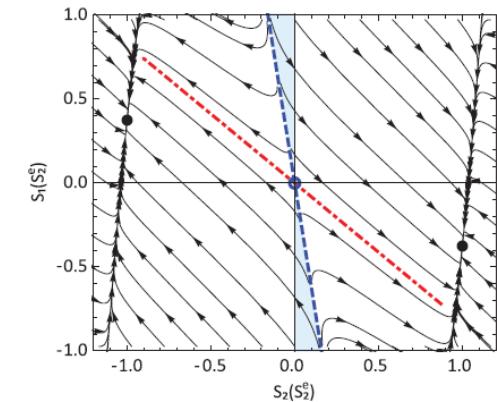


CREDIT: NIELS BOHR ARCHIVE, PHOTOGRAPH BY ERIK GUSTAFSON, COURTESY AIP EMILIO SEGRÉ VISUAL ARCHIVES, MARGRETHE BOHR COLLECTION

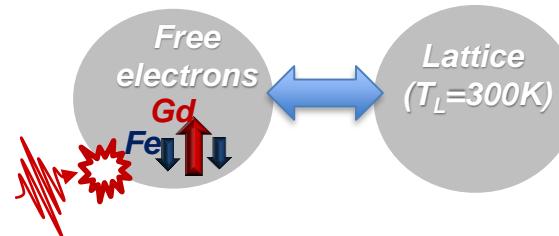
Physicists Wolfgang Pauli, left, and Niels Bohr demonstrating a "tippe top" toy in 1954. Tippe tops flip upside down to spin on their handle and are part of the "Secret Science of Toys" festival at the Fleet Science Center on Jan. 21.



I. Radu et al, *Nature* **472** 205-208 (2011).

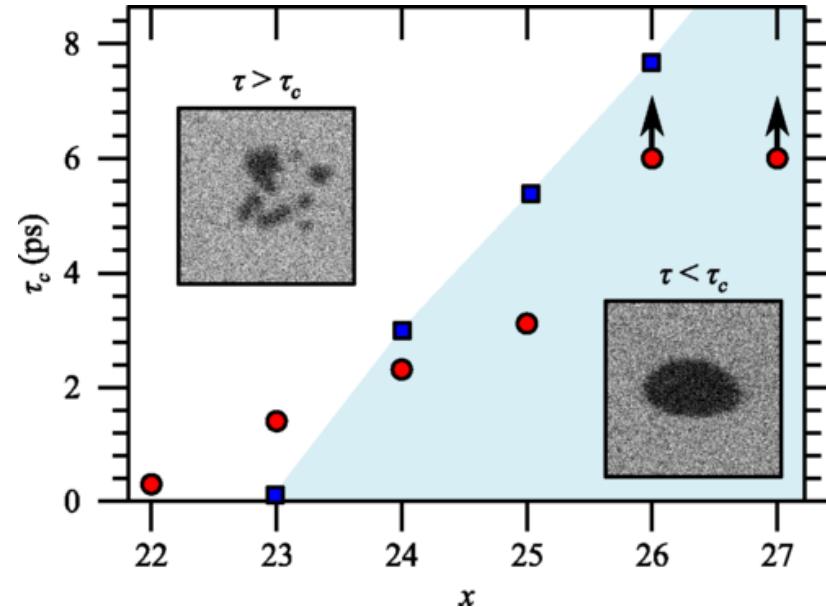
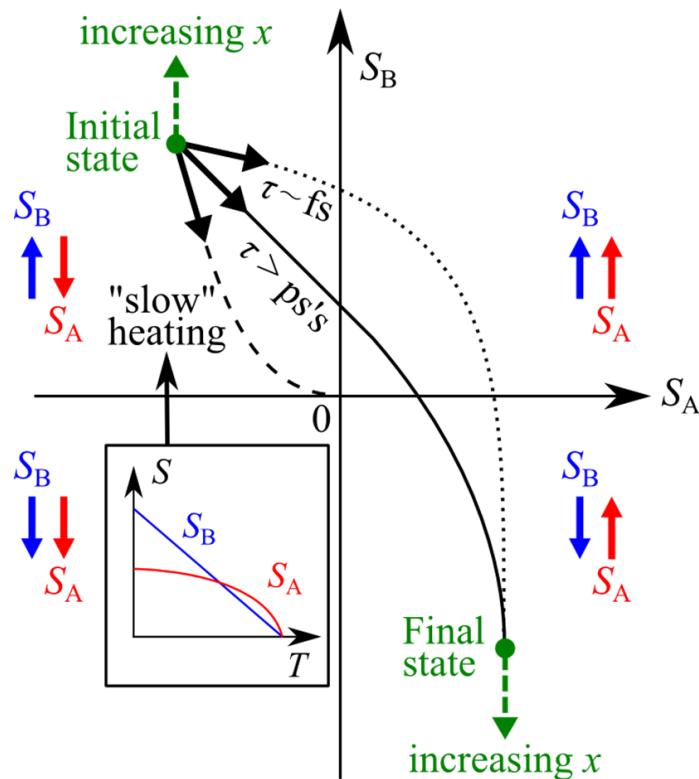


J. Mentink et al, *Phys. Rev. Lett.* **108**, 057202 (2012).



- 1) Hot electrons ($t < \tau_{e-ph}$)
- 2) Different demagnetizations ($t < \tau_{Fe-Gd}$)
- 3) Strongly non-equilibrium state ($t \sim \tau_{e-ph}$ and $t < \tau_{Fe-Gd}$)
- 4) Relaxation

