Ultrafast demagnetization dynamics observed at $M_{2,3}$ edges of hcp Co - first principles calculations

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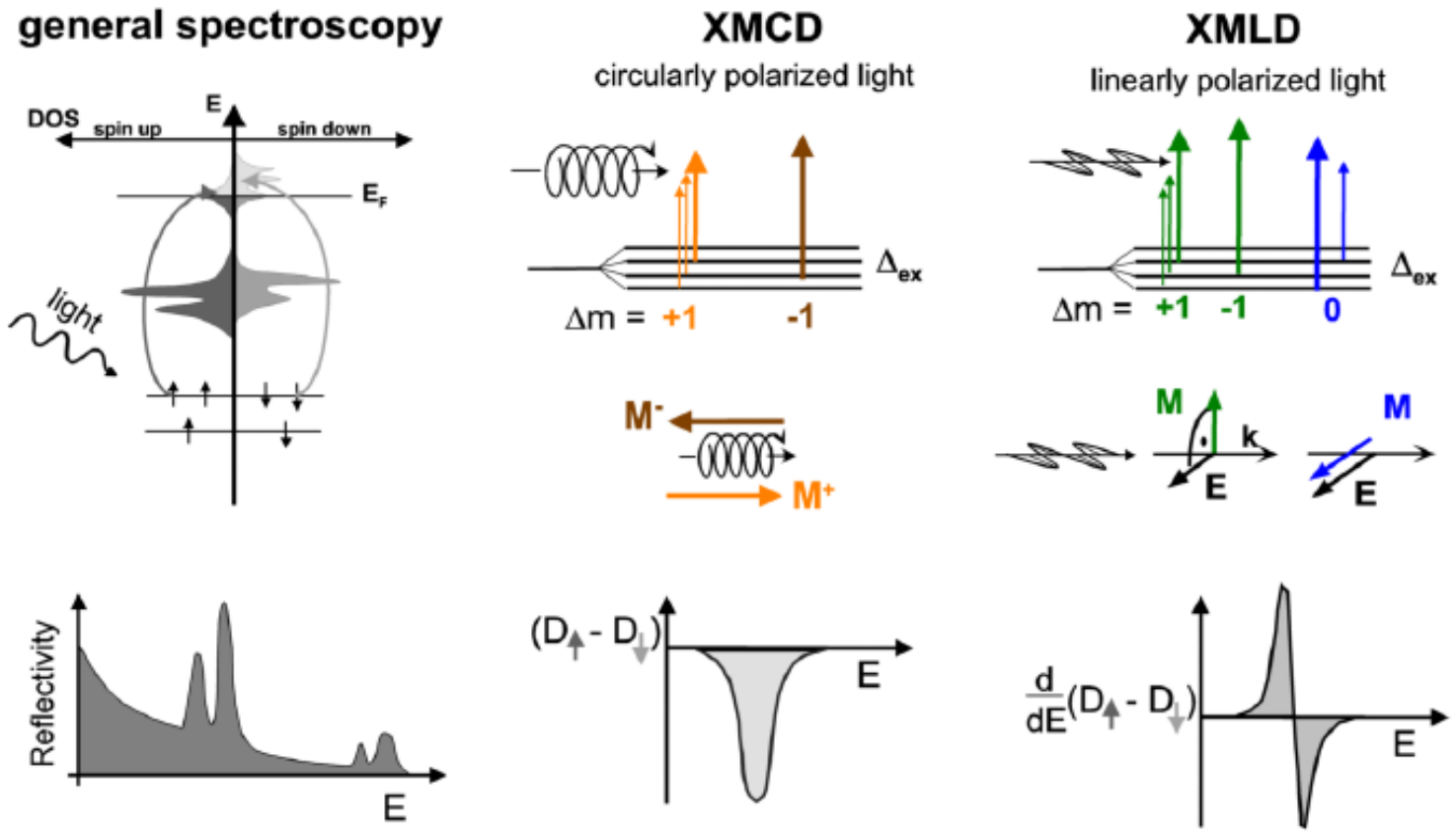
Motivation: magneto-transport linear and quadratic in $\vec{M}$

	conductivity	magneto-optics	X-ray
$\varepsilon_{ij}^{(1)}$, $i \neq j$	Hall effect	MOKE, magnetic circular dichroism and birefringence	XMCD
$\varepsilon_{ij}^{(2)}$, $i \neq j$	longitudinal Hall (quadratic Hall)	quadratic MOKE	quadratic XMCD
difference of diagonals, $\varepsilon_{ii}^{(2)} - \varepsilon_{jj}^{(2)}$ $(i \neq j)$	AMR	Voigt effect, magnetic linear dichroism (MLD), Schäfer-Hubert	XMLD

