Fundamentals of light-driven ultrafast spin dynamics in magnetic materials

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Vetenskapsrådet

SEVENTH FRAMEWORK PROGRAMME

femtoSpin

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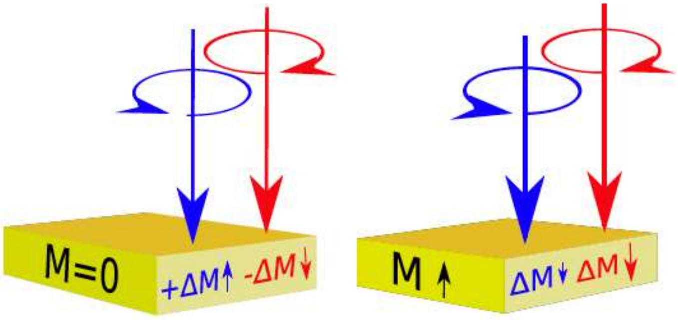
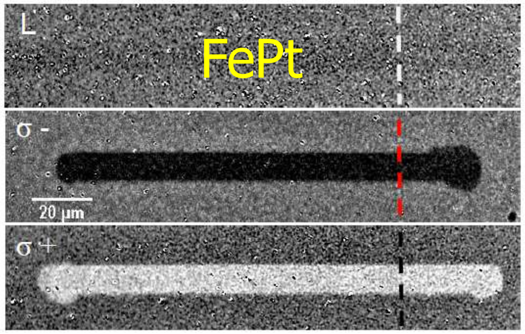
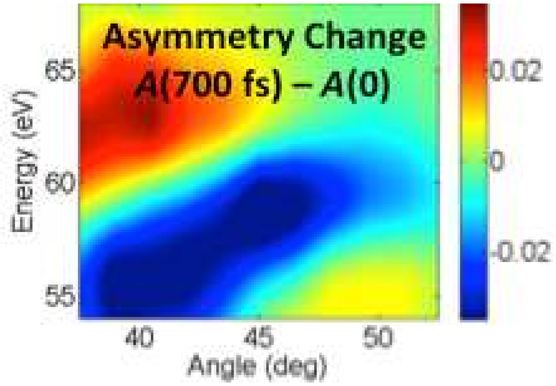
SPIN PHENOMENA
INTERDISCIPLINARY CENTER
SPICE

Outline

- All-optical switching – experimental results
- *Ab initio* theory for laser-imparted *opto-magnetism*
  & multiscale theory for all-optical switching
- Ultrafast laser-induced demagnetization
  - what do we know after 20 years?
- Fundamental mechanisms - Exchange splitting collapse,
  magnon excitation & spin transport

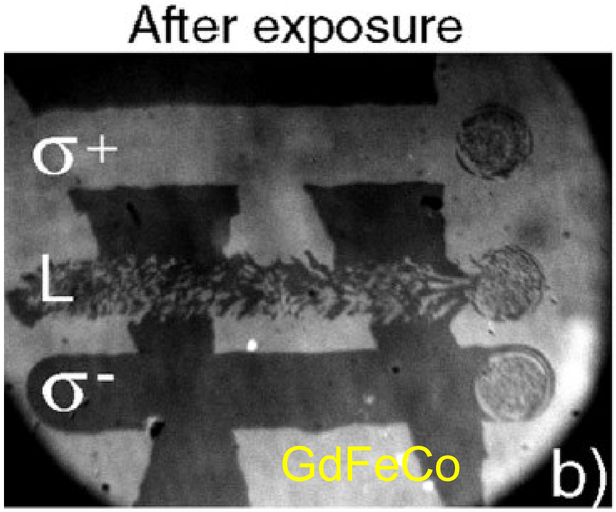


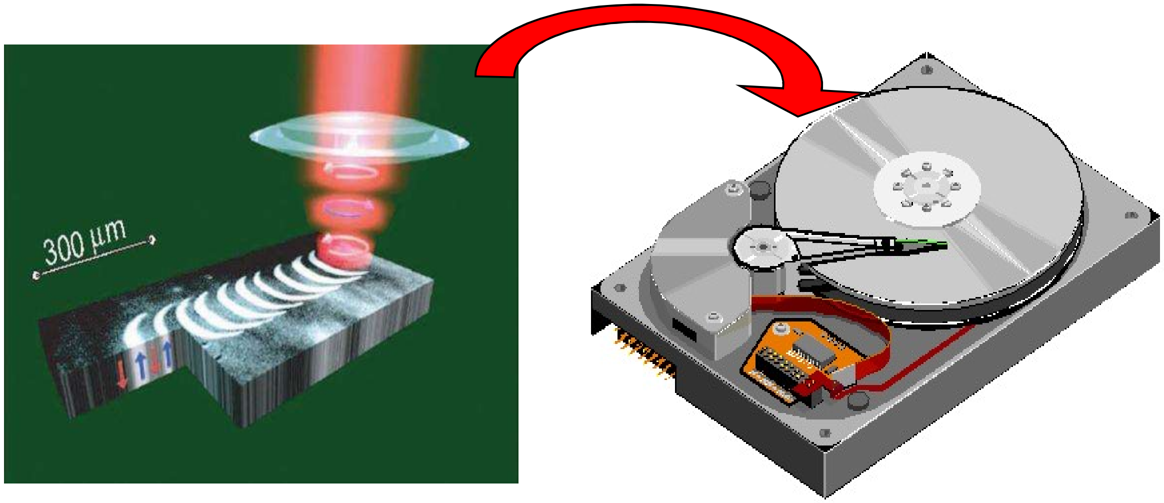




All-optical writing of magnetic domains

Laser-induced magnetization reversal: all-optical helicity-dep. recording



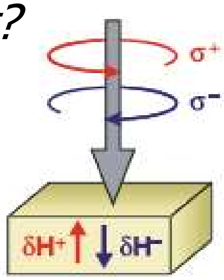


Stanciu et al, PRL **99**, 047601 (2007)

- All-optical writing on GdFeCo medium
- Due to *inverse Faraday effect?*

➡ $\vec{M}^{ind} \propto v_{IFE} \cdot E_i E_j^*$

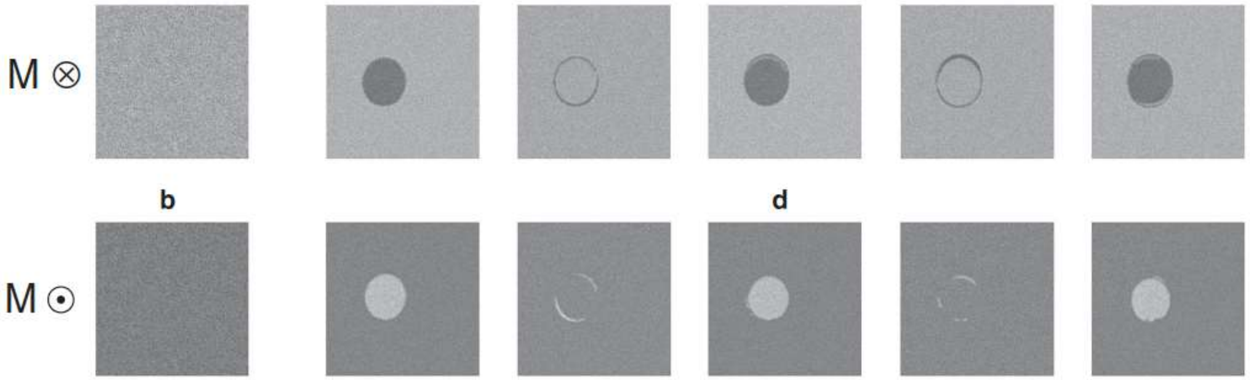
nonlinear effect



- Erasing & writing with circ.-pol. fs pulses
- Approx. 10^3 times faster recording ?

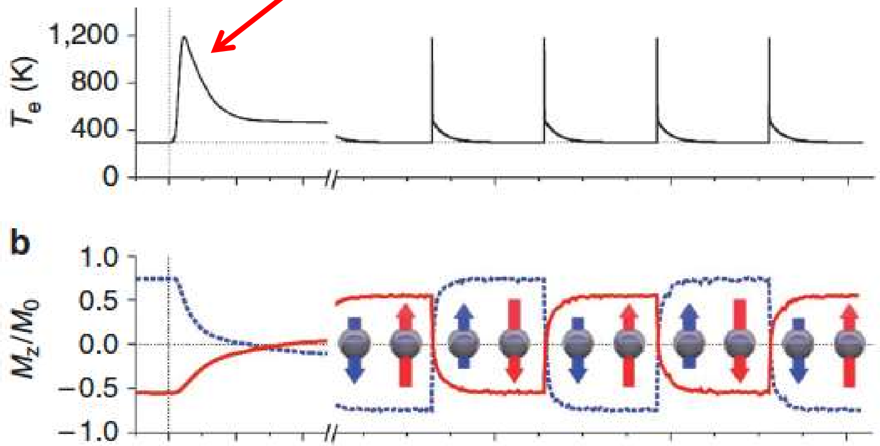
Helicity-independent toggle switching in GdFeCo

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

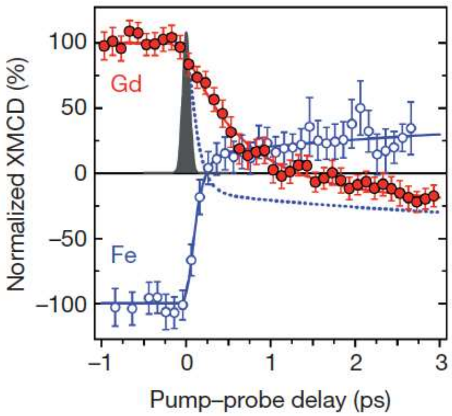


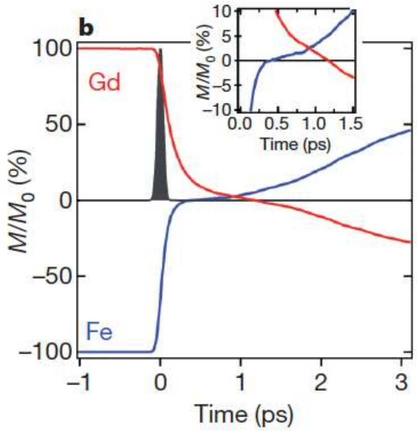
Atomistic LLG simulations coupled to two temp. model, distinct Fe and Gd dynamics

T_e



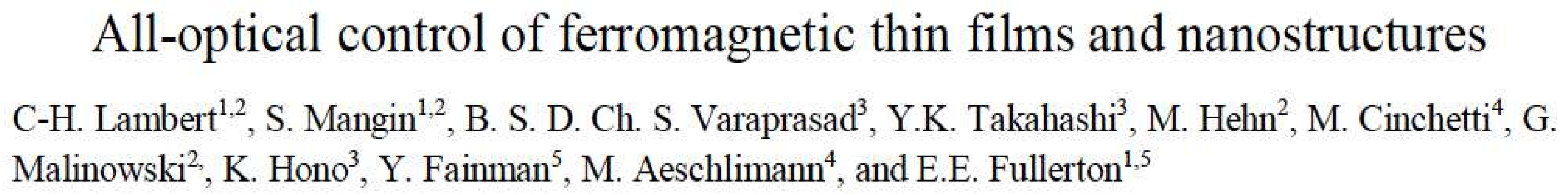
100 fs pulses





Radu et al, Nature **472**, 205 (2011)

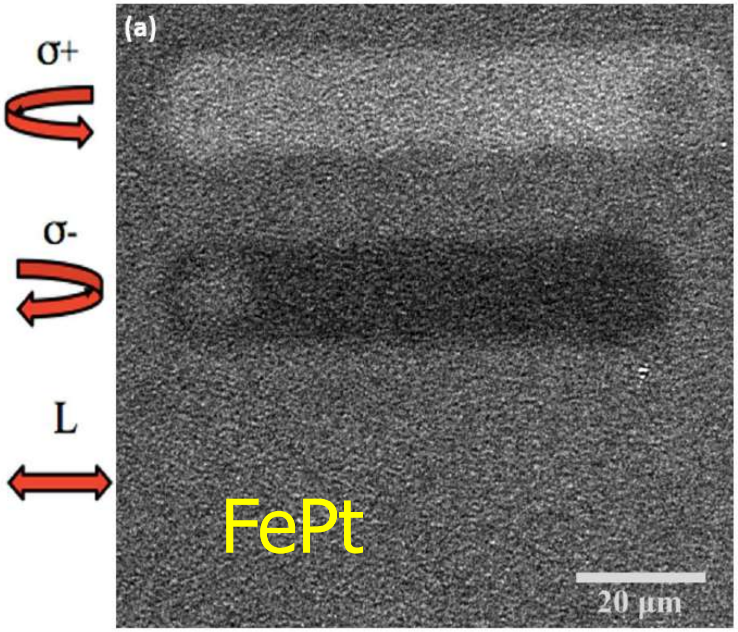
Helicity-dependent reversal of magnetization



All-optical control of ferromagnetic thin films and nanostructures

C-H. Lambert[1,2], S. Mangin[1,2], B. S. D. Ch. S. Varaprasad[3], Y.K. Takahashi[3], M. Hehn[2], M. Cinchetti[4], G. Malinowski[2], K. Hono[3], Y. Fainman[5], M. Aeschlimann[4], and E.E. Fullerton[1,5]

Science **345**, 1337 (2014)

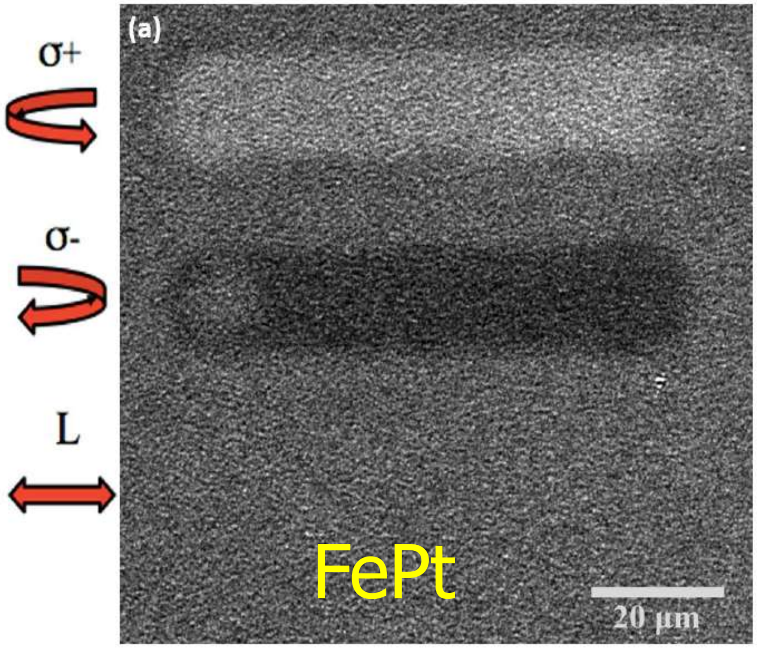


AO-HD reversal works for synthetic antiferromagnets and ferromagnets & recording material FePt

Mangin et al, Nature Mater. **13**, 286 (2014)

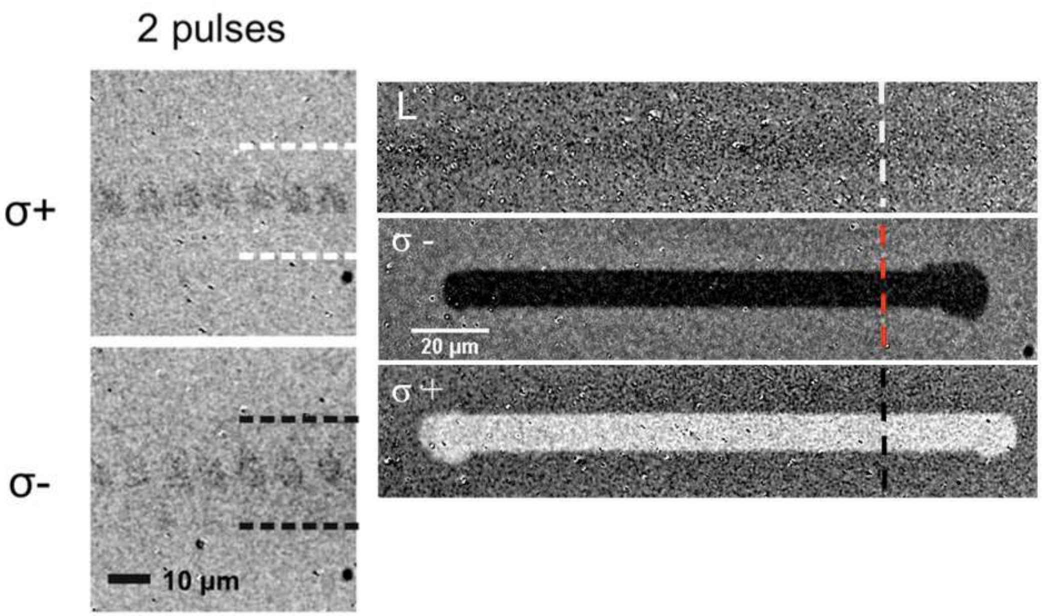
Towards few shot switching of recording media

Continuous laser motion



Lambert et al, Science **345**, 1337 (2014)

Repeated single shots



John et al, Sci. Rep. **7**, 4114 (2017)

Microscopic origin?