Femtosecond vs picosecond pulse excitation



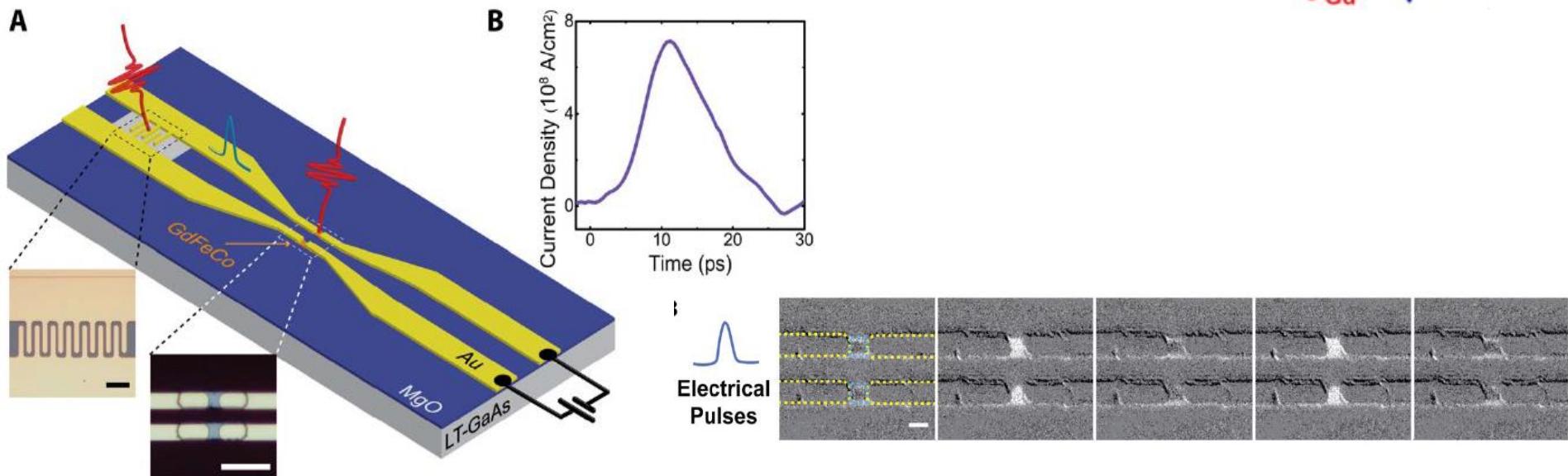
C. Davies et al, arXiv:1904.11977 (2019).
Phys. Rev. Applied 13, 024064 (2019).

**Sublattices are brought out of mutual equilibrium,
if excited faster than the Fe-Gd exchange interaction**

Ultrafast magnetization reversal by picosecond electrical pulses

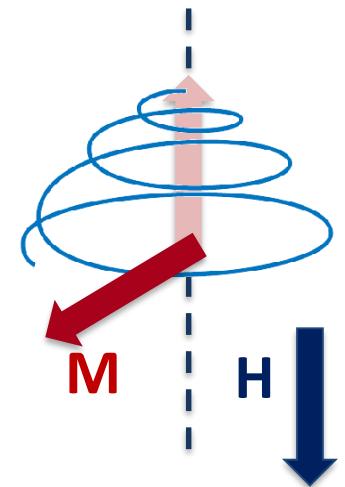
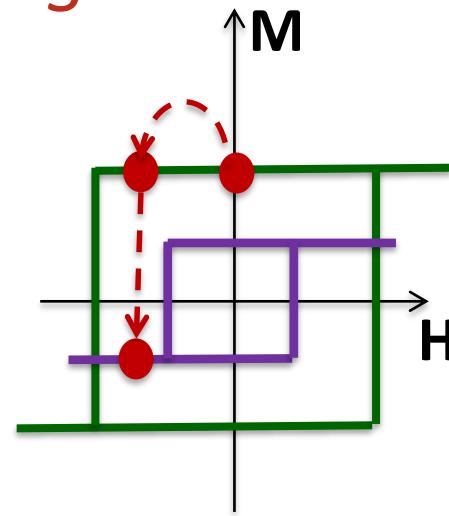
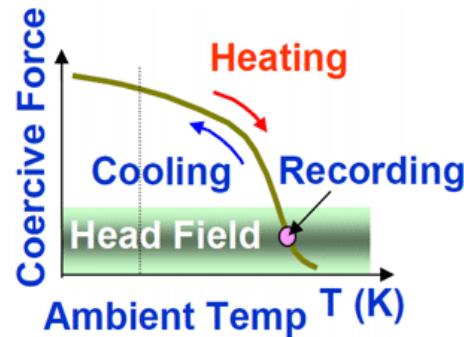
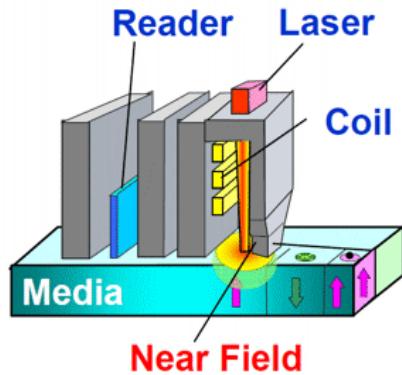
Yang Yang,^{1,*†} Richard B. Wilson,^{2,*†} Jon Gorchon,^{3,4,*} Charles-Henri Lambert,³ Sayeef Salahuddin,^{3,4} Jeffrey Bokor^{3,4†}

Sci. Adv. e1603117 3 (2017).



Ultrafast heating as a stimulus!

State-of-the-art in data storage: *heat assisted magnetic recording*



$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \mathbf{M} \times \mathbf{H}_{eff} - \frac{\lambda}{M^2} \mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{eff})$$

Magnetization reversal

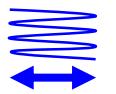
- with a magnetic field in a “wrong” direction;
- without any magnetic field and solely with heat;
- with no heat, no field.