- different photon energy ranges: dc (i.e. $\omega = 0$), visible, X-ray
- what part of permittivity/conductivity tensor is measured (diag./off-diag.)
- but: symmetry arguments equal for **all** quadratic magneto-transport effects
- presented arguments are equally valid both for permittivity $_{ij}$ and conductivity σ_{ij} tensors as they are related by $_{ij} = \delta_{ij} + (i/\omega)\sigma_{ij}$.

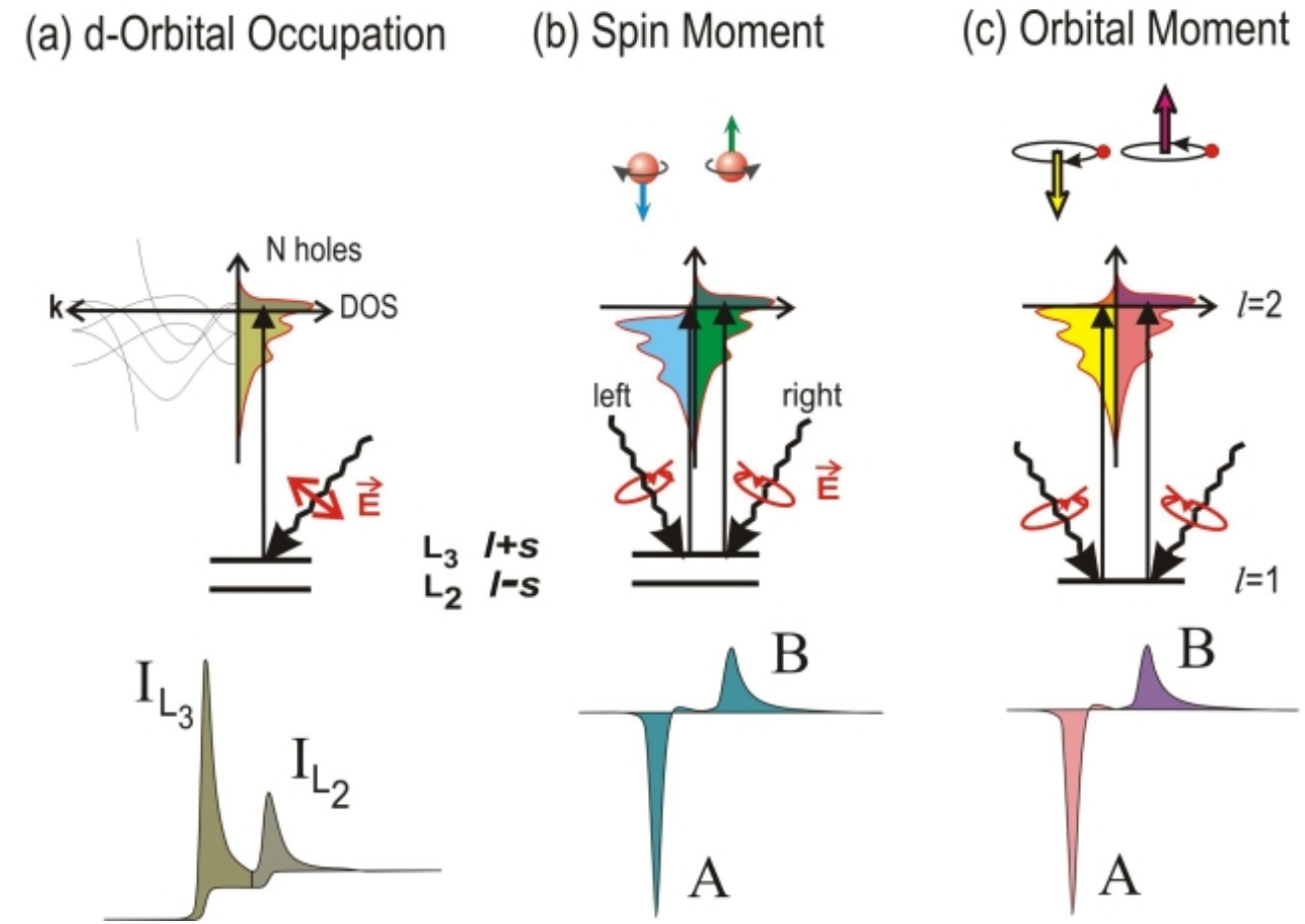
Electronic structure probed by magneto-optical spect.