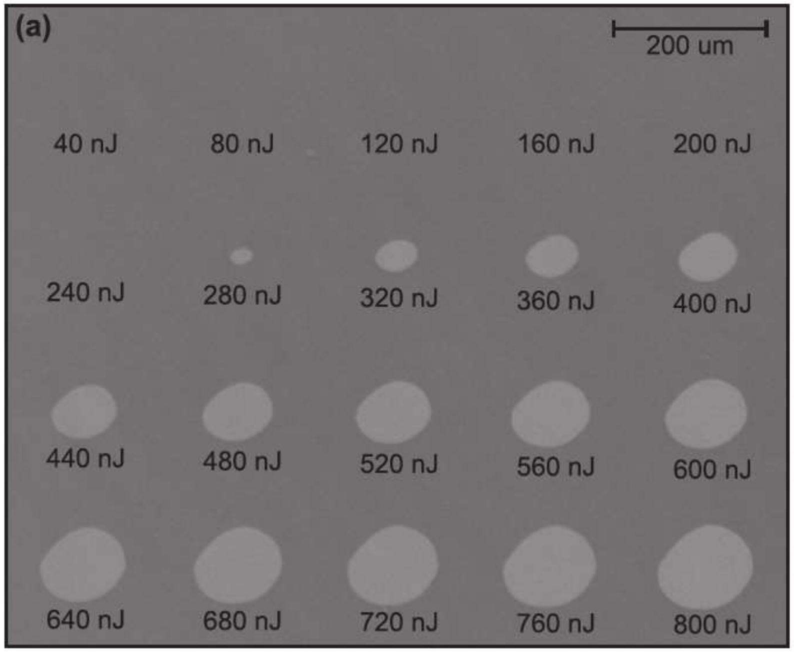
$$P_{abs} \propto \mathrm{Re}[i\omega\varepsilon_{ij}(\omega) \cdot E_i E_j^*]$$

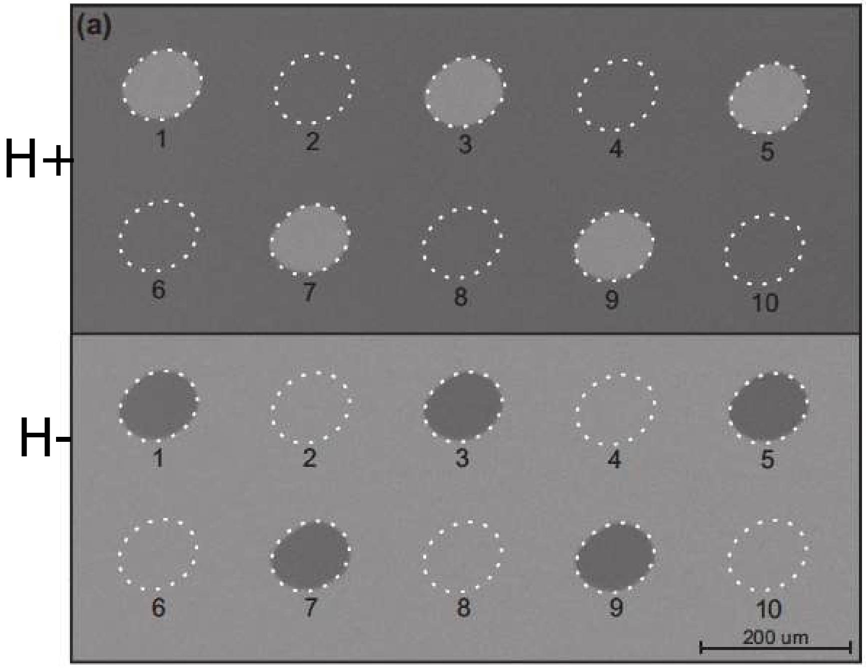
- Helicity dependent laser heating (MO effect, $n^+ \neq n^-$)?
- Helicity dependent induced magnetization (IFE) ?

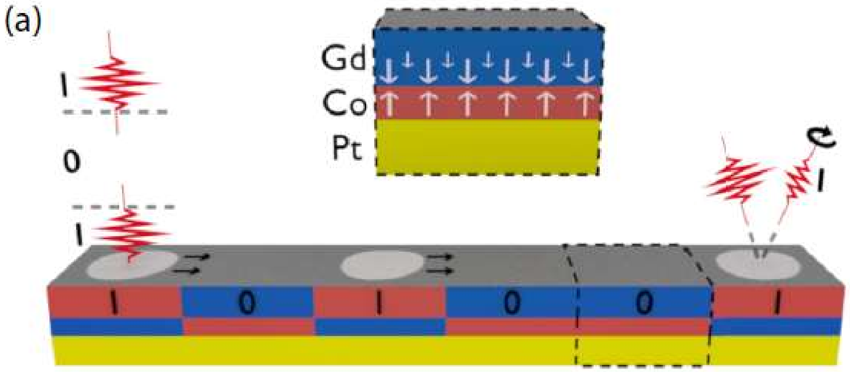
$$\vec{M}^{ind} \propto v_{IFE} \cdot E_i E_j^*$$

All-optical single-pulse toggle switching

Lalieu et al, PRB **96**, 22041R (2017)



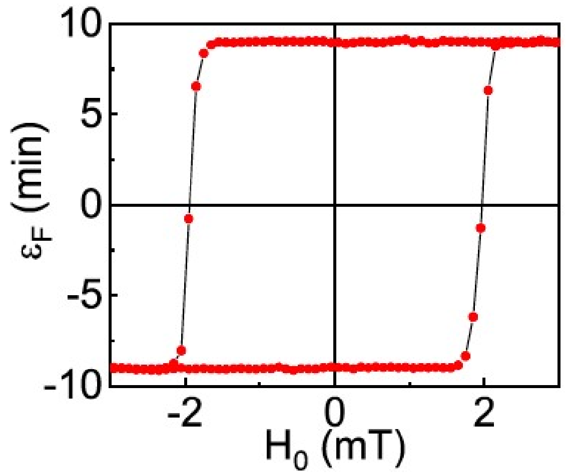


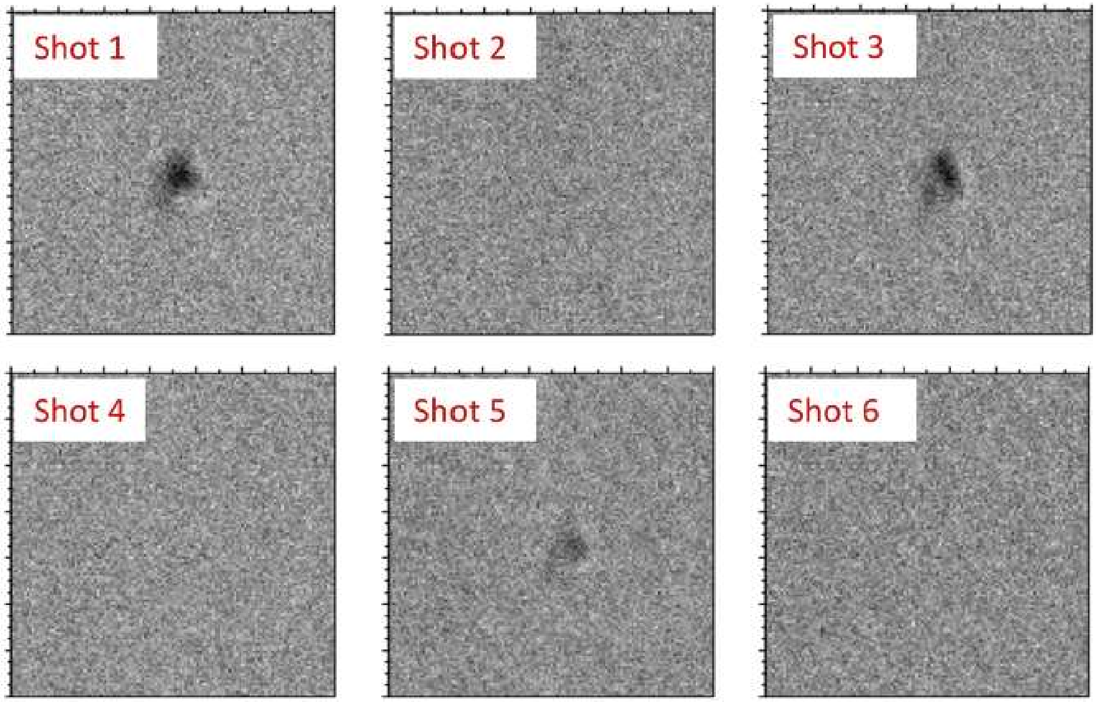


with linearly polarized light – suggests *heat-driven* mechanism, ferri-magnetic Gd-Co interlayer exchange

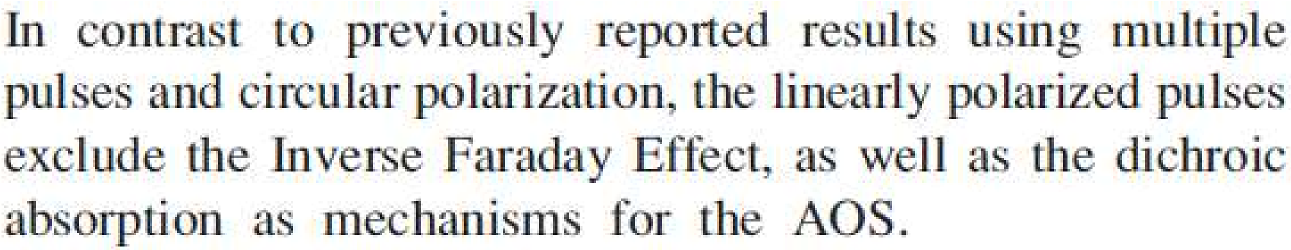
Towards single shot all-optical switching

Vomir, Albrecht & Bigot, Appl. Phys. Lett. **111**, 242404 (2017)





Pt: 1 nm
Co: 0.7 nm
Pt: 0.7 nm
Co: 0.7 nm
Pt or Ta: 2 nm



In contrast to previously reported results using multiple pulses and circular polarization, the linearly polarized pulses exclude the Inverse Faraday Effect, as well as the dichroic absorption as mechanisms for the AOS.

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Fundamentals: Need theoretical modeling

Ab initio calculations

induced magnetization (IFE)

exchange constants, damping,

n±, dichroic absorption, C_e

Micromagnetic modeling

Atomistic LL-Gilbert or

microscopic LL-Bloch equation,

Two-temp. model (T_e, T_L)

1) Dichroic absorption: Kubo linear response theory

$$\boxed{\sigma_{\alpha\beta}(\omega) = -\frac{ie^2}{m^2\hbar V}\sum_{nn'}\frac{f(\epsilon_n)-f(\epsilon_{n'})}{\omega_{nn'}}\frac{\Pi^{\alpha}_{n'n}\Pi^{\beta}_{nn'}}{\omega-\omega_{nn'}+i/\tau}}$$

lifetime

(Relativistic ab initio DFT calculations)

$$\boldsymbol{\epsilon}(\omega) = \mathbf{1} + \frac{4\pi i}{\omega}\boldsymbol{\sigma}(\omega) \Rightarrow n_{\pm}^{2}(\omega) = \varepsilon_{xx}(\omega) \pm i\varepsilon_{xy}(\omega)$$

Thin film geometry: $\mu^{\pm} = \frac{2\omega}{c}\mathrm{Im}[n^{\pm}]$, $\underbrace{P^{\pm}(z,t) = \mathrm{Re}[n^{\pm}]I \cdot e^{-\mu^{\pm}z}}_{\text{Hel. dep. absorption* and thus heating}}$

(spin-orbit effect)

Hel. dep. absorption* and thus heating

2. Theoretical understanding of IFE ?

Pitaevskii (1961): thermodynamic theory (non-absorb. medium)

IFE proportional to Faraday Effect (?)

$$\vec{M}^{ind} \propto \frac{\partial \varepsilon_{ij}}{\partial \vec{H}} E_i E_j^* \propto v_{IFE} \cdot E_i E_j^* \quad \longleftrightarrow \quad \theta_F \propto \frac{\partial \varepsilon}{\partial H} Hd = v_{FE} \cdot Hd$$

$$(v_{IFE} = v_{FE})$$

Precise quantum theory required; metals are absorbing!

⇒ Pitaevskii's expression not applicable

Quantum approach:
- response theory quantum formulation
- *Ab initio* materials specific calculations

Other theories: Pershan et al, PR **143** (1966), Hertel JMMM **303** (2006)

Density-matrix theory – Quantum expression

Liouville-von Neumann eq. $$i\hbar \frac{d\hat{\rho}(t)}{dt} = [\hat{H}(t),\hat{\rho}(t)] - i\hbar\Gamma\hat{\rho}(t)$$

With circularly pol. light as perturbation to *H* $[H' \propto p \cdot A(t)]$

Compute induced: $$\boldsymbol{M}_{\text{ind}} = \mu_B \mathrm{Tr}\{(\hat{\boldsymbol{L}} + 2\hat{\boldsymbol{S}})\hat{\rho}^{[2]}\}$$

$$\boxed{M_{\text{ind}} = (\mathcal{K}_{\text{o}} + \mathcal{K}_{\text{dA}} + \mathcal{K}_{\text{dB}} + \text{c.c.})E_0^2}$$

$$\mathcal{K}_{\text{o}} = \frac{e^2}{m^2\omega^2}\sum_{n\neq m;l} M_{mn} \frac{\frac{p^+_{nl}p^-_{lm}(f_m - f_l)}{E_l - E_m + i\hbar\Gamma_{lm} - \hbar\omega} - \frac{p^-_{nl}p^+_{lm}(f_l - f_n)}{E_n - E_l + i\hbar\Gamma_{nl} - \hbar\omega}}{E_n - E_m + i\hbar\Gamma_{nm}},$$

$$\mathcal{K}_{\text{dA}} = \frac{e^2}{m^2\omega^2}\sum_{nl} M_{nn}\left[\frac{p^+_{nl}p^-_{ln}(f_l - f_n)}{(E_l - E_n + i\hbar\Gamma_{ln} - \hbar\omega)^2} + \frac{p^-_{nl}p^+_{ln}(f_l - f_n)}{(E_n - E_l + i\hbar\Gamma_{nl} - \hbar\omega)^2}\right],$$

$$\mathcal{K}_{\text{dB}} = \frac{e^2}{m^2\omega^2}\sum_{nl} \frac{M_{nn}}{\hbar\omega} \frac{p^+_{nl}p^+_{ln}(f_n - f_l)(i\hbar\Gamma_{ln} - \hbar\omega)}{(E_l - E_n)^2 + (\hbar\Gamma_{ln} + i\hbar\omega)^2}.$$