Photo-magnetism of Co-substituted iron garnet

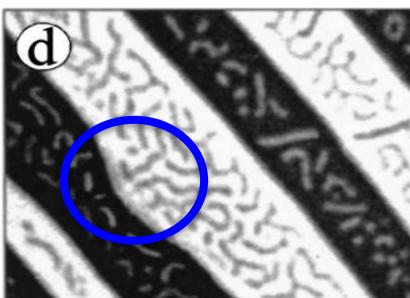
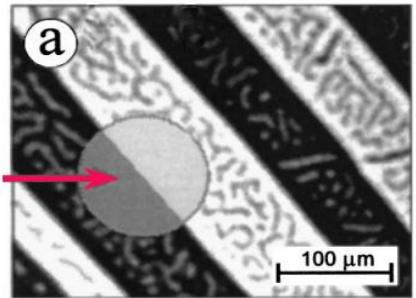
laser CW:



E||[110]



E||[1-10]

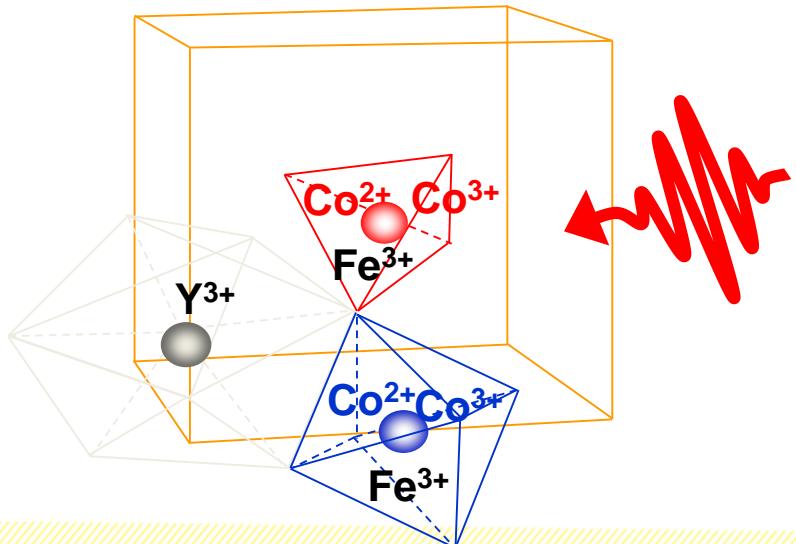


$\text{Y}_2\text{CaFe}_{3.9}\text{Co}_{0.1}\text{GeO}_{12}$ / GGG (001)

A.Chizhik et al. *PRB*, 57 (1998).

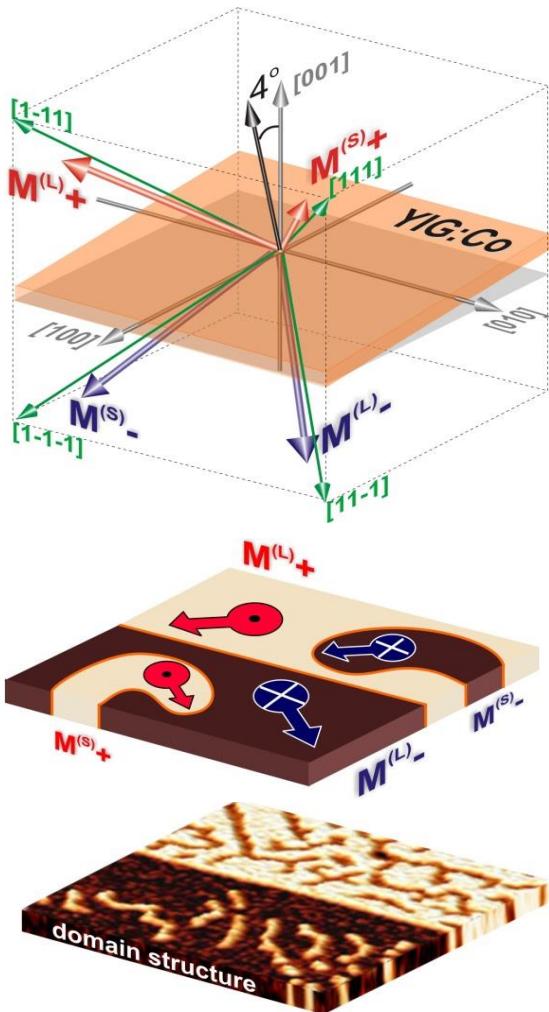
A.Stupakiewicz et al. *PRB*, 64 (2001).