→ most sensitive to magneto-crystalline anisotropy: XMLD

JMMM 272, 2146 (2004), PRL 104, 187401 (2010), PRB 84, 054448 (2011)

XMCD: sum rules:



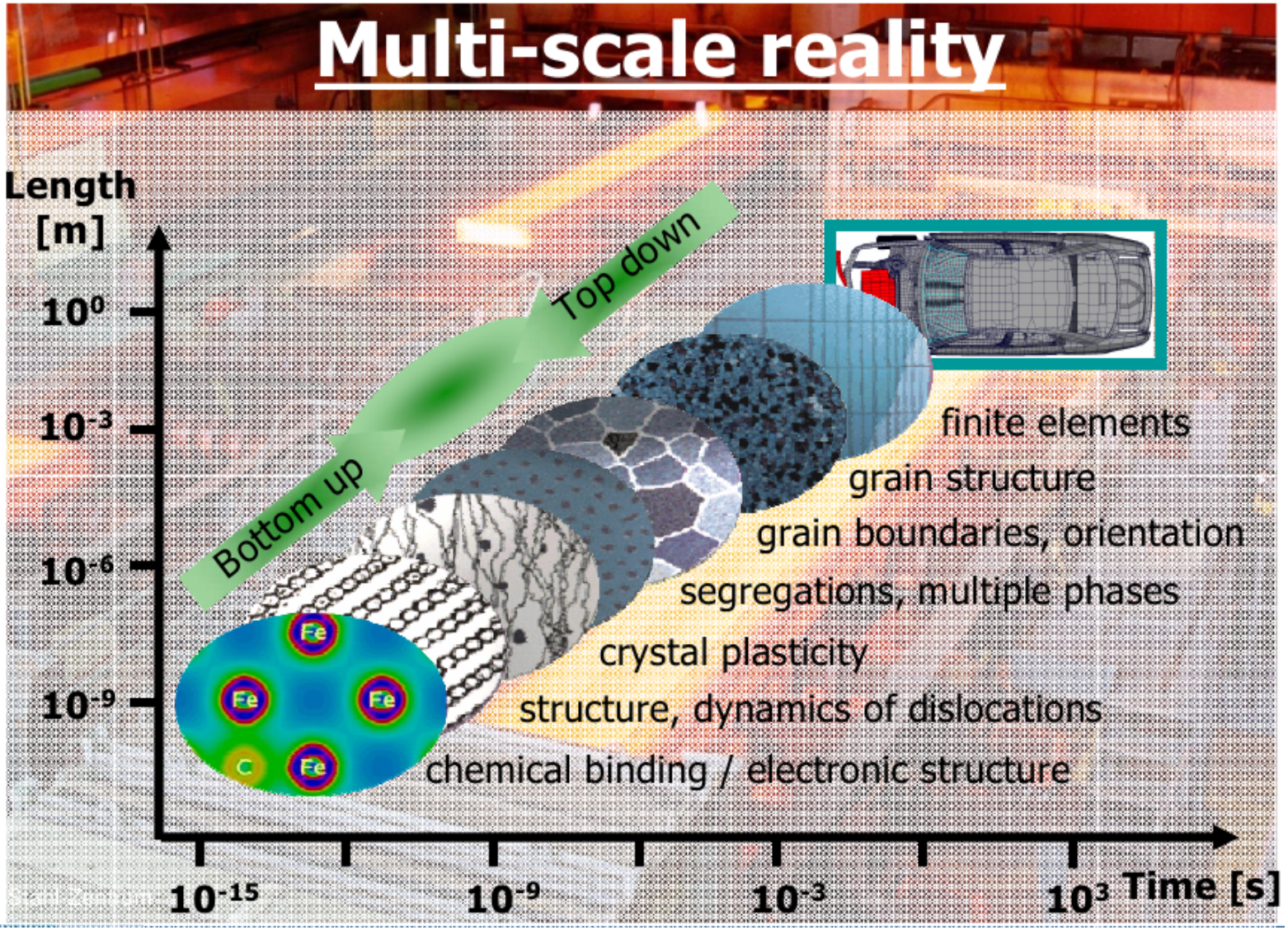
$$N_h = \left\langle I_{L_3} + I_{L_2} \right\rangle / C$$