Requires the quantum states in the solid, *n*, *m* and band energies

Ab initio, relativistic DFT, ASW code

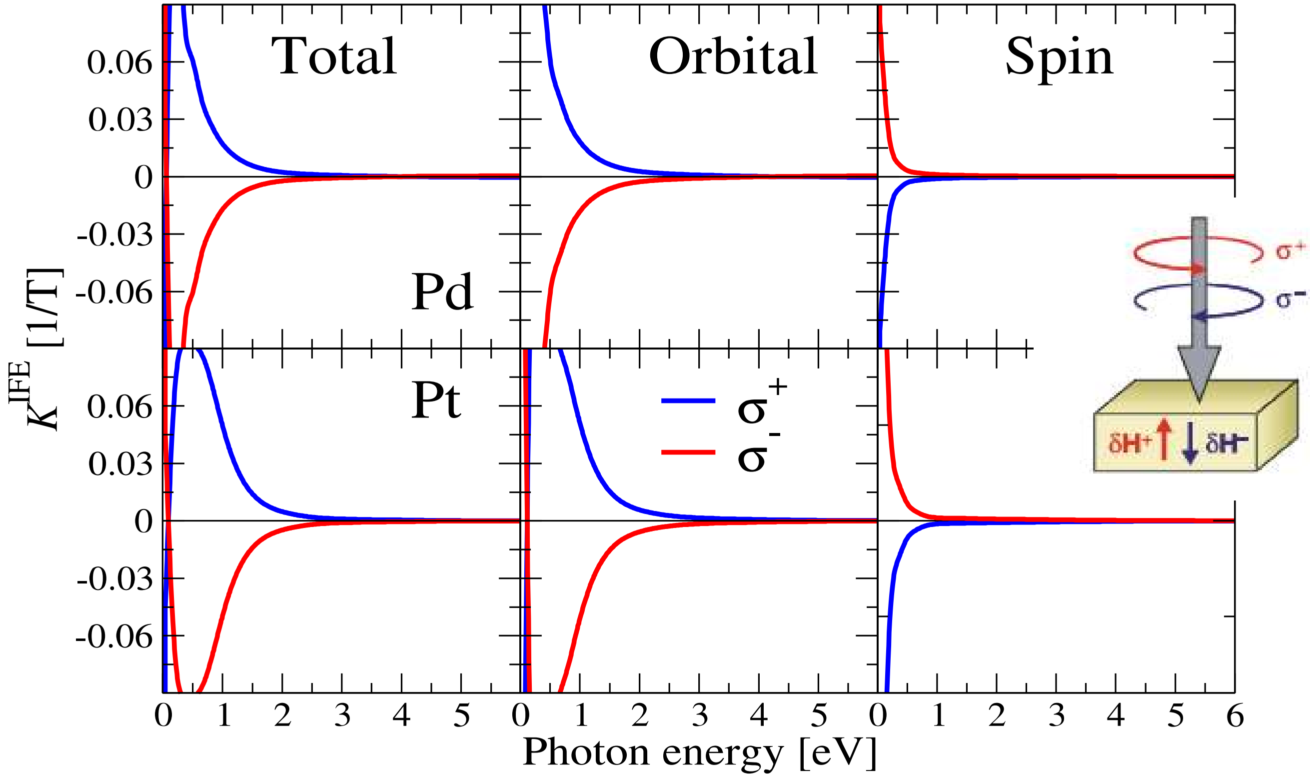
$$\vec{M} = \vec{L} + 2\vec{S}$$

Contains electronic Raman and Rayleigh scattering

Battiato, Barbalinardo & Oppeneer, Phys. Rev. B **89**, 014413 (2014)

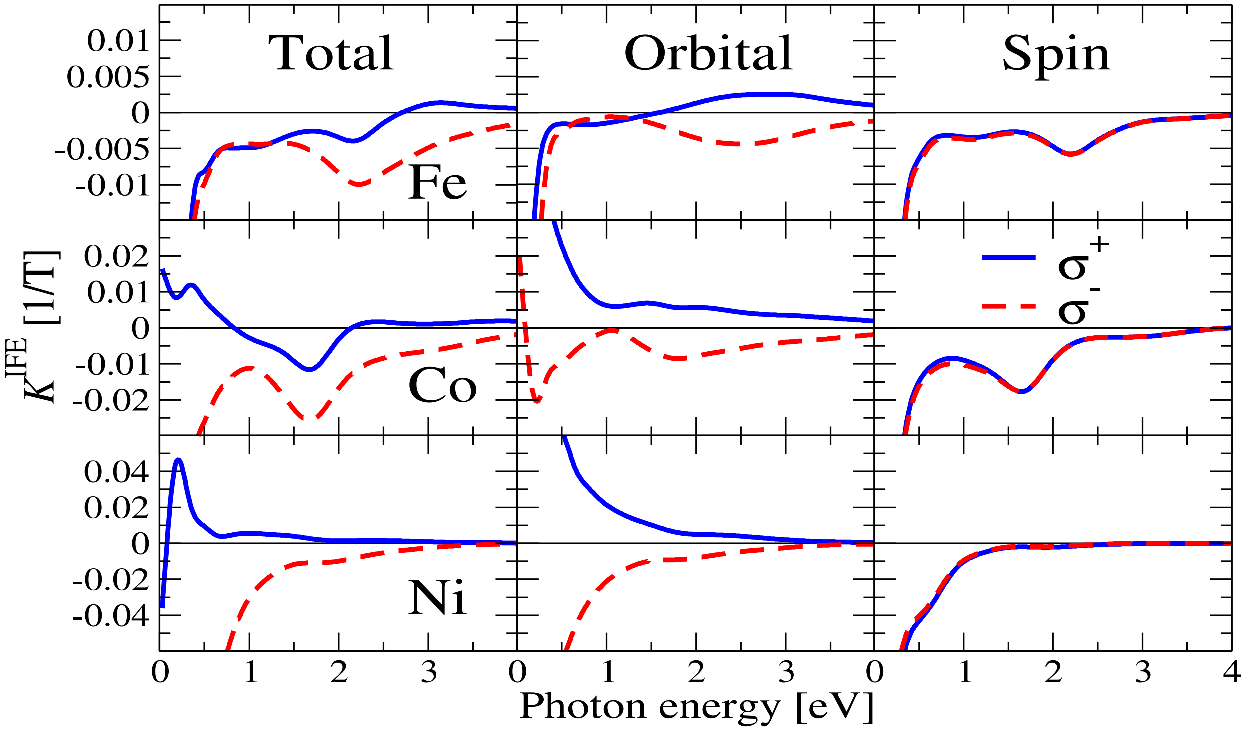
Nonmagnetic metals

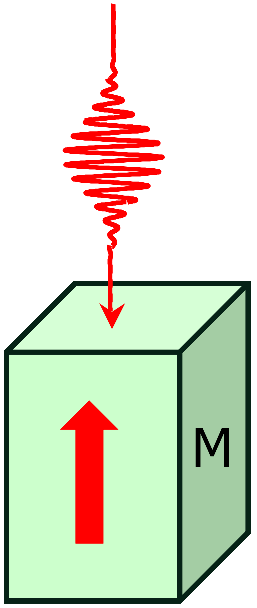
$$M_{ind}(\omega)=[K_L^{IFE}(\omega)+K_S^{IFE}(\omega)]I/c\ ,\quad I=\varepsilon_0 c E_0^2/2 \qquad (\hbar\Gamma=0.03\ Ry)$$



- *Antisymmetric* in helicity for nonmagnetic materials
- Opposite effect of spin and orbital IFE contributions
- Large orbital contribution in heavy metals as Pt (large SOC)

Elemental ferromagnets





- Spin contribution *not* antisymmetric in helicity
- Total IFE is *asymmetric* for ferro (ferri-)magnets

Berritta, Mondal, Carva & Oppeneer, Phys. Rev. Lett. **117**, 137203 (2016)

Freimuth, Blügel & Mokrousov, PRB **94**, 144432 (2016)

What is the laser-induced optomagnetic field ?!

$$M_{ind}(\omega)=K_{IFE}(\omega)I/c \quad , \quad I=\varepsilon_0 c E_0^2/2$$

($h\nu$ = 1.55 eV, I = 1.0 GW/cm^2)

Material	Optomagn. field
Ni	-30 T
Fe	-50 T
Co	-100 T
Cu	±100 T
Pd	±2 T
Pt	±30 T
Au	±300 T
FePt	-300 T

← Calc. Zeeman field needed for $|M_{ind}|$

Longitudinal configuration

} Larger for heavier elements

- Optomagnetic field is material & $h\nu$ dependent
- Larger for materials with large spin-orbit interaction
- Can give a sizable contribution to all-optical switching
- Prediction/design of suitable materials possible

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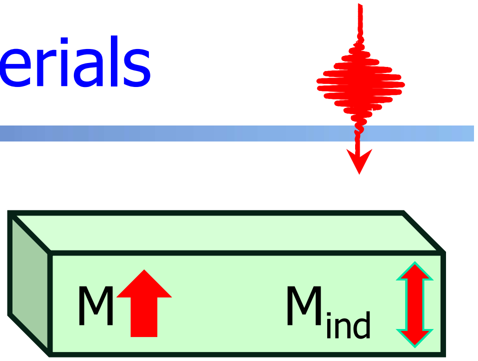
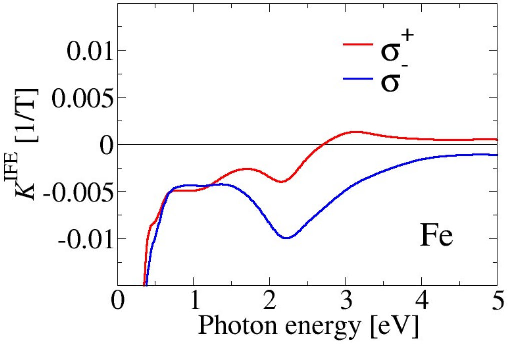
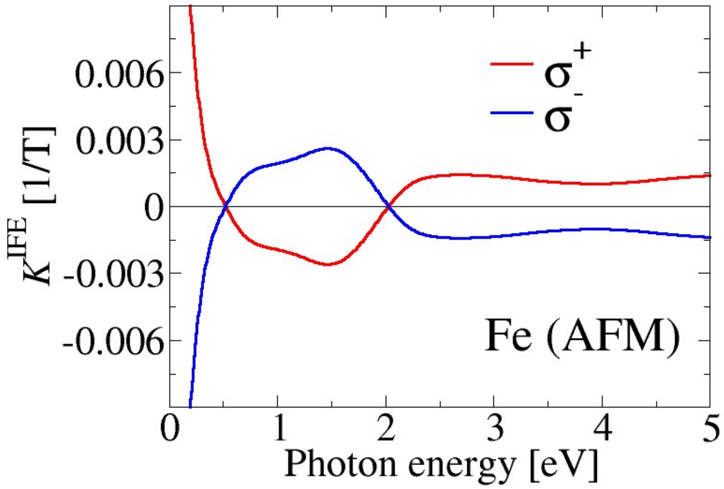
Antiferromagnetic materials

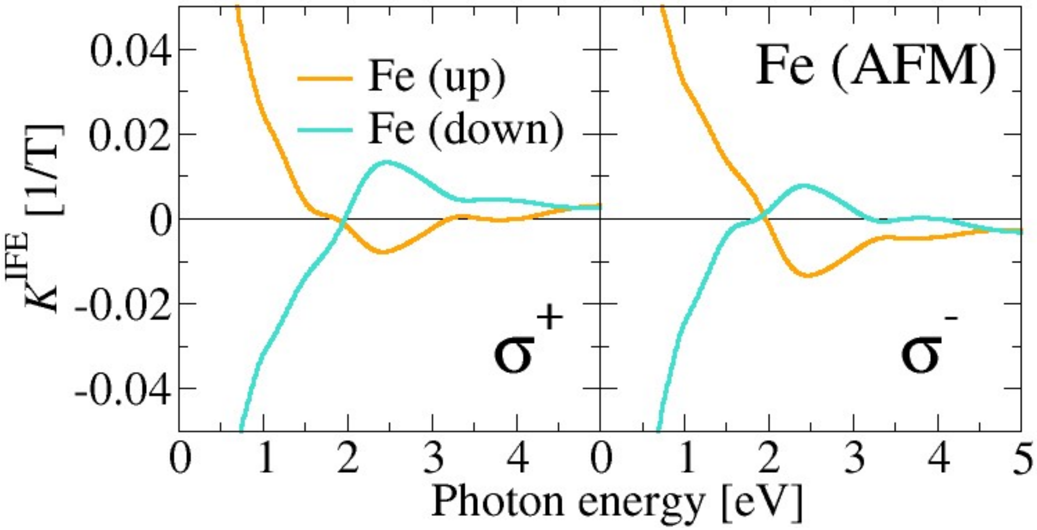






$$M^{Fe} = \pm 1.635 \mu_B$$

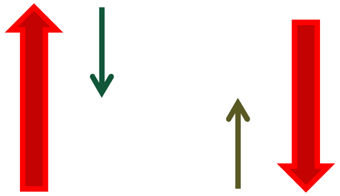
Small effect, antisymmetric?



I=1 GW/cm² ω=2.5 eV

$$M_{ind}^{\Uparrow}(\sigma^+) = -6.4 \times 10^{-4} \mu_B$$

$$M_{ind}^{\Downarrow}(\sigma^+) = 1.1 \times 10^{-3} \mu_B$$

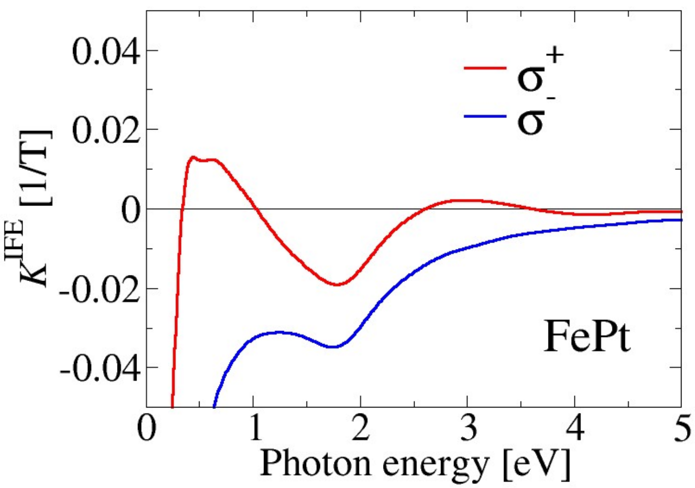


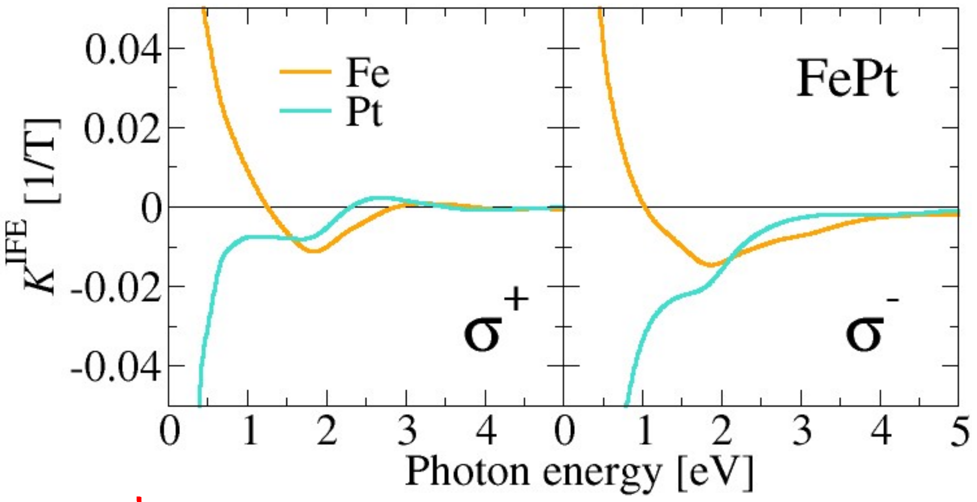
$$\vec{M}^{ind} \propto v_{IFE} \cdot E_i E_j^*$$

- Staggered induced magnetization

FePt in longitudinal configuration

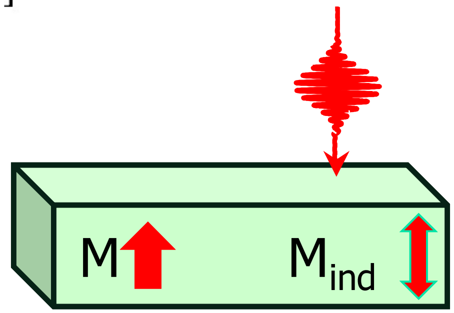
$$M_{ind}(\omega)=[K_L^{IFE}(\omega)+K_S^{IFE}(\omega)]I/c \quad , \quad I=\varepsilon_0 c E_0^2/2 \qquad (\hbar\Gamma=0.03\ Ry)$$





$$M^{Fe}=2.84\mu_B$$

$$M^{Pt}=0.41\mu_B$$



I=1 GW/cm^2
ω=1.55 eV

$$M_{ind}^{\sigma^+}=-1.5m\mu_B$$

$$M_{ind}^{\sigma_z^-}=-3.3m\mu_B$$

Ferromagnet: asymmetric in helicity

FePt: large contribution on Pt, related to large SOC

Multiscale modeling approach for FePt

Ab initio calculations

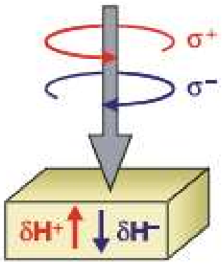
C_e, $n\pm$, induced magn.