Light-induced slow ($\sim \mu\text{m/sec}$) motion of domain wall



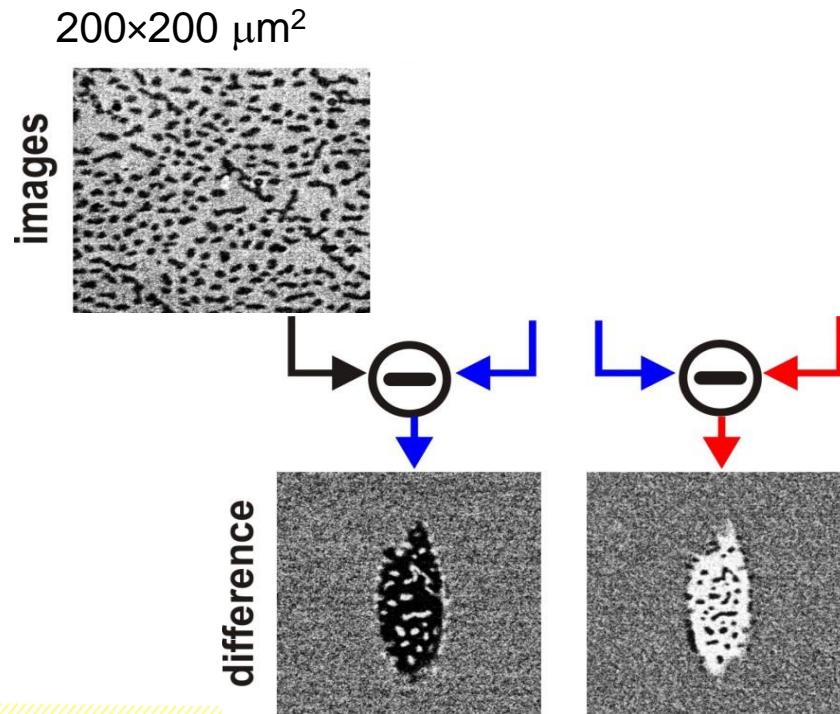
**Recording?
Heating?
Speed?**

Photo-magnetic recording in iron garnet



$\text{Y}_2\text{CaFe}_{3.9}\text{Co}_{0.1}\text{GeO}_{12}$ on GGG (001)
thickness $d=7.5 \mu\text{m}$ (grown by LPE)

A. Stupakiewicz et al,
[arXiv:1609.05223](https://arxiv.org/abs/1609.05223)
Nature **542**, 71 (2017).

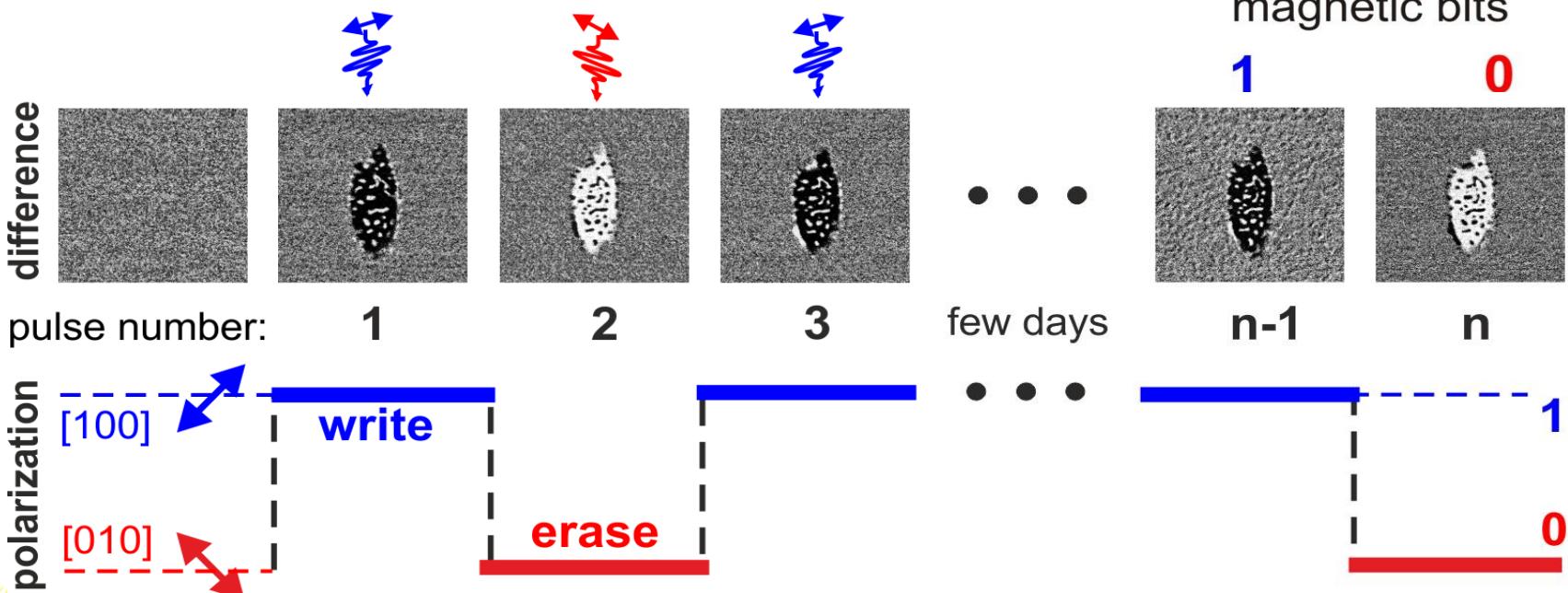
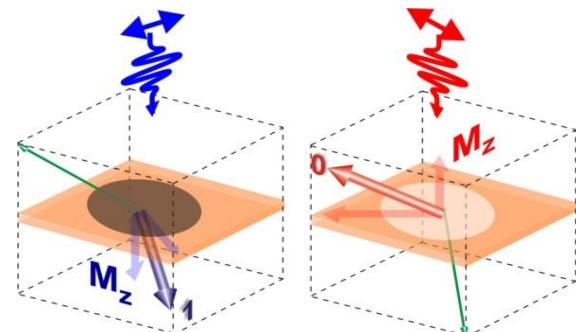