$$m_s = \mu_B \langle -A + 2B \rangle / C$$

$$m_o = \mu_B \langle A + B \rangle / 3C$$

http://ssrl.slac.stanford.edu/stohr/xmcd.htm

Length and time scale



First-principles calculations - utilizing HPC

Many chemical and physical properties could be calculated from first-principles (ab initio) electronic structure calculations based on solving of:

Fundamental quantum theory, Schrödinger/Dirac equations

&

Density functional theory*

The only input:

- atomic number of constituents
- some structural information

* in 1998 awarded by Nobel prize in Chemistry, W. Kohn & J. Pople

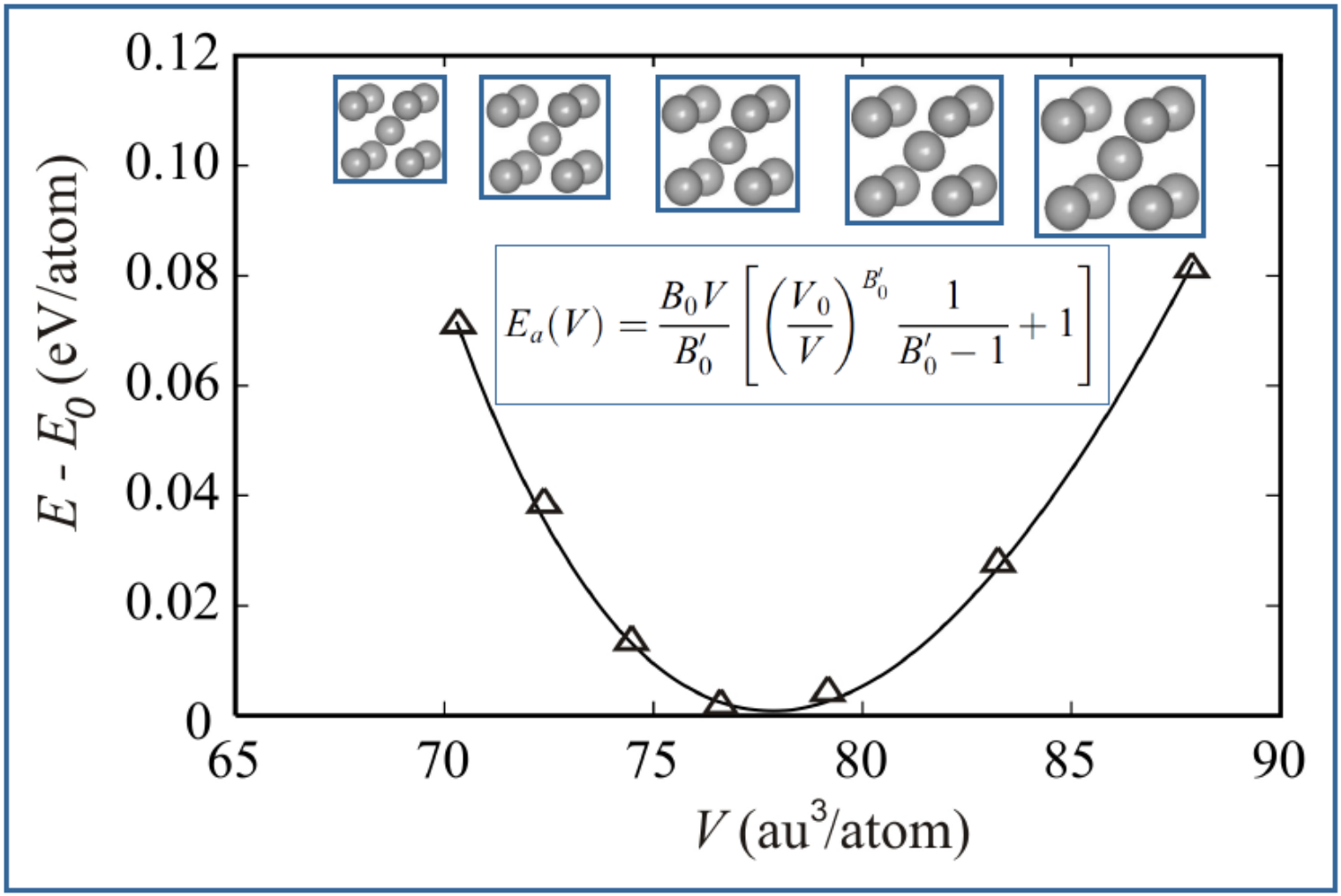
Quantum-mechanical description

Many-body (~10^{23} per cm^3) Schrödinger equation:

$$H\,\Psi(\{\mathbf{r}_i\},\{\sigma_i\}) \;=\; E\,\Psi(\{\mathbf{r}_i\},\{\sigma_i\})$$

10^{3*23}-dimensional many-body wave function
- analytic solution does *not* exist
- numerical solutions only