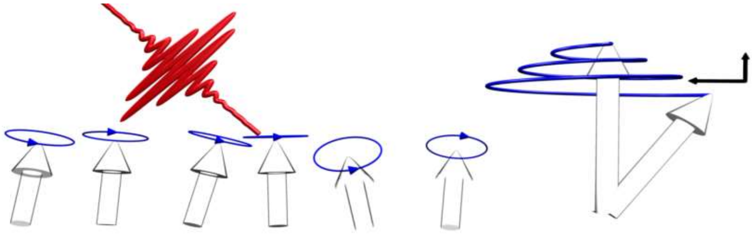
Micromagnetic model - LLB

Rate model

Transition probabilities

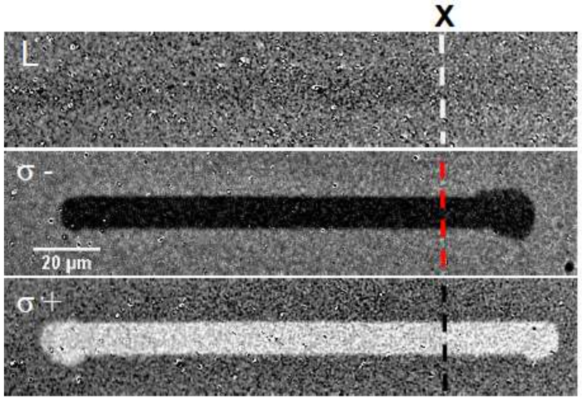
Experiment





Relativistic DFT (LSDA)

$$\frac{d\mathbf{m}}{dt} = -\gamma \mathbf{m} \times \mathbf{H}_{eff} + \gamma \alpha_{\|} \frac{\mathbf{m} \cdot \mathbf{H}_{eff}}{m^2} \mathbf{m} - \gamma \alpha_{\perp} \frac{\mathbf{m} \times (\mathbf{m} \times \mathbf{H}_{eff})}{m^2}$$

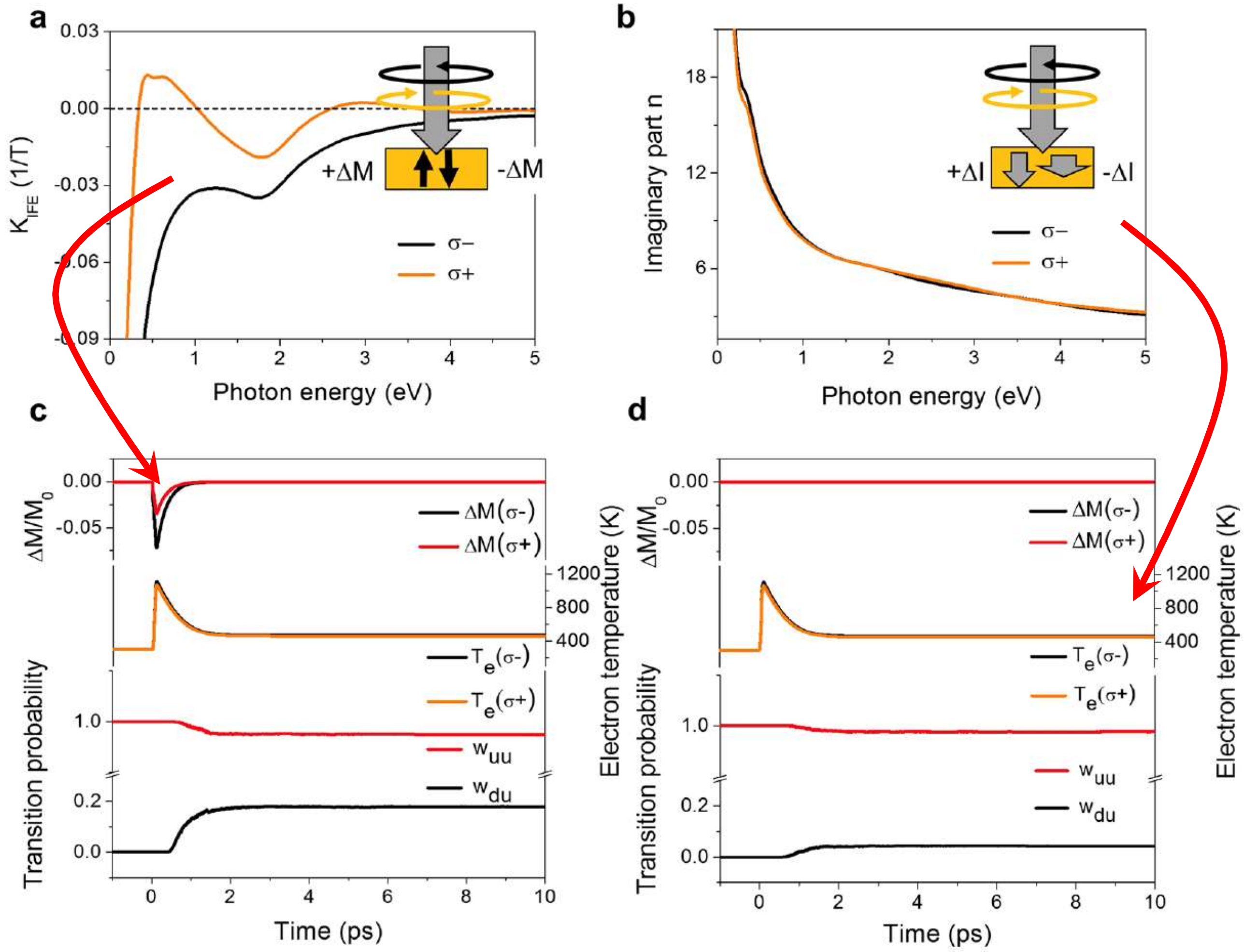


(with U. Nowak, O. Chubykalo-Fesenko, M. Münzenberg)

Magnetic recording material FePt

Helicity-dep. induced magnetization

Increased electron temperature T_e



Inverse Faraday effect:

$\Delta M = -7.1\%\ M_s\ (\sigma+)$ and $-3.5\%\ (M_s)\ (\sigma -)$

MCD effect:

$\Delta T = 32$ K (for $T_{e,max} = 1100$K)

for spin up and spin down

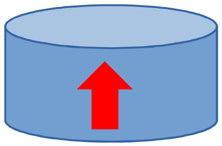
Two-temperature model

energy dissipation to lattice

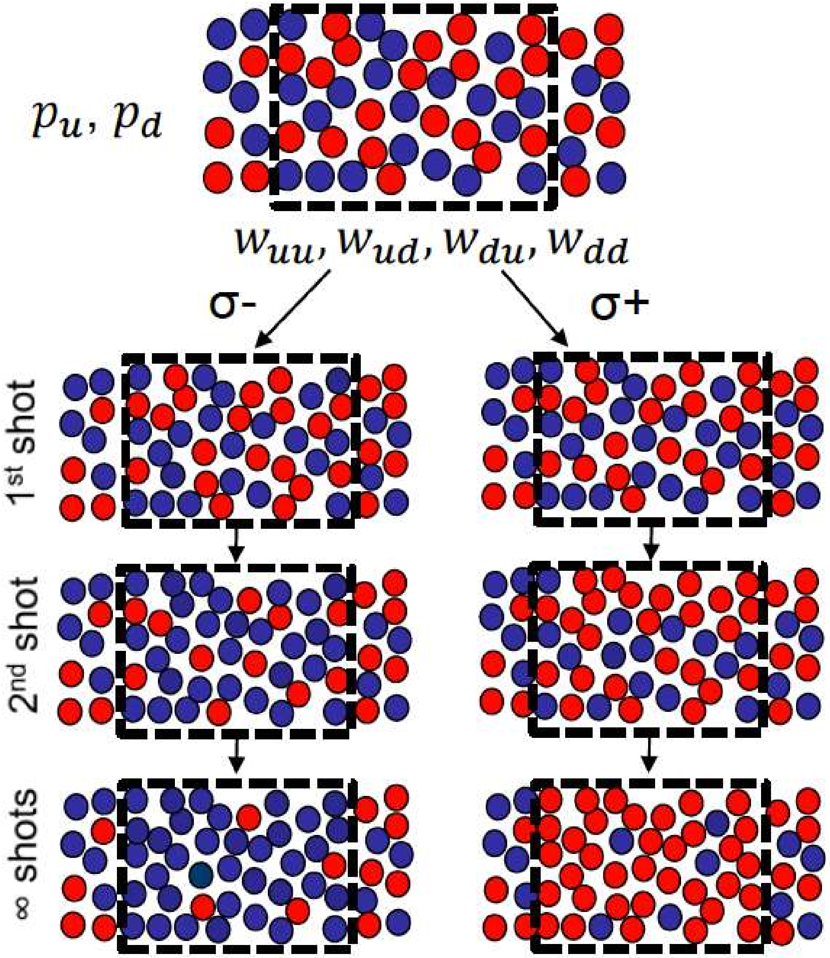
Transition probabilities:

$W_{uu}, W_{du}, W_{dd}, W_{ud}$

And: *stochastic rate model*



Stochastic rate model for repeated laser shots



$$\begin{aligned} p_u^{i+1} &= p_u^i w_{uu} + p_d^i w_{du} \\ &= w_{du} + p_d^i (w_{uu} - w_{du}) \end{aligned}$$

$$(p_u + p_d = 1)$$

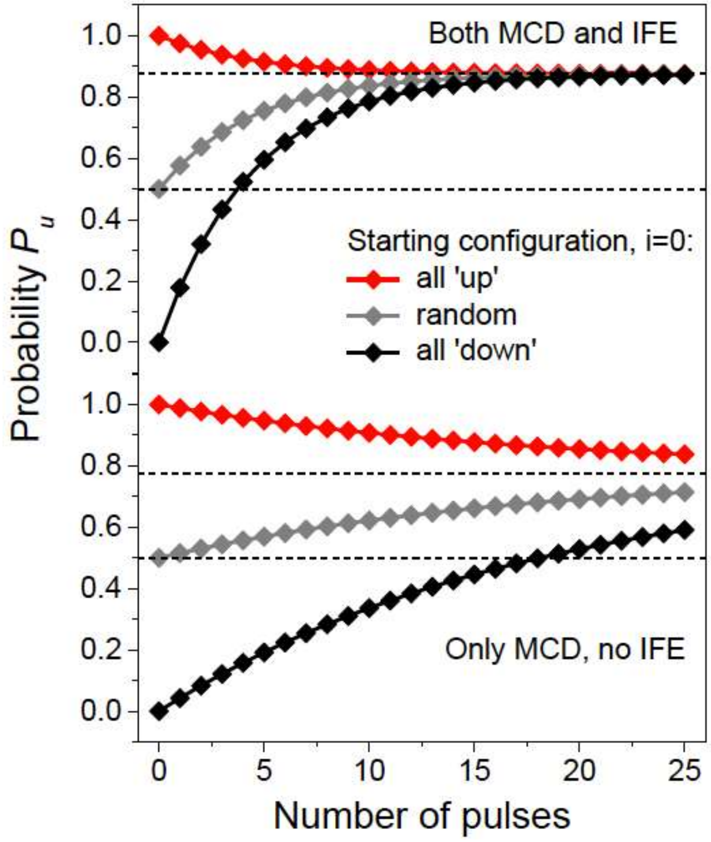
$$p_u(n \to \infty) = w_{du} \frac{-1}{w_{uu} - w_{du} - 1} = \frac{w_{du}}{w_{ud} + w_{du}}$$

High T_e , no asymmetry:
$w_{uu} = w_{ud} = 0.5 \Rightarrow p_u = 0.5$

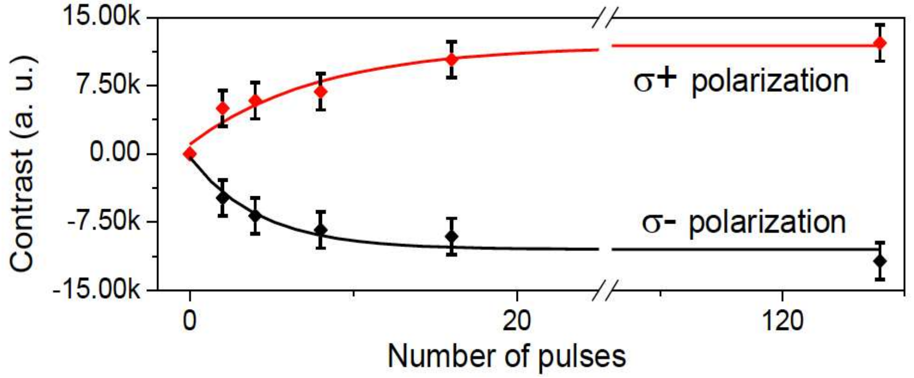
IFE: asymmetric, e.g. $w_{uu} > w_{ud}$

With calculated transition probabilities

Simulation



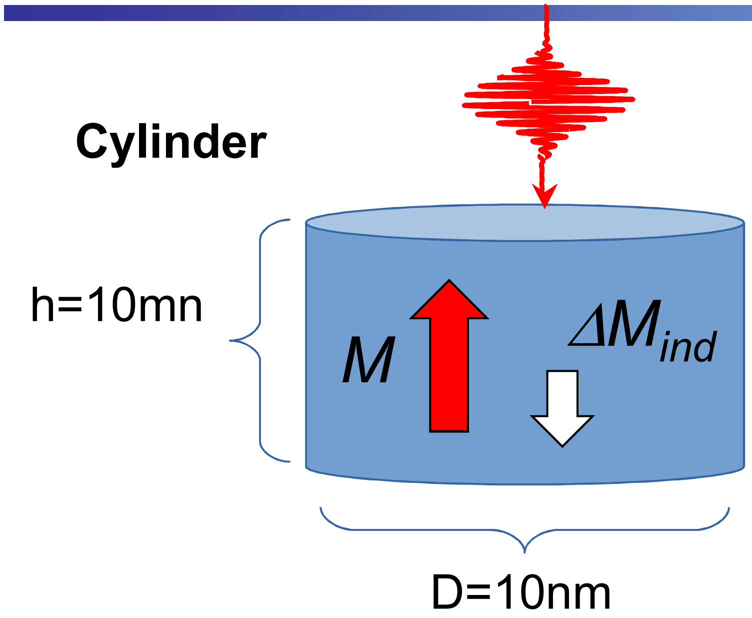
Experiment



- Only heating by helicity-dep. absorption does not give fast switching (not deterministic)
- IFE can provide switching with few laser pulses and possibly with single pulse

John, Berritta, Hinzke, Nowak, Chubykalo-Fesenko, Oppeneer, McCord, Münzenberg et al, Sci. Rep. **7**, 4114 (2017)

All-optical switching of Fe nanoparticle



Pulse duration	30	fs
Pump fluence	5-10	mJ/cm^2
C_e/T	222	J/(m^{3}K^2)
C_{ph}	2.3e6	J/(m^{3}K)
$G_{e\text{-}ph}$	6.6e17	J/(m^{3}sK)

Material	Fe	
Magnetic moment	2.22	μ_B
Exchange (J)	7.05e-21	J
Anisotropy (K)	5.65e-25	J

Atomistic Heisenberg spin Hamiltonian & LLG simulations:

$$H = -\frac{1}{2}\sum\sum J_{ij}\vec{s_i}\vec{s_j} - \sum K_i(\vec{s_i}\cdot\vec{n_i})^2 - \sum \mu_i\vec{B}\cdot\vec{s_i}$$

with York VAMPIRE code (S. Ruta, R. Chantrell)

(calc. by M. Berritta, J. Hurst)

Atomistic LLG simulations

Atomistic spin dynamics simulations – Landau-Lifshitz- Gilbert EOM

$$\frac{\partial s_i}{\partial t} = -\frac{\gamma_i}{1+\lambda_i^2}\left[\vec{s}_i \times \vec{H}_i^{eff} + \lambda \vec{s}_i \times \left(\vec{s}_i \times \vec{H}_i^{eff}\right)\right]$$

$$\lambda - microscopic\ damping$$

$$\gamma - gyromagnetic\ ratio$$

Effective field

$$\vec{H}^{eff} = -\frac{1}{\mu_i}\frac{\partial H}{\partial \vec{s}_i} + \vec{H}^{th}$$