change perspective

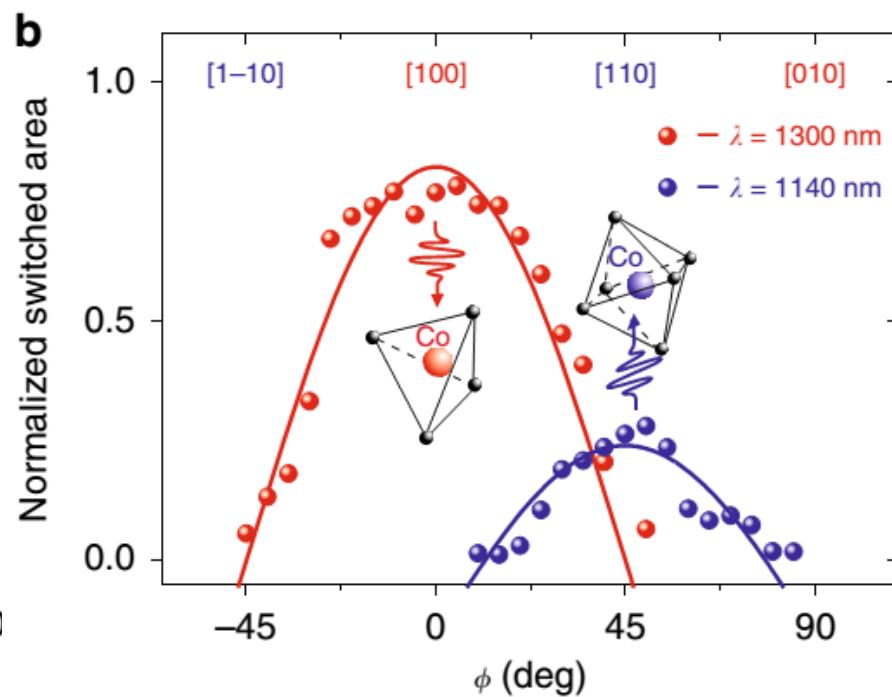
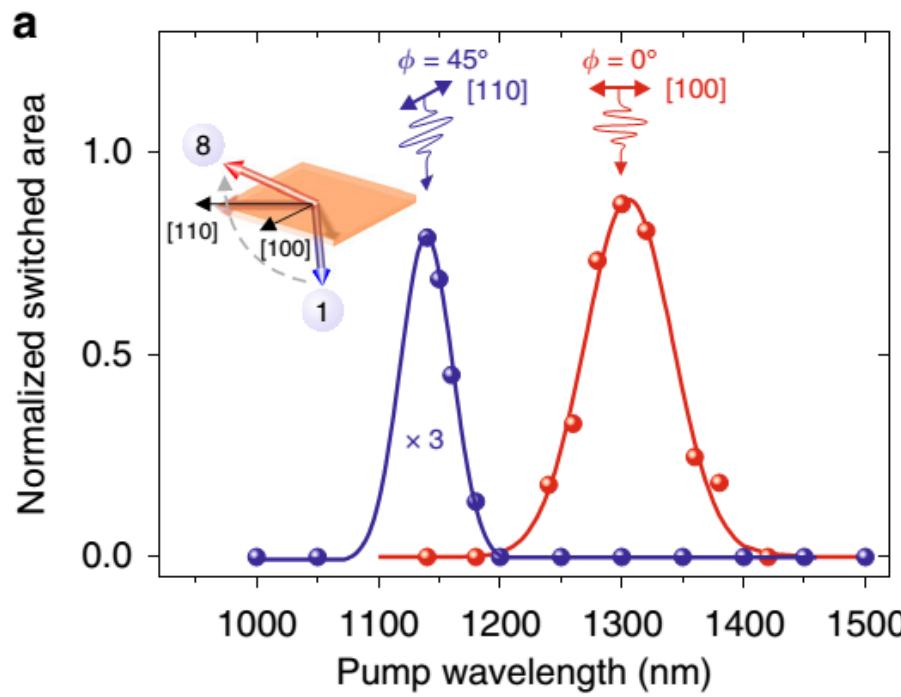
Photo-magnetic recording

- ✓ single pulse
- ✓ repeatable switching
- ✓ zero applied field
- ✓ room temperature