- **1 Fe atom**, 10 data-points/dimension ... $10^{3*(26+1)}$ bytes
  (1 DL-DVD ~ 10^{10} bytes) ... 10^{71} DVDs ... "mission impossible"!
- such a solution is unnecessary and would also be a non-sense !
- further approximations are necessary !

DFT calculation - Simple Example

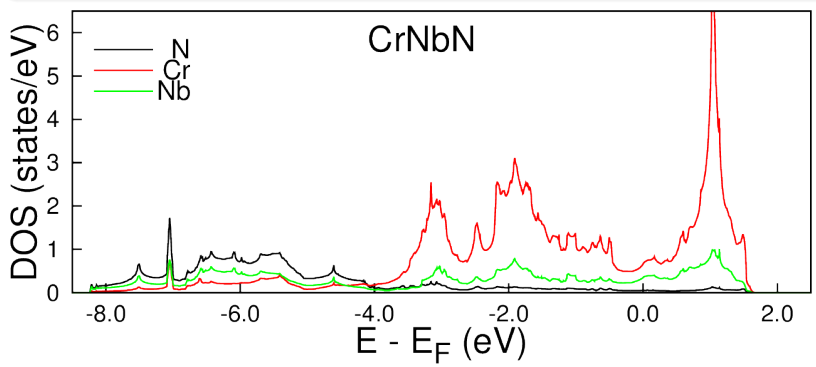


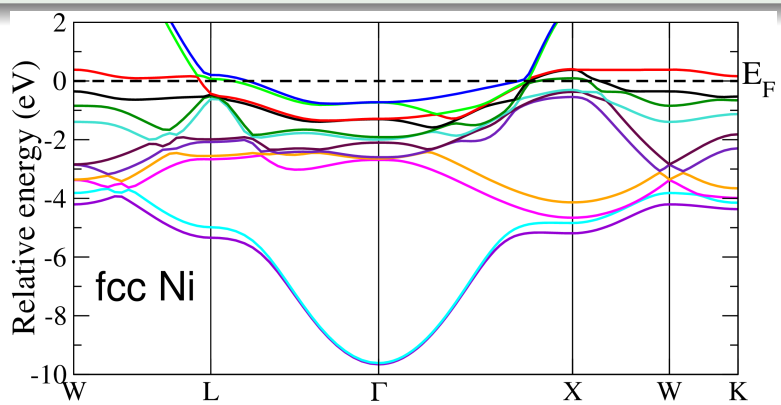
ATOMS ⟺ **POSITIONS** ⟺ **PROPERTY**

Quantities from *Ab initio* calculations

Selected list of quantities

- Density of states, bandstructure, Fermi level, band gaps
- Structural stability of phases
- Magnetic order, non-magnetic, ferro-, antiferro-, spin-spirals, transition temperatures
- Elasticity, is it brittle or ductile? Young moduli, strength
- Defects, lattice vibrations, thermal expansion
- Optical and magneto-optical properties Kerr, Faraday, Voigt x-ray absorption
- Relativistic effects, superconductivity