Coupling to thermal (Langevin) bath:

$$\langle H^{th}_{i,\alpha}(t) H^{th}_{j,\beta}(t')\rangle = \frac{2\lambda_i k_b T \mu_i}{\gamma_i}\delta_{ij}\delta_{\alpha\beta}(t-t')$$

$$\alpha, \beta = x, y, z$$

2 temp.model

$$C_e\frac{dT_e}{dt} = -G_{el}(T_e - T_l) + P(t)$$

$$C_l\frac{dT_l}{dt} = G_{el}(T_e - T_l)$$

pulse length

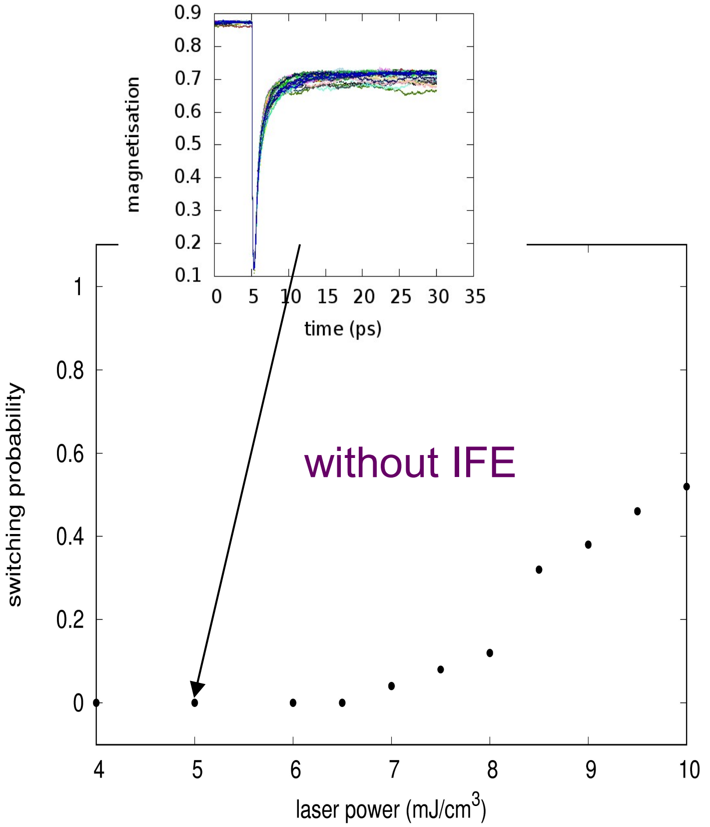
$$\Delta M_{ind} = K^{IFE}(\omega)\frac{I(t)}{c}$$

$$I(t) = \begin{cases} B_{pa} I_{max} exp\left[-2.77\left(\frac{t-t_0}{\tau_{pulse}}\right)^2\right] & for\ t < t_0 \\ B_{pa} I_{max} exp\left[-2.77\left(\frac{t-t_0}{\tau_{IFE}}\right)^2\right] & for\ t > t_0 \end{cases}$$

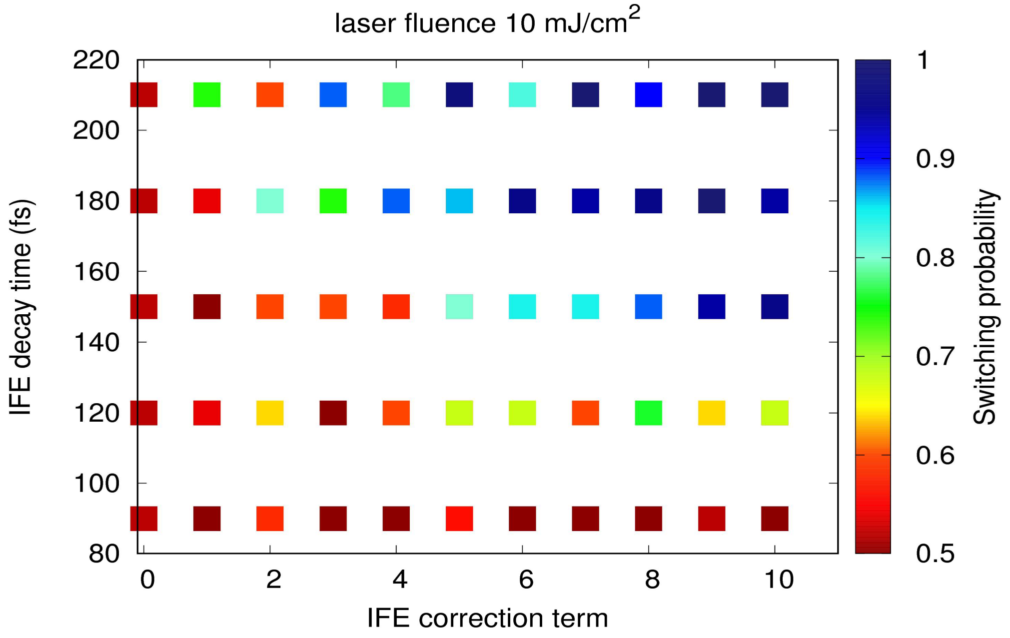
B_{pa} is a correction factor due to plasmonic antenna;
t_0 is the time corresponding to the peak of laser pulse.

strength

Switching probability with / without IFE



with IFE



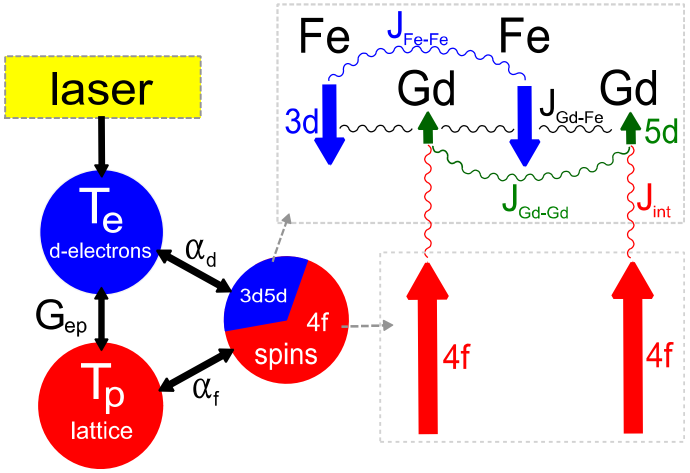
➡ Max 50% switching probability due to heating

➡ High switching probability with high IFE correction term and large IFE decay time

- ➢ Longer pulse length helps IFE assisted switching

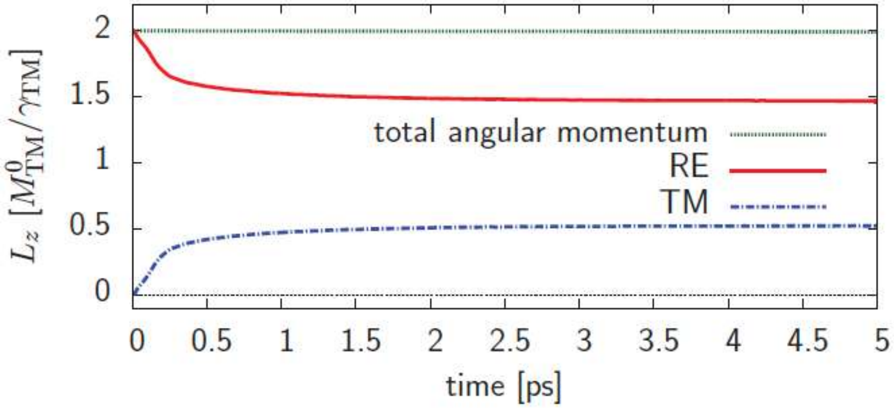
Hel.-independent *thermal* switching in GdFeCo

Distinguish 4f and 5d spins/orbitals

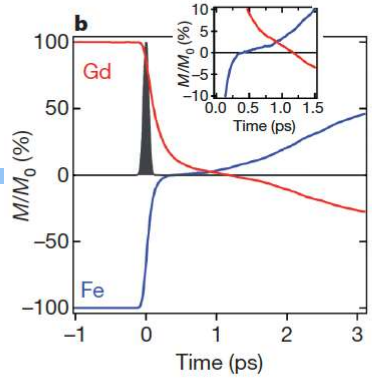


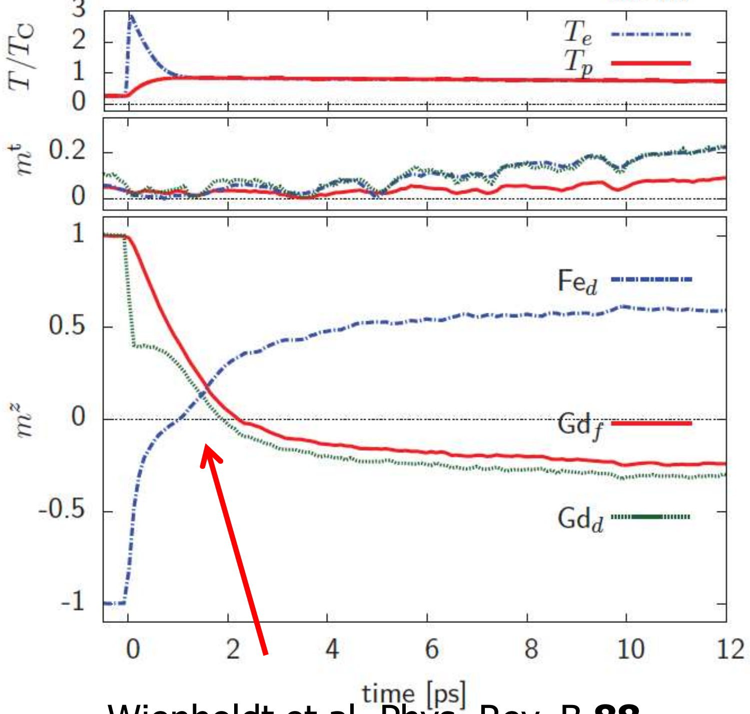
$$\mathcal{H} = -\sum_{\langle ij\rangle} \frac{J_{ij}}{2} \mathbf{S}_i \cdot \mathbf{S}_j - \sum_{i\in \mathrm{Gd}} J_{\mathrm{int}} \mathbf{S}_i \cdot \mathbf{S}'_i - d_z \sum_i (S_i^z)^2$$

Large 5d-4f J ≈ 130 meV



Radu et al,
Nature **472**,
205 (2011)





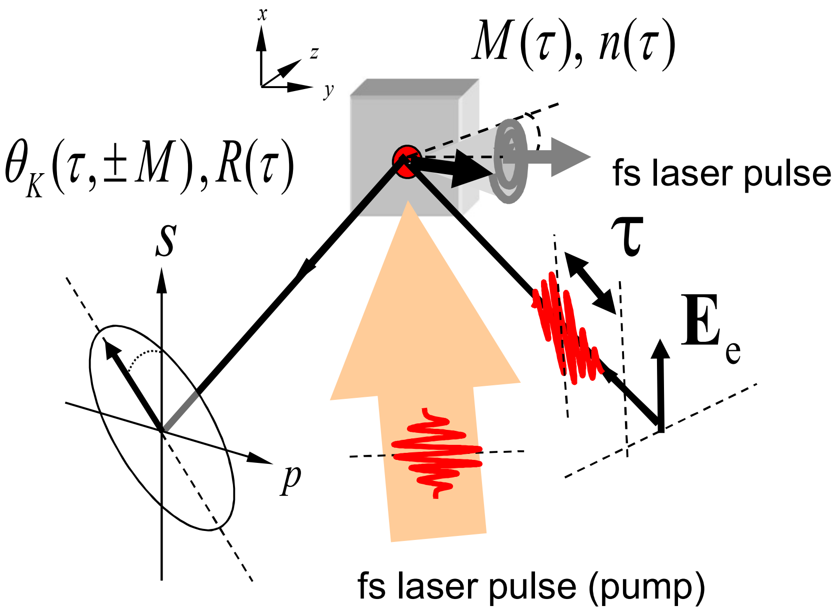
Wienholdt et al, Phys. Rev. B **88**,
020406R (2013)

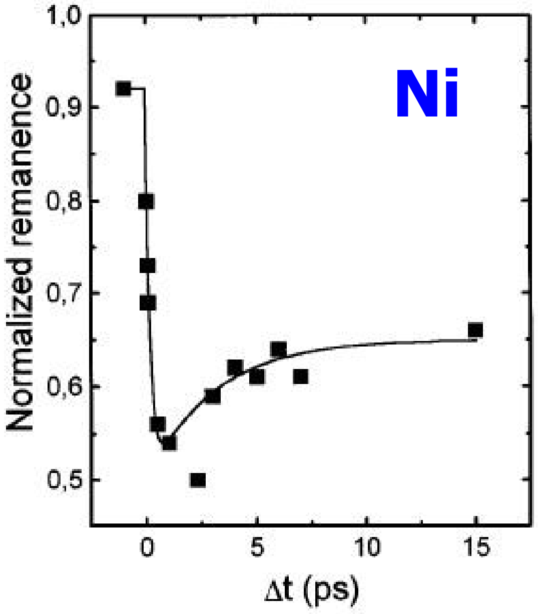
Angular momentum transfer
(dissipationless toy model)

cf. Bergeard et al, Nat. Comm. **5**, 3466 (2014)

Ultrafast laser-induced demagnetization

Surprising discovery, which lead to field of *ultrafast magnetism*





Beaurepaire, Merle, Danois &
Bigot, PRL **76**, 4250 (1996)

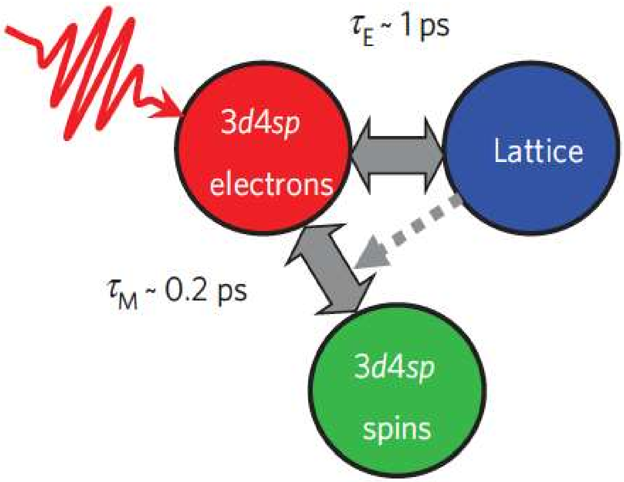
- quenching of the magnetization in <250 fs

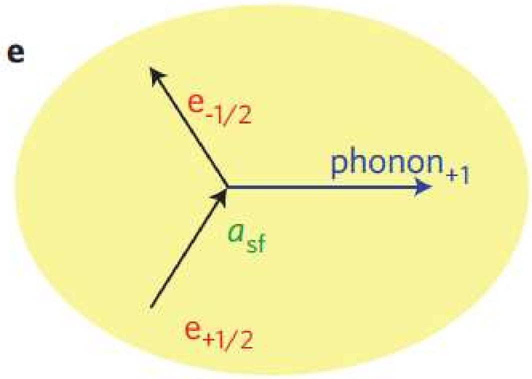
Eric Beaurepaire
†24th April, 2018

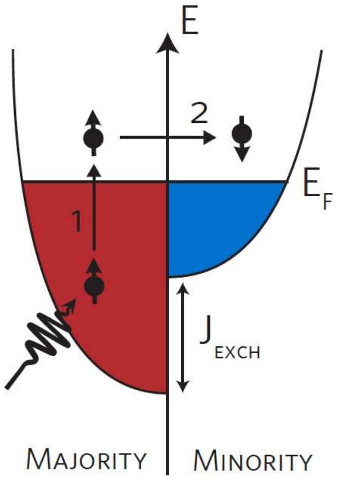
Jean-Yves Bigot
†2nd May, 2018

Elliott-Yafet with fast reduction of exchange splitting?

Koopmans et al, PRL **95** (2005), Koopmans et al, Nature Mater. **9**, 259 (2010)







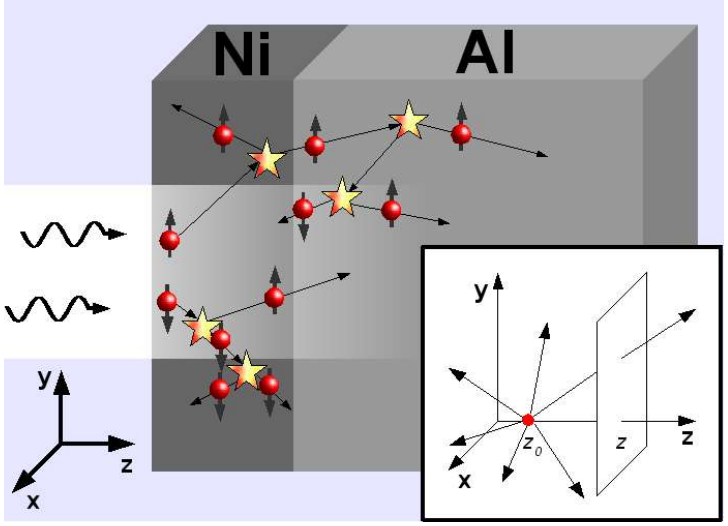
Energy transferred to lattice

Elliott-Yafet spin-flips, leading to dynamical quenching of exch. splitting

Schellekens & Koopmans, PRL **110**, 217204 (2013)

Mueller et al, PRL **111,** 167204 (2013) [μT- model]

Superdiffusion as mechanism of ultrafast demagnetization?