A. Stupakiewicz et al,
[arXiv:1609.05223](https://arxiv.org/abs/1609.05223)
Nature 542, 71 (2017).



Selection rules for magnetic writing with light on iron-garnet

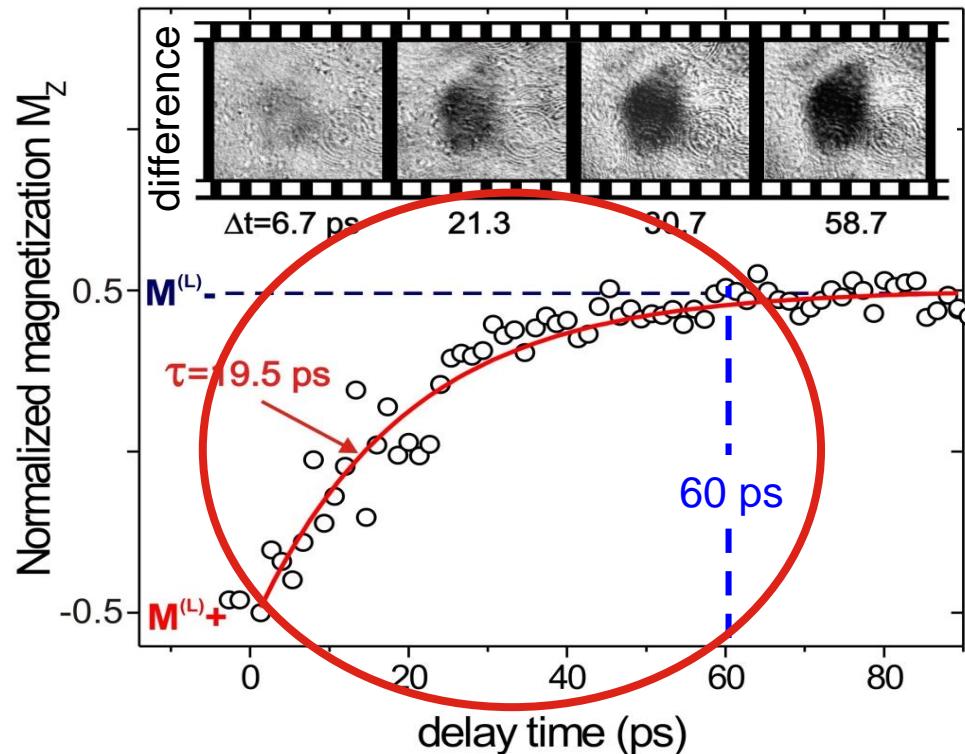


Optical resonant excitation of tetrahedral and octahedral
Co²⁺ sublattices

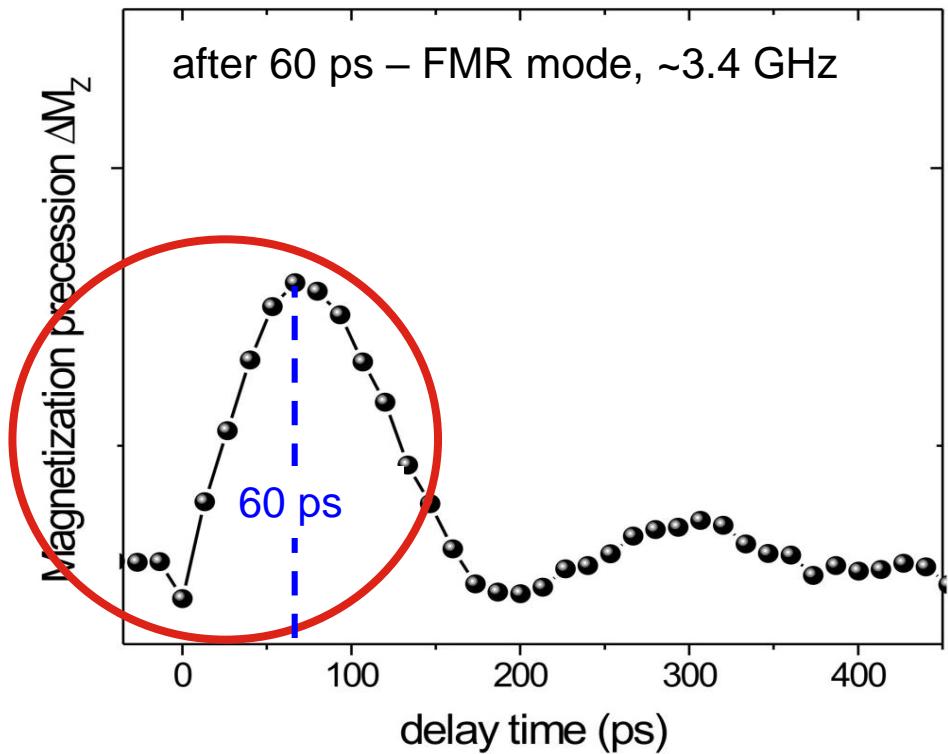
A. Stupakiewicz et al, *Nature Comm* **10**, 612 (2019).

Dynamics of the laser-induced switching in garnet

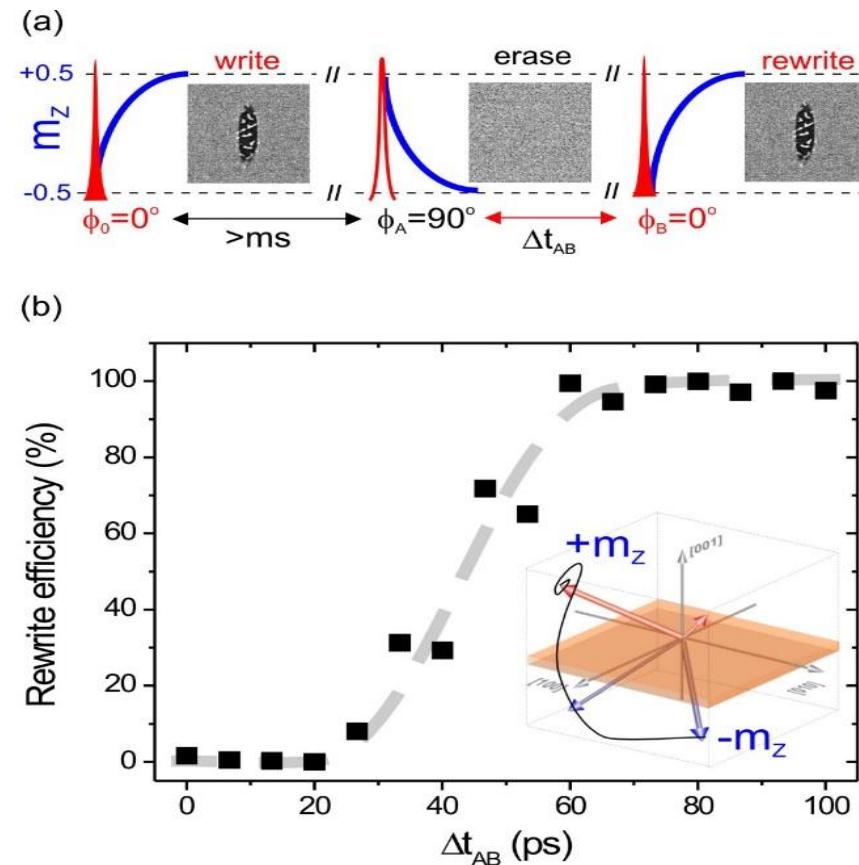
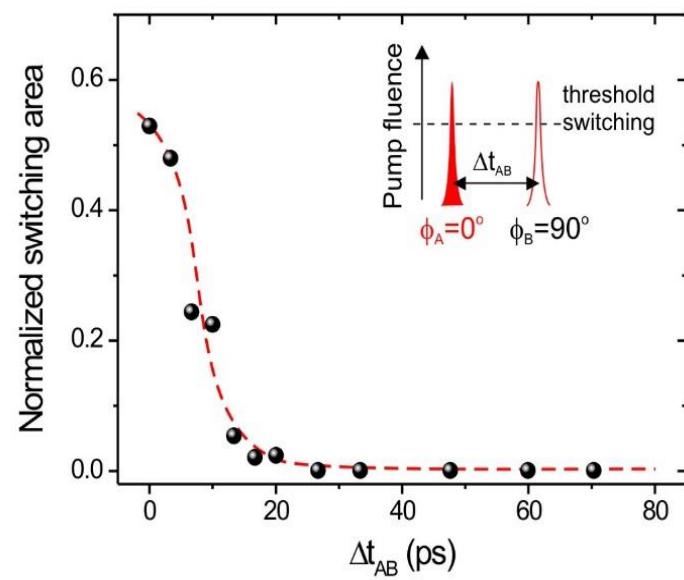
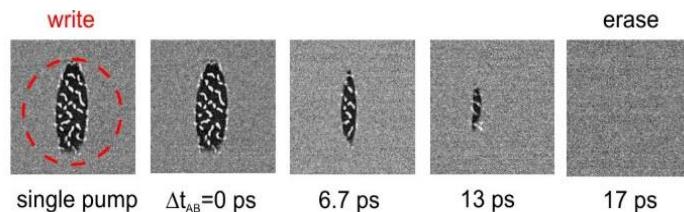
switched after the first quarter-period