The principle of XMLD

Microscopic description:

- XMCD($\approx \mathbf{M}$) probes FM ordering only, sum rules gives us orbital and spin moment[a]
- XMLD($\approx \mathbf{M^2}$) probes FM and AFM ordering, anisotropy[b]
- XMLD$= \frac{T_\parallel - T_\perp}{T_\parallel + T_\perp} = \frac{R(H_\parallel) - R(H_\perp)}{R(H_\parallel) + R(H_\perp)} \approx \frac{\omega d}{c} Im[n_\parallel - n_\perp] \approx \frac{\omega d}{2cn} Im[\varepsilon_\parallel - \varepsilon_\perp - \frac{\varepsilon_{od}^2}{\varepsilon_\perp}]$
- $\varepsilon_\perp = \frac{1}{2}(\varepsilon_+ + \varepsilon_-)$ then XMLD$\approx \varepsilon_\parallel - \frac{1}{2}(\varepsilon_+ + \varepsilon_-)$, high symmetry directions (4-fold) symmetry $\varepsilon_\pm = \varepsilon_{xx} \pm i\varepsilon_{xy}$
- XMLD in reflection for p-polarized light (not-normalized):

$$\mathbf{XMLD} = \mathbf{R_{pp}(M_x)} + \mathbf{R_{pp}(-M_x)} - \mathbf{R_{pp}(M_y)} - \mathbf{R_{pp}(-M_y)}$$

This cancels the contributions linear in magnetization.

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[a] Thole et al, PRL 68, 1943 (1992), Carra et al, PRL 70,694 (1993)

[b] v.d. Laan PRL (1999), H. Kuneš and P. M. Oppeneer, PRB (2003)

Methodology: FLAPW calculations (WIEN2k code)

For the 3p edges:

- Relativistic (spin-orbit) SCF cycle (WIEN2k)
- Routines for
  1. Linear response theory - Kubo formula tensor $\varepsilon_{ij}(\omega)$
  2. Momentum matrix elements ($M_i = \langle n' \overrightarrow{k} | \overrightarrow{p} . \overrightarrow{e} | n \overrightarrow{k} \rangle^2$), *i.e.* among all bands and over all k-points
  3. Joint density of states - absorptive part of ε_{ij}
  4. In-house Kramers-Krönig relations $\rightarrow$ complex ε_{ij} tensor elements
- XAS, XMCD, XMLD, etc. based on Yeh's 4-vector formalism[a]
  1. Optical response of multilayered structure for various polarization vectors (**E**), incidence wave vector (**k**) with respect to magnetization direction (**M**) and crystal axis
  2. Obtaining energy dependence of reflectivity (R_{pp}, R_{sp}, R_{ps}, R_{ss}) and transmitivity (T_{pp}, T_{sp}, T_{ps}, T_{ss}) for $\rightarrow$ LMOKE, TMOKE, QMOKE, MLD, Schäffer-Hubert (Voigt) effect etc. for X-ray as well as for valence states.
  3. including grazing incidence

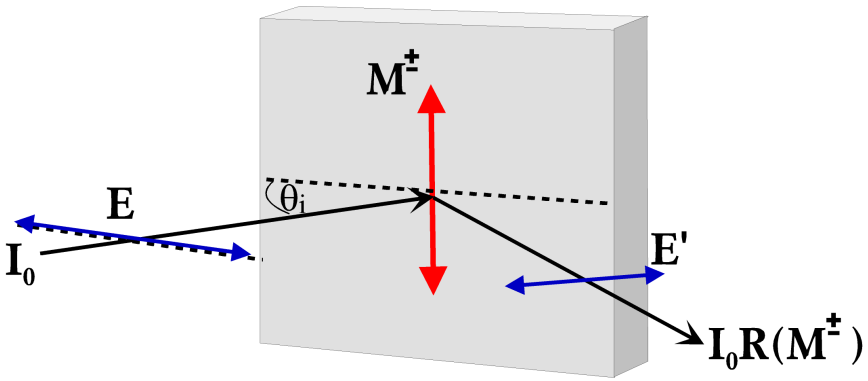
[a] Yeh (1980), Višňovský (1991)

Experimental setup & calculations on equal footing