Superdiffusive spin-dynamics:

$$\frac{\partial n_\sigma^{tot}}{\partial t} + \frac{n_\sigma^{tot}}{\tau_\sigma} = \left(-\frac{\partial}{\partial z}\hat{\phi}_\sigma + \hat{I}\right)\left(\hat{S} n_\sigma^{tot} + S_\sigma^{ext}\right)$$

Change in e-density

Reaction term

Flux outflow

Source term

Battiato, Carva, Oppeneer, PRL **105**, 027203 (2010)

Distinct from ballistic and diffusive spin-transport

$$\left\langle r^2(t)\right\rangle \propto t^{2/\gamma}$$

Ballistic: γ=1

Diffusive: γ=2

Here: $1 \le \gamma \le 2 \quad and \quad \gamma = \gamma(t)$ “Superdiffusive regime”

Demagnetization through creation of an ultrafast spin current

Bergeard et al, PRL **117** (2016)
Razdolski et al, Nat. Comm. **8** (2017)

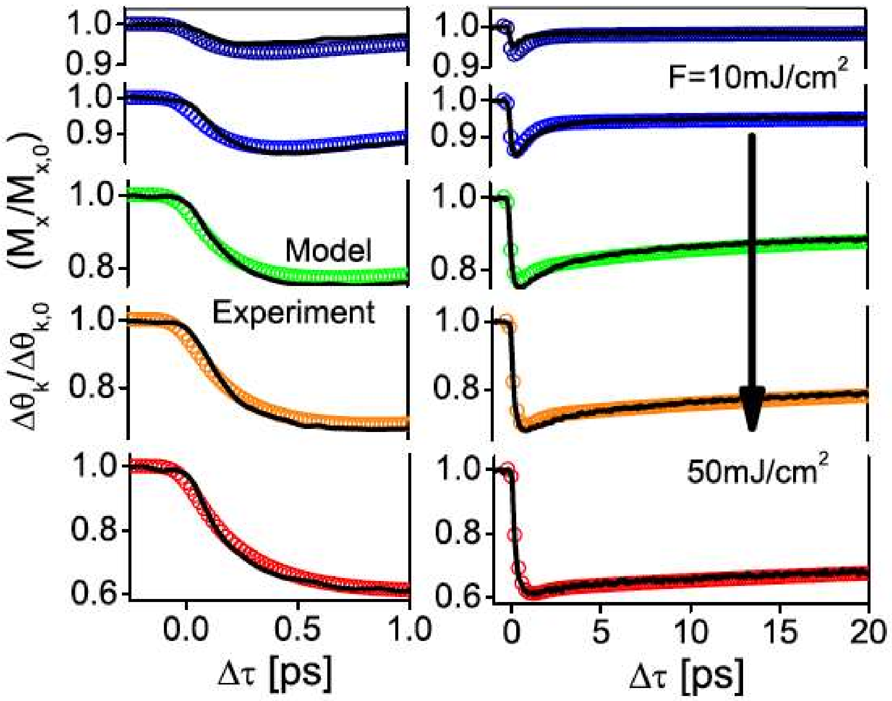
Rudolf et al, Nature Comm. **3**, 1037 (2012)
Kampfrath et al, Nature Nano. **8**, 256 (2013)
Eschenlohr et al, Nat. Mater. **12**, 332 (2013)

LLB simulations for demagnetization of Ni

Landau-Lifshitz-Bloch equation

$$\dot{\mathbf{m}} = \gamma[\mathbf{m}\times\mathbf{H}_{\mathrm{eff}}] + \frac{\gamma\alpha_{\parallel}}{m^2}[\mathbf{m}\cdot\mathbf{H}_{\mathrm{eff}}]\mathbf{m} - \frac{\gamma\alpha_{\perp}}{m^2}[\mathbf{m}\times(\mathbf{m}\times\mathbf{H}_{\mathrm{eff}})],$$

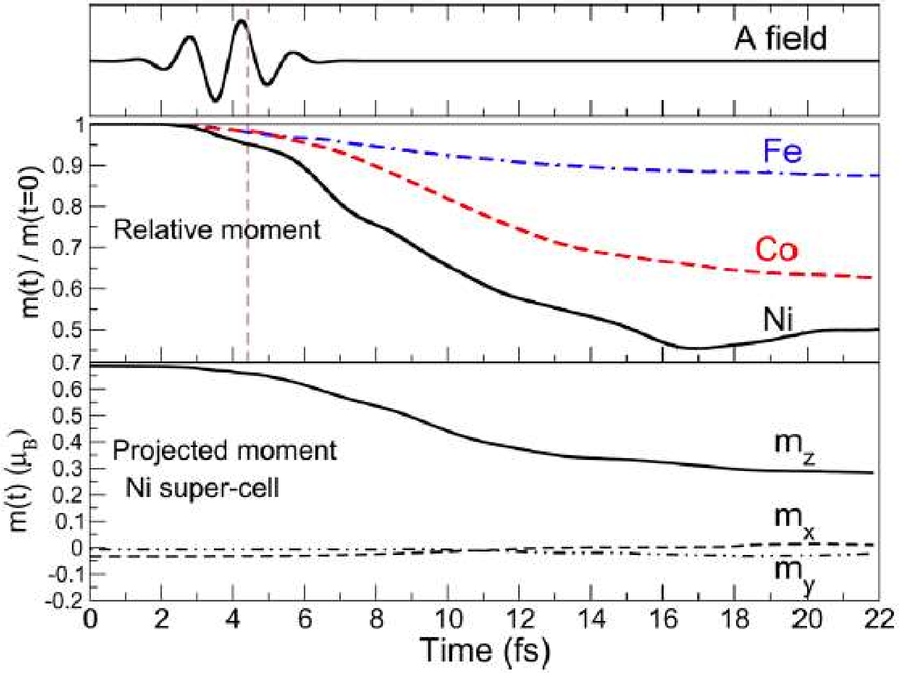
Spin dissipation rate in $\alpha_{||}$

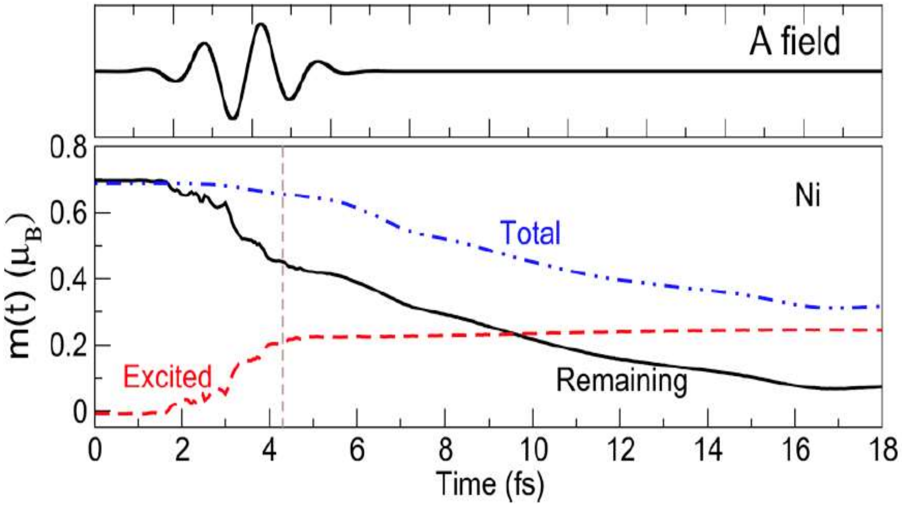


Atxitia et al, PRB **81**, 174401 (2010)

⇨ thermal long./trans. dissipation mechanism

Time-dependent DFT calculations





Very fast electronic quenching of m_z

Spin-orbit mediated long. spin flips during & after excitation, S -> L

Krieger, Dewhurst, Elliott, Sharma, & Gross, J. Chem. Theo. Comp. **11**, 4870 (2015)

Elliott, Krieger, Dewhurst, Sharma & Gross, New J. Phys. **18**, 013014 (2016)

(talk of Sangeeta Sharma)

Ab initio calculation of Elliott-Yafet demagnetization

Generalized energy dependent-spin-flip Eliashberg function

$$\alpha^2_{SF}F(E,\Omega) = \frac{1}{2M\Omega}\sum_{\nu,n,n'}\int dk \int dk' \; g^{\nu,\uparrow\downarrow}_{kn,k'n'}(q)\,\delta(\omega_{q\nu} - |\Omega|)\,\delta(E^{\uparrow}_{kn} - E)\,\delta(E^{\downarrow}_{k'n'} - E)$$

Spin-resolved transition rates: $S^{\sigma\sigma'} = \int_0^\infty d\Omega\; \alpha^2_{\sigma\sigma'}F(E,\Omega) f_\sigma(E)(1-f_{\sigma'}(E))(1+2N(\Omega))$

NEQ, FD

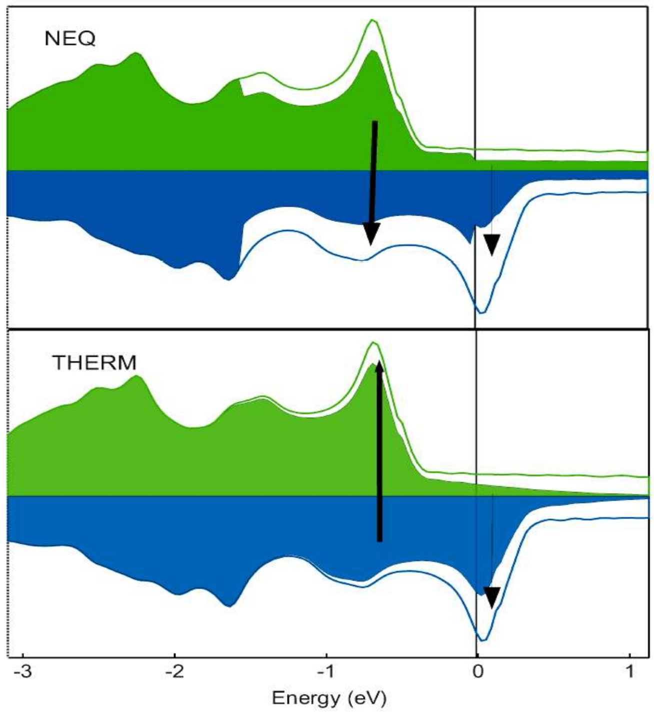
NEQ "deep" minority holes are created, gives more moment-reducing transitions

Faster demagnetization for NEQ state than for electron thermalized state

Carva, Battiato & Oppeneer, PRL **107**, 207201 (2011)

Essert & Schneider, PRB **84**, 224405 (2011)

Carva, Battiato, Legut & Oppeneer, PRB **87**, 184425 (2013)



Computed effective electron-phonon demagnetization

Total SF probability: $P_S = (S^- + S^+) / \sum_{\sigma\sigma'} S^{\sigma\sigma'}$

Demagnetization ratio: $D_S = (S^- - S^+) / \sum_{\sigma\sigma'} S^{\sigma\sigma'}$ $\longleftrightarrow$ $dM/dt = 2\mu_B (S^- - S^+)$

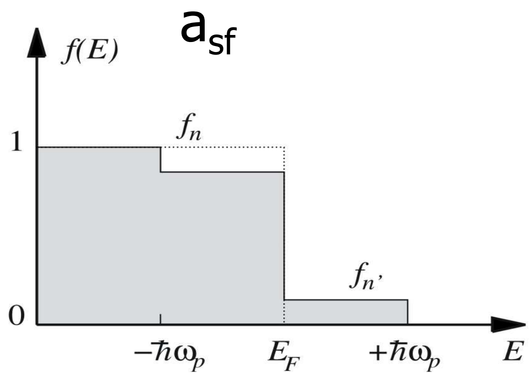
	$P_S^{b^2}$	P_S	D_S
Ni (low T)	0.07	0.04	0
Ni (T_e=1500K)	0.08	0.05	0.002
Ni (T_e=3000K)	0.11	0.07	0.003
Ni (T_e=5000K)	0.12	0.10	0.004
Ni (NEQ)	0.12	0.09	0.025

$\Delta M = 0.08 \mu_B / ps$

(~3% / 250fs)

$\Delta M = 0.1 \mu_B / 250\, fs$

(~17% / 250fs)

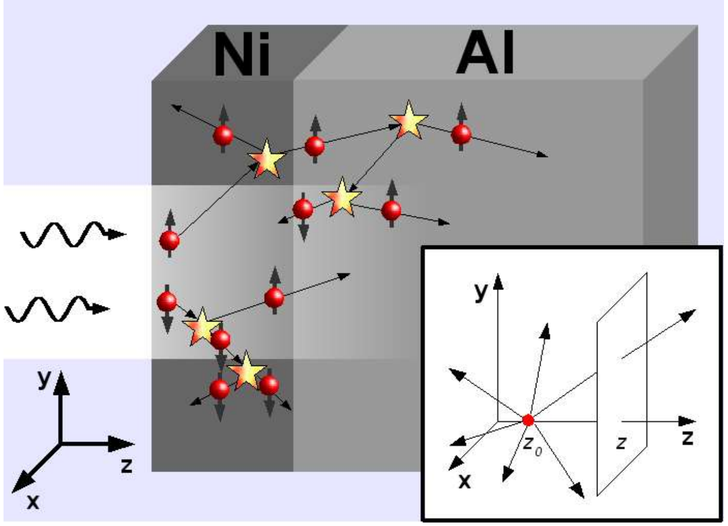


nonthermal

Large SF probability $P_s \neq$
large demagnetization $\sim D_s$

Carva, Battiato & Oppeneer,
PRL **107**, 207201 (2011)

Superdiffusion as mechanism of ultrafast demagnetization?



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Change in e-density | Reaction term | Flux outflow | Source term

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Rudolf et al, Nature Comm. **3**, 1037 (2012)
Kampfrath et al, Nature Nano. **8**, 256 (2013)
Eschenlohr et al, Nat. Mater. **12**, 332 (2013)

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Time-evolution of NEQ superdiffusive transport

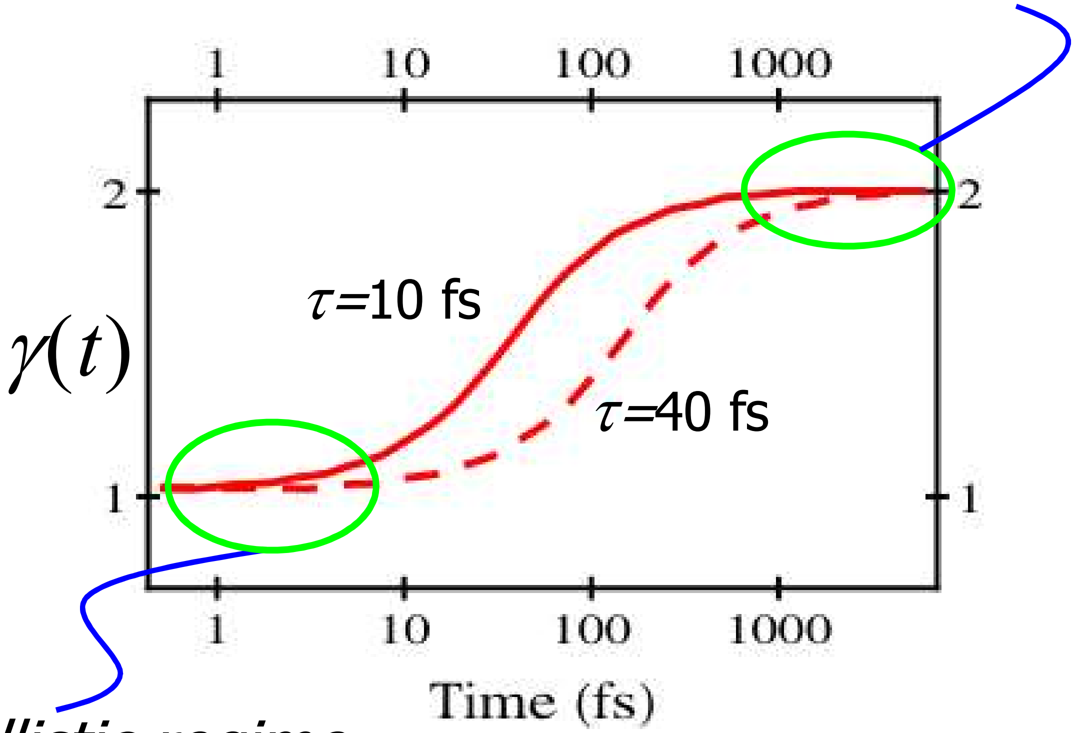
Average displacement

$$\left\langle r^2(t)\right\rangle = K_w t^{(2/\gamma)}$$

Anomalous diffusion exp. $\gamma(t)$

To compare to ballistic & diffusive transport: switch-off inelastic scattering (i.e. no thermalization)