high Gilbert damping: $\alpha = 0.2$



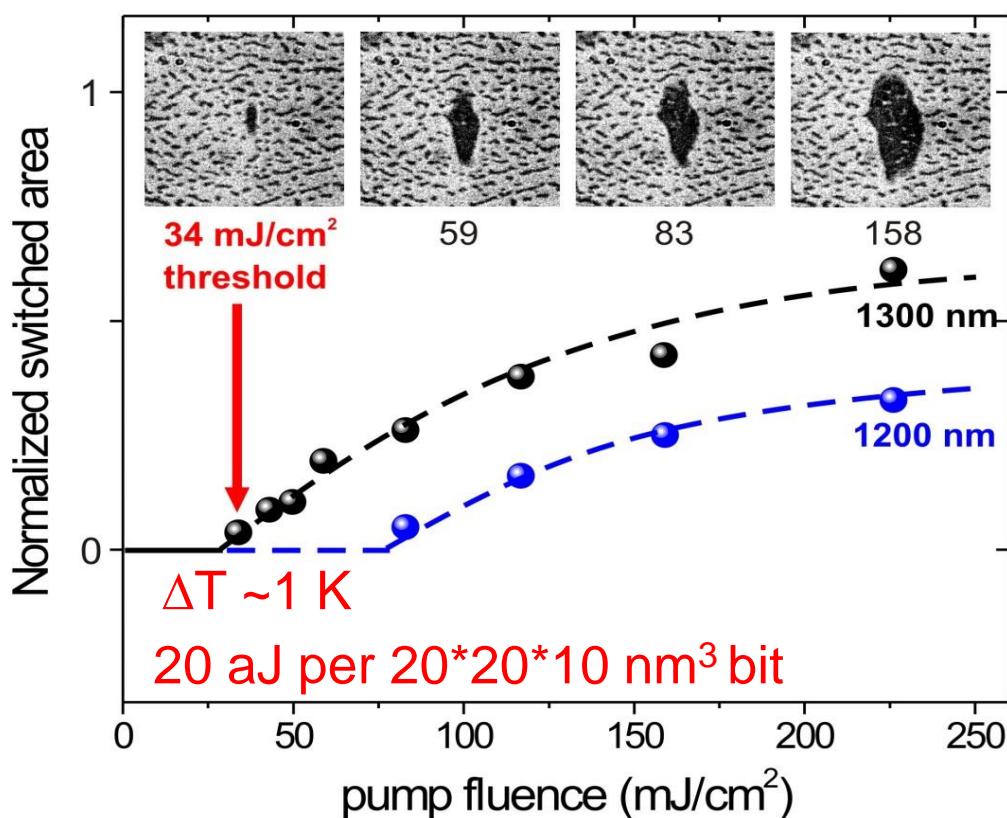
Fundamental limits of repetition rate of magnetic writing on $\text{Y}_3\text{Fe}_5\text{O}_{12}:\text{Co}$



Rewriting at the frequency of 20 GHz (50 ps per bit)!

K. Szerenos et al. *Phys. Rev. Applied* **12**, 044057 (2020).

Efficiency of the photo-magnetic recording



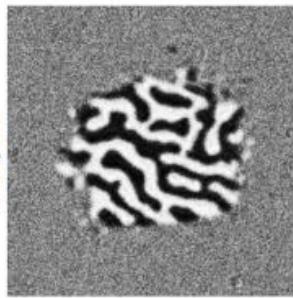
Optical resonant excitation
of tetrahedral Co^{2+}



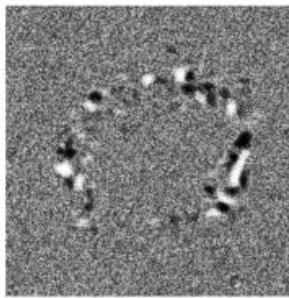
Ultrafast all-optical toggle writing of magnetic bits without relying on heat