Diffusive regime



Ballistic regime

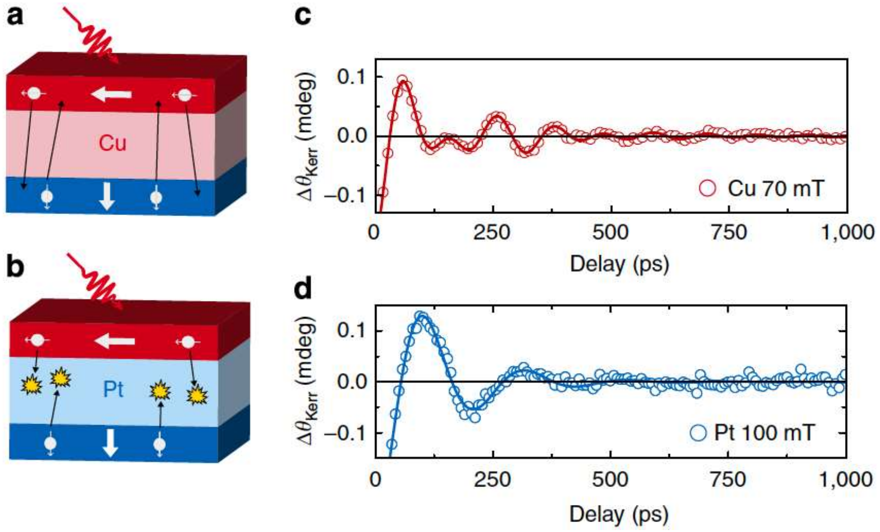
Only elastic lifetime τ= 10 or 40 fs

- Cross-over to diffusive hot e transport ~500 fs

Recent observations of spin transport

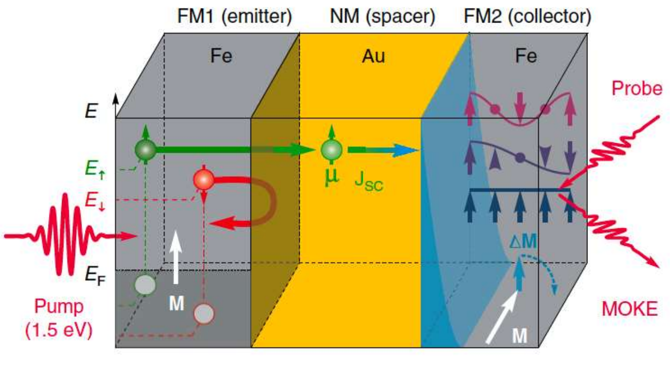
Ultrafast spin-transfer torque driven by fs laser excitation

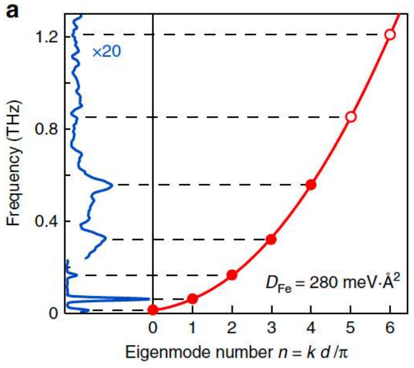
Schellekens et al, Nat. Commun. **5**, 4333 (2014)



Excitation of standing spin waves by STT-spin current

Razdolski et al, Nat. Commun. **8**, 15007 (2017)





Theory: Posters of K. Carva, U. Ritzmann

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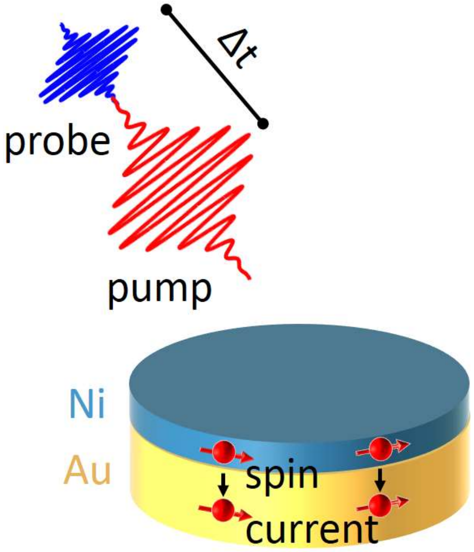
Quantifying superdiffusive spin currents in metals

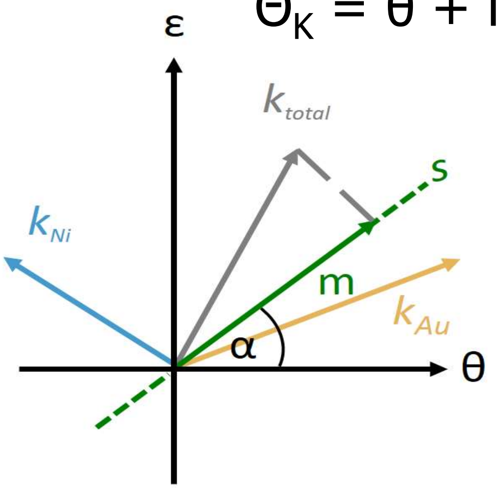
- How fast? How much?

10 nm Ni on 150 nm Au
Use *Complex MOKE* to detect spins in Au & Ni
Compare with *ab initio* & superdiffusive theory

Absence of SD currents suggested in other work!



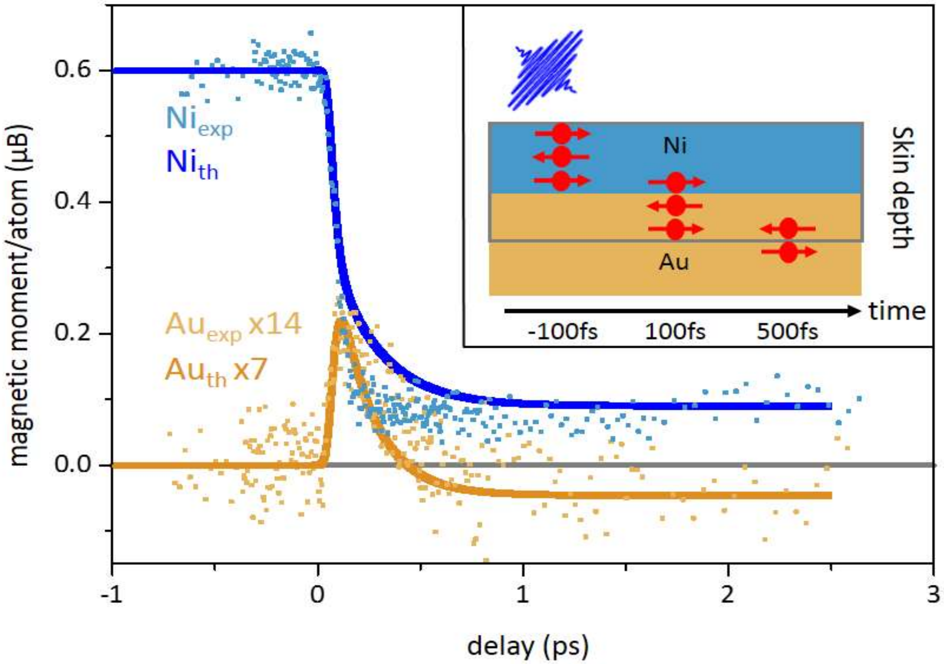
$\Theta_K = \theta + i\varepsilon$



can choose a detection axis *s* such that only Ni or Au is detected

With M. Aeschlimann (Kaiserslautern), S. Mathias (Göttingen)
B. Koopmans (Eindhoven)

Observation of very fast spin current



SD transport theory: full curves

Hofherr et al, PRB **96**, 100403R (2017)

- Very fast decay in Ni ~40 fs, fast spin injection in Au, ~42±35 fs
- Well explained by superdiffusive spin transport in Au
- But: only 0.25 μ_B/Au atom detected of 0.52 μ_B/Ni atom loss
- Spin flips in Ni and Ni/Au interface contribute to loss in Ni
- *Spin injection efficiency* ≥50%

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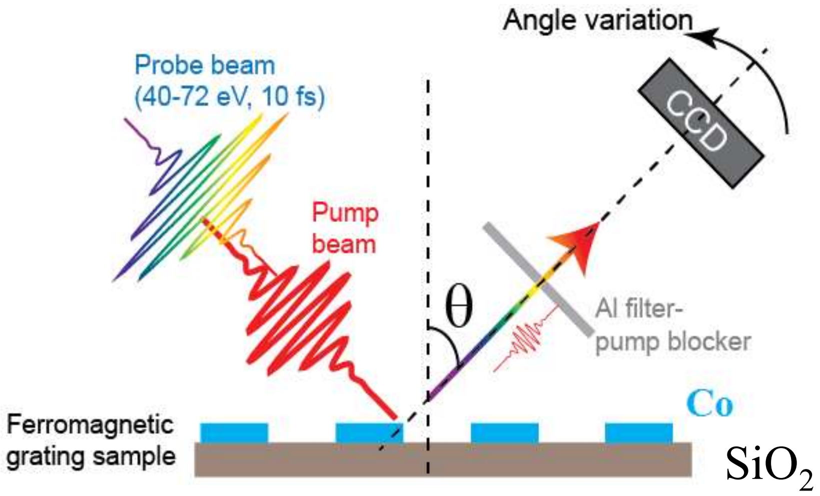
Ultrafast laser-induced demagnetization theories

Elliott-Yafet e-phonon scattering ?	Koopmans et al., PRL **95** (2005)
Spin-orbital interaction S ->L	Zhang & Hübner, PRL **85** (2000)
Fast magnon excitation ?	Carpene et al., PRB **78** (2008)
Coulomb-exchange e-e scattering ?	Krauss et al., PRB **80** (2009)
Laser induced (relativistic) spin-flips	Zhang et al., Nat.Phys. **5** (2009)
Chem. potential adjustment μT	Mueller et al., PRL **111** (2013)
Spin-orbit inter. + opt. excitation	Krieger et al., JCTC **11** (2015)
Fast S -> L -> lattice transfer	Töws & Pastor, PRL **115** (2015)
Superdiffusive spin transport	Battiato et al., PRL **105** (2010)

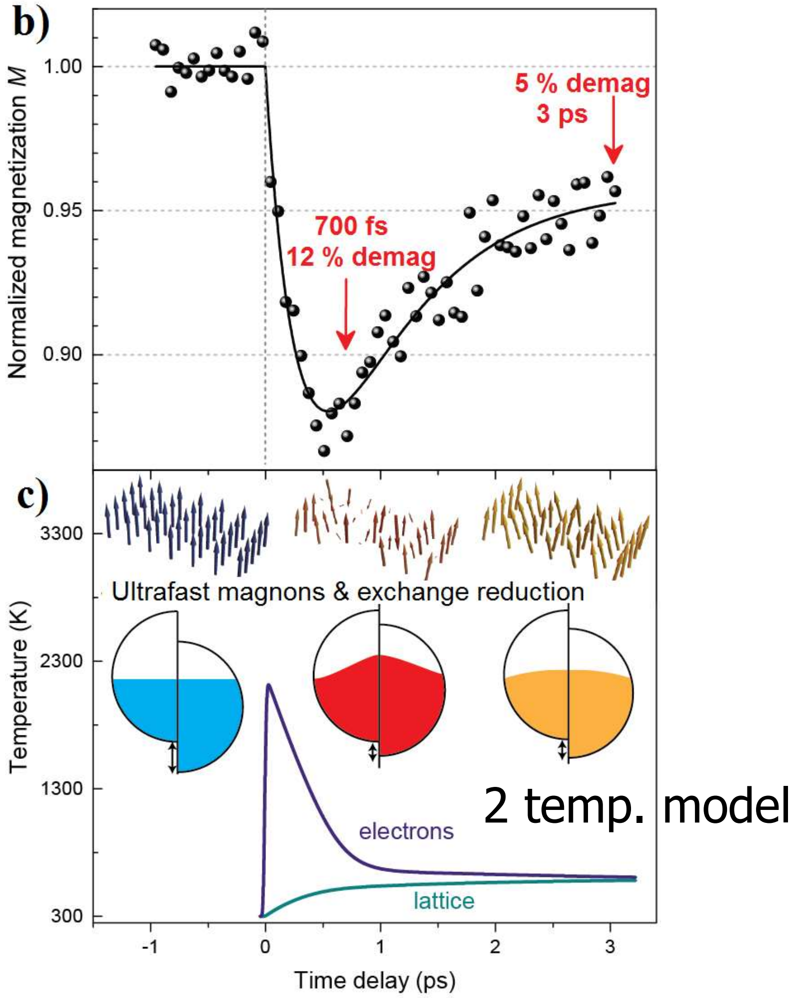
➔ scrutinize mechanisms with further experiments & calculations

Demagnetization of ferromagnet on an insulator?

Angle, energy and time-resolved XUV T-MOKE



- 10 nm Co / SiO_2
- **No** superdiffusion
- Electrons thermalized at ~400fs
- Thermal effects 700 fs & 3 ps
- Measure Co hcp M edge ~60 eV

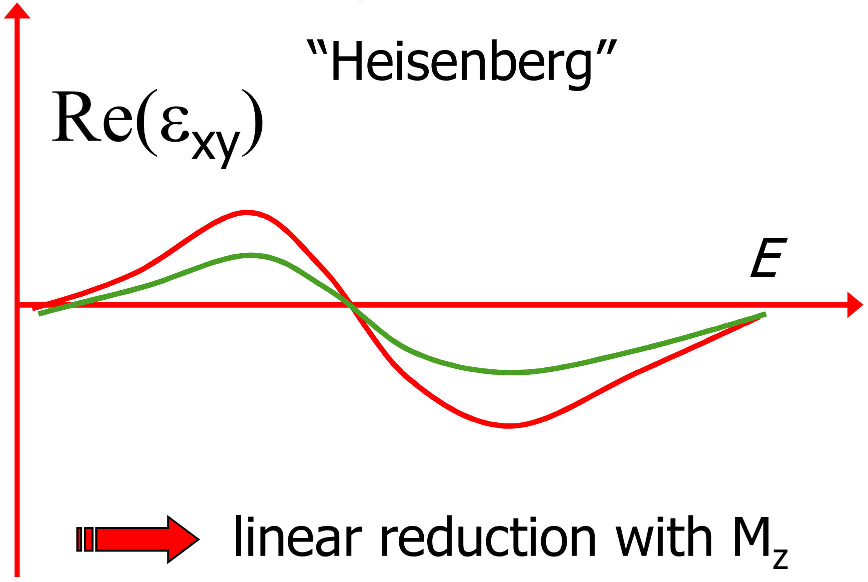


What is the main contribution to demagnetization?

With Boulder (Murnane), Kaiserslautern (Aeschlimann) groups

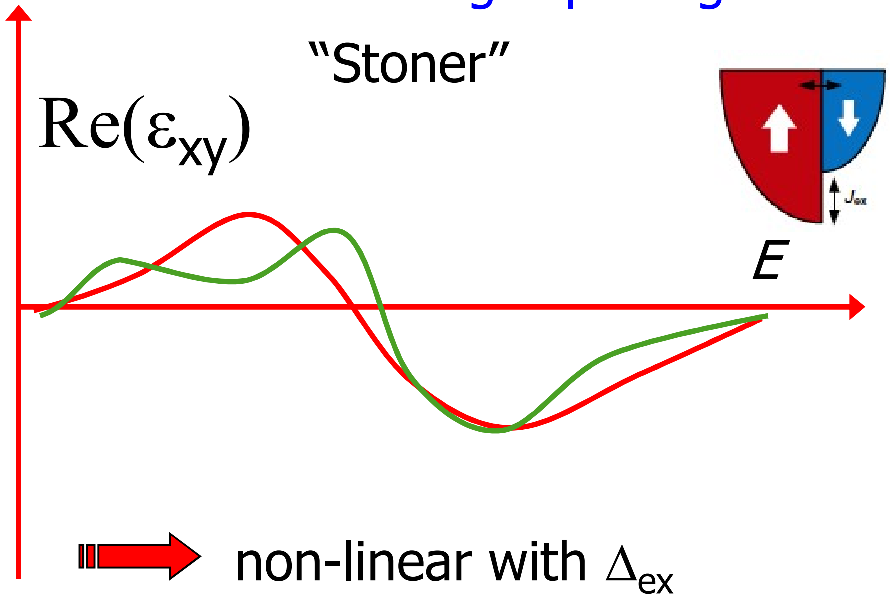
Magnon excitation vs. exchange reduction

Frozen spin wave



linear reduction with M_z

Reduced exchange splitting



non-linear with Δ_{ex}

Oppeneer et al, Z. Phys. B **88**, 309 (1992)

Can distinguish effects of these two mechanisms on MOKE

Predicted by Erskine & Stern, PRB **12**, 5016 (1975)

Determine possible demagnetization mechanisms

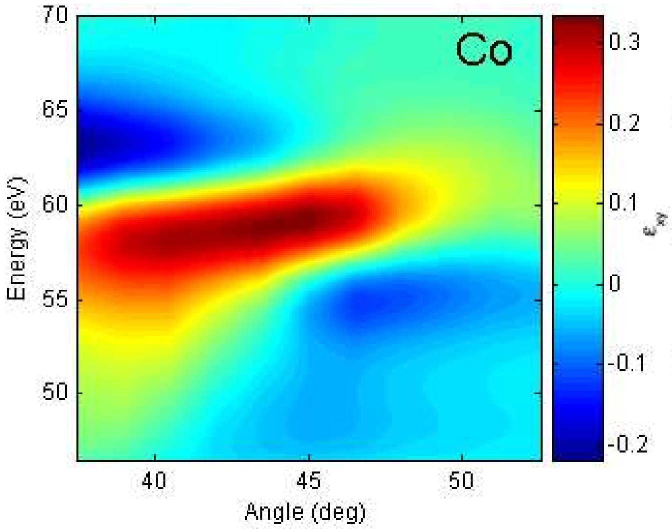
1. Measure whole *energy and angle* dependent T-MOKE asymmetry at 700 fs and 3 ps and determine change in A(t) wrt A(t=0)

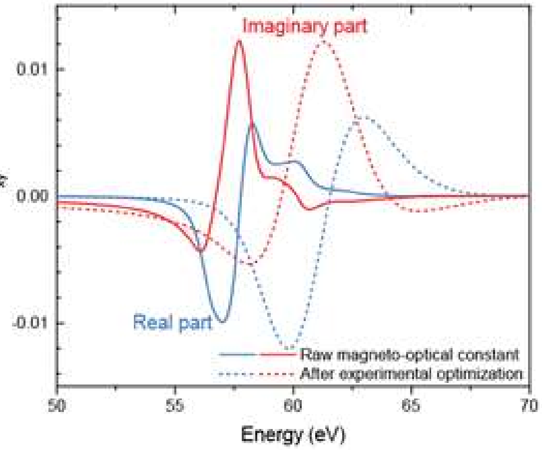
$$A = 2\,Re\left[\frac{\sin 2\theta_i\,\epsilon_{xy}}{n^4\cos\theta_i^2 - n^2 + \sin\theta_i^2}\right] = 2\,\mathrm{Re}[\mathrm{F}(\theta,n)\,\epsilon_{xy}] = 2\,Re[\mathrm{F}(\theta,n)]Re[\epsilon_{xy}] - 2Im[\mathrm{F}(\theta,n)]Im[\epsilon_{xy}]$$

2. Compute *ab initio* ε_{xy} for several cases: 1) frozen magnon excitations, 2) reduced exchange splitting (spin-flips), 3) increased electron temperature T_e - construct the change in A(t) wrt A(t=0) –> least square fit with experiment

$$A(t) = \frac{R(M+) - R(M-)}{R(M+) + R(M-)}$$

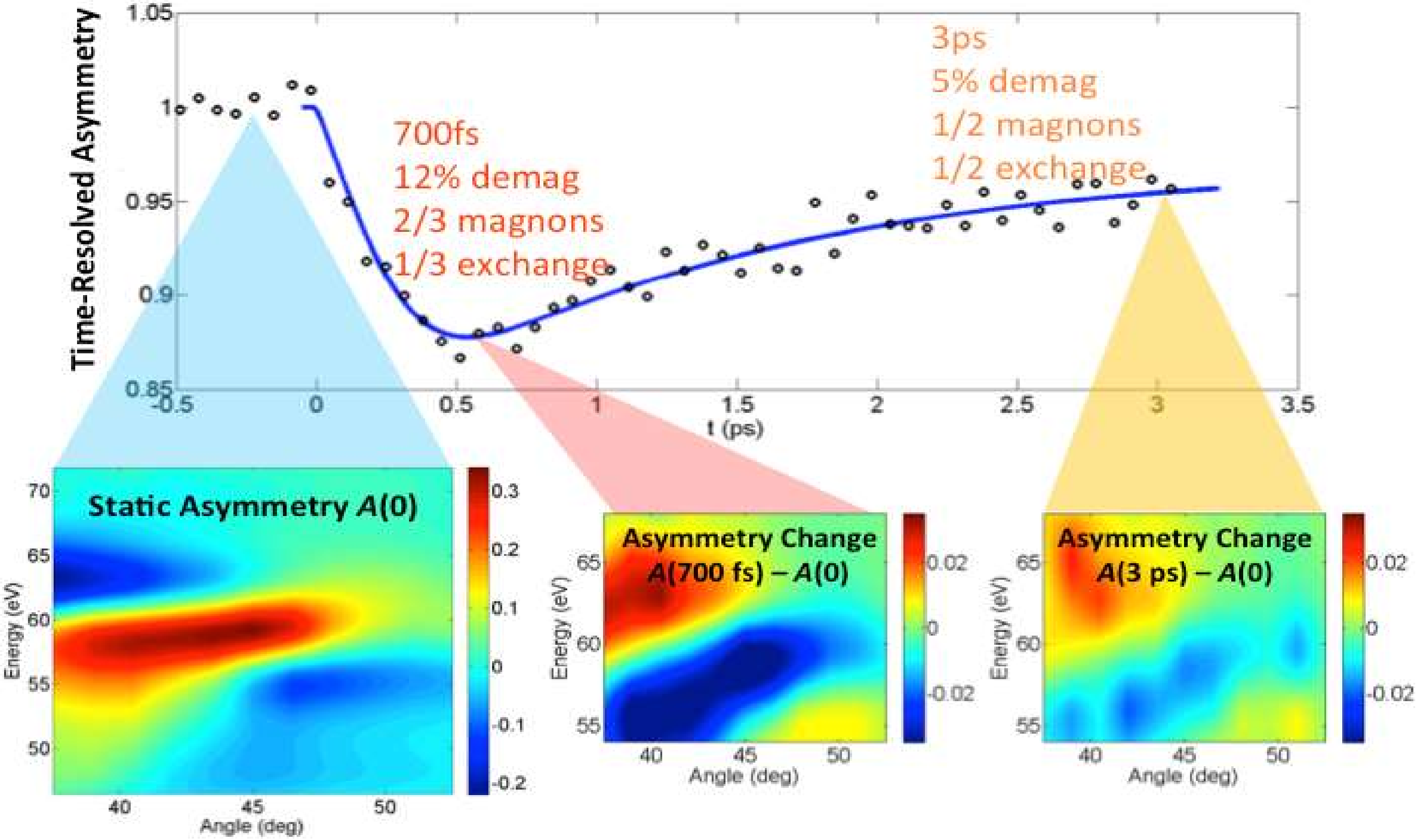
Asymmetry





Turgut et al, PRB **94**, 220408R (2016)

Results of time-, energy, and angle-resolved T-MOKE



For Co

- Surprisingly small contribution from spin-flips (exch. split reduction)
- Larger effect is due to fast magnon excitation => reduction of M_z

Turgut, Zusin, Legut, Carva, Legut, Oppeneer, Murnane et al, PRB **94**, 220408R (2016)

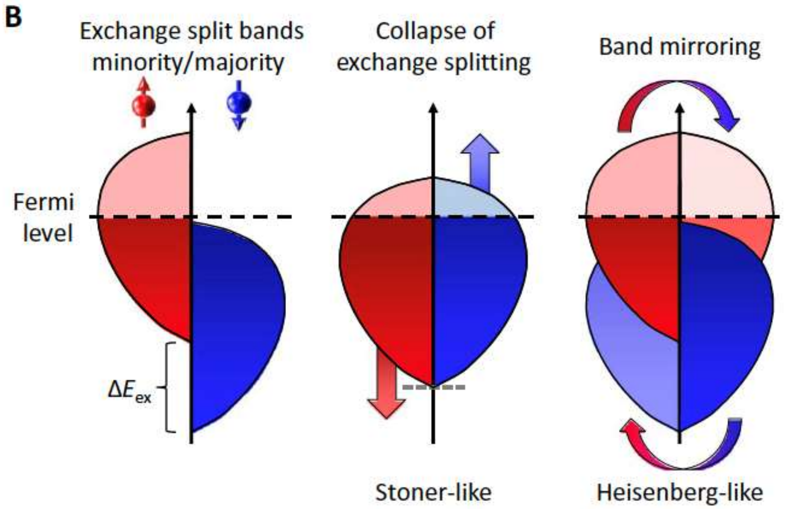
Ultrafast demagnetization of Fe and Co - recent exps.

Ultrafast magnon generation in Co,

not reduction of exch. splitting

(spin, energy & time resolved ARPES)

Eich et al, Sci. Adv. **3**, e1602094 (2017)



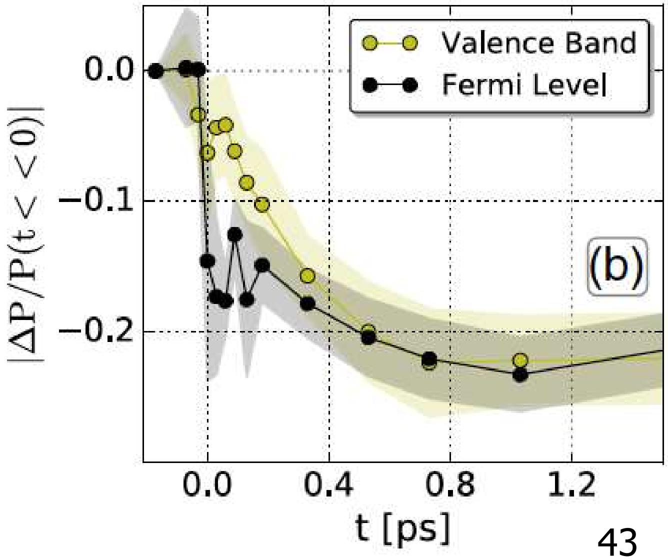
Observation of energy-dep. spin-polar. reduction in Fe

and band mirroring

(Spin, energy & time resolved ARPES)

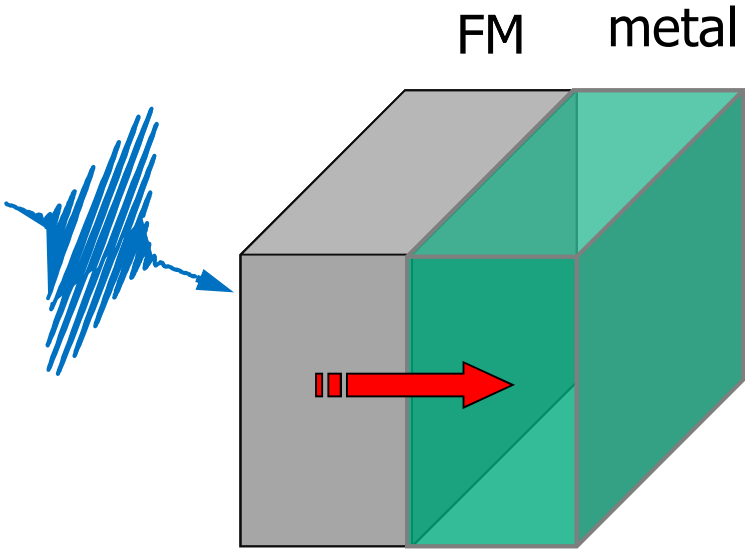
Gort et al, PRL **121**, 087206 (2018)

→ Non-rigid atomic moment

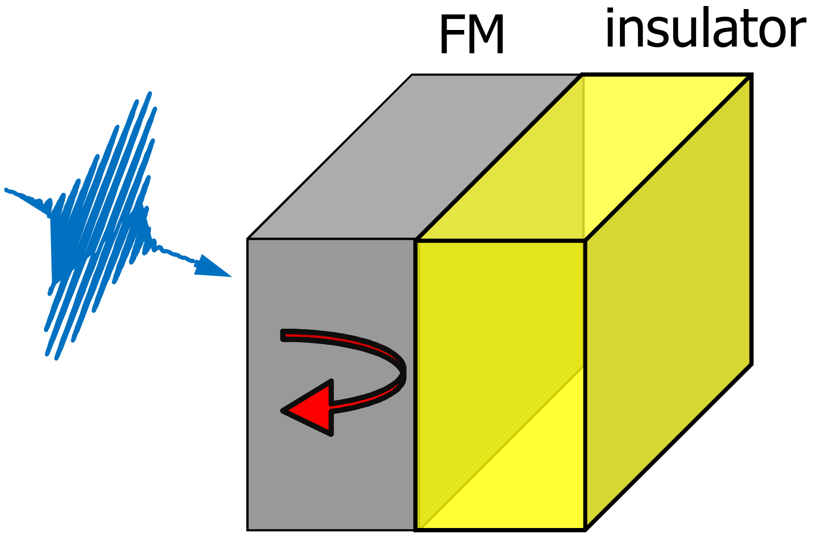


Ultrafast demangetization: an emerging picture

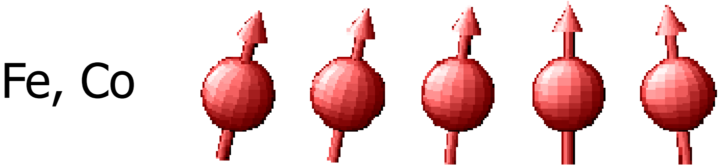
<u>Importance of the substrate</u>



- Strong & fast demagnetization
- Superdiffusive spin currents can contrib. significantly
- Other contributions smaller



- Smaller & slower demagnetization
- No spin currents possible
- Mainly spin wave generation and lesser el.-phonon spin flips contribute



Summarizing …

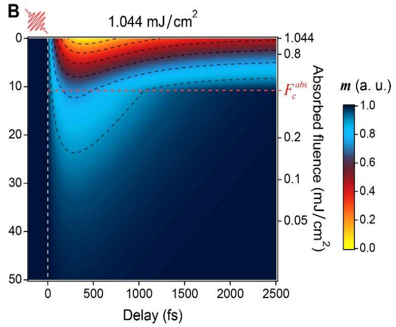
- *IFE: prediction of large, materials' specific opto-magnetic field*
  => that can be utilized for fast magnetic switching
- AO HD switching: Simulations give insight in details of switching
  (pulse length etc)
- Superdiffusive transport plays key role in demagnetization of Ni/Au
  (quantification of spin current pulse)
- Laser-excited Co (Fe): influence of fast magnon generation, less strong
  reduction of exchange splitting.
- Ni: possibly stronger effect of exchange reduction, non-rigid moment?

Outlook: Future directions …

Optimization of spin-current injection in nonmagnetic metal & STT

Modeling of excited spin-waves & switching in trilayers

Include depth-dependence of demagnetization & T_e



You et al, PRL **121**, 077204 (2018)

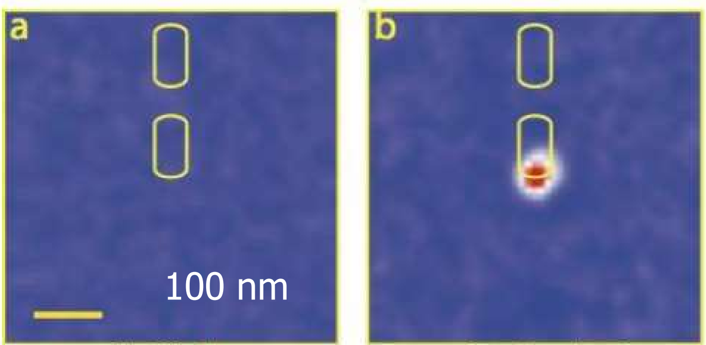
Ultrafast demagnetization & spin transport in lanthanide magnets

Ultrafast demagnetization in antiferromagnets

IFE: Materials' optimization, ab initio & atomistic simulations

Combine with hot electron (spin) currents

AOS on the nanoscale: optimization of plasmonic near field & IFE material



initial single shot

Liu et al, Nano Lett. **15**, 6862 (2015)

Theory for non-rigid atomic moment

Include ab-initio theory for non-equilibrium beyond 2TM

Acknowledgements & thanks to


Pablo Maldonado


Karel Carva


Ulrike Ritzmann


Marco Berritta


Leandro Salemi

Stefan Mathias (U-Göttingen), Martin Aeschlimann (U-Kaiserslautern)

Dominik Legut (U-Ostrava)

Oksana Chubykalo-Fesenko (CSIC, Madrid)

Margaret Murnane, Henry Kapteyn
Patrik Grychtol, Emrah Turgut (Boulder)

Marco Battiato (U-Singapore)

Claus Scheider, Roman Adam (Jülich)

Martin Weinelt, Tobias Kampfrath (FU-Berlin)

Pavel Baláz (Charles Uni, Prague)


Ritwik Mondal


Jerome Hurst

Söhnke Wienholdt, Denise Hinzke
Ulrich Nowak (U-Konstanz)

Sergiu Ruta, Roy Chantrell (York University)

Markus Münzenberg (U-Greifswald)