T. Zalewski¹, A. Maziewski¹, A. V. Kimel² & A. Stupakiewicz¹

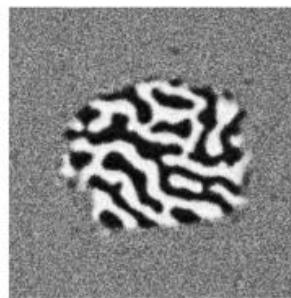
pulses: n=1



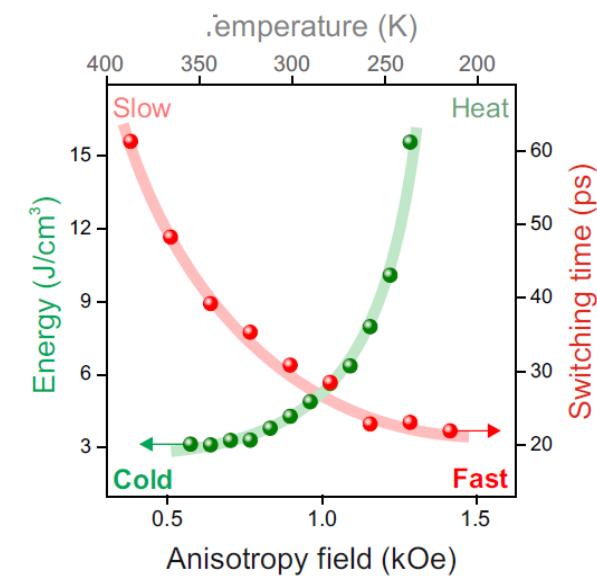
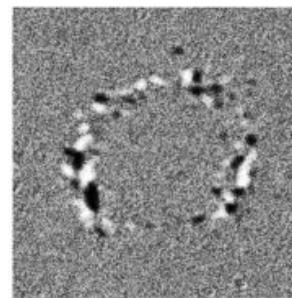
n=2



n=3



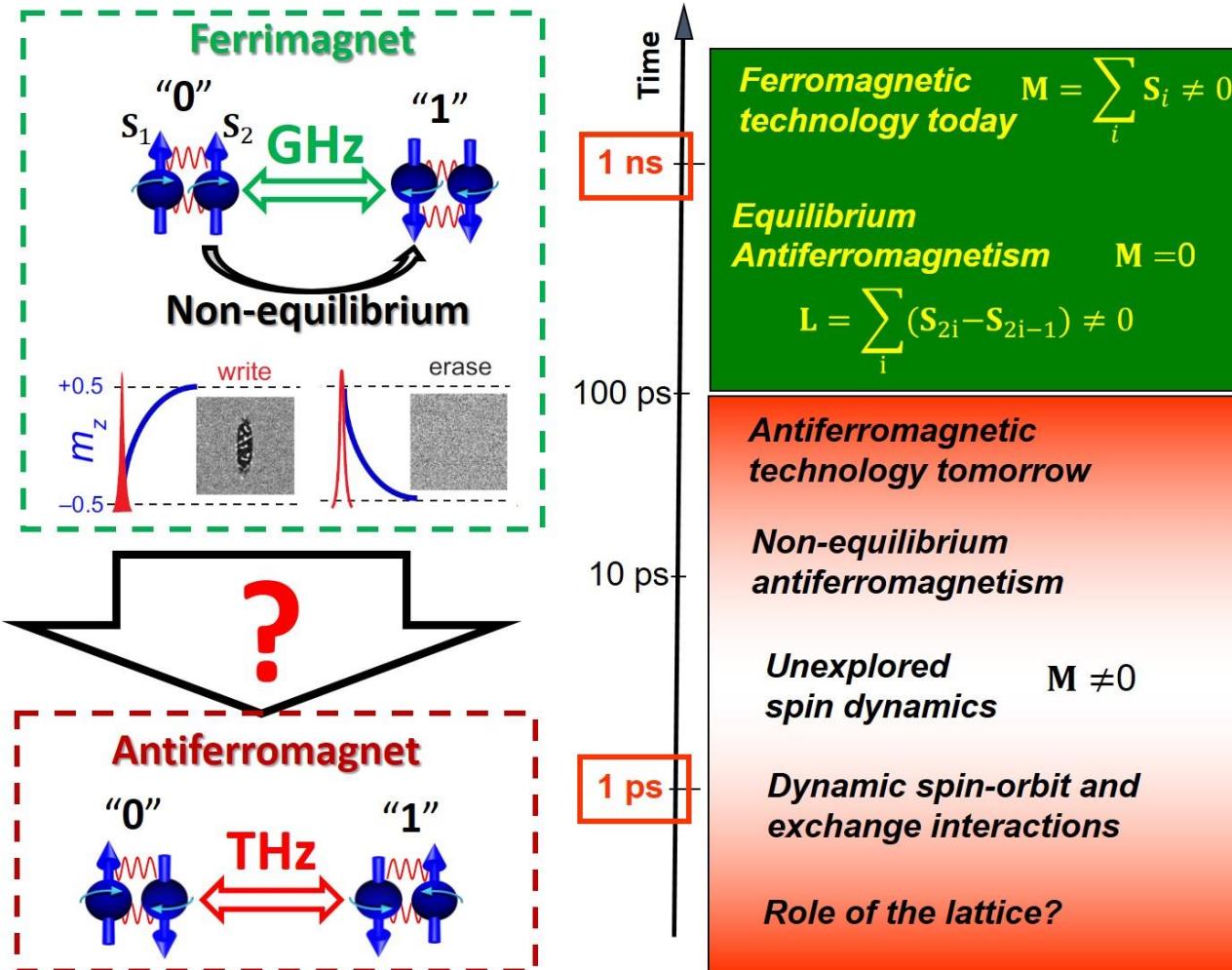
n=4



Summary

- Understanding magnetization reversal is the key for understanding the fundamental limits on the rate of writing of magnetic bits.
- Ultrafast magnetization reversal is a counter-intuitive process.
- If a stimulus is ultrafast, magnetization can be reversed with a “wrong” magnetic field, solely with heat or even without any heat.

Outlook – ultrafast antiferromagnetism as the next challenge



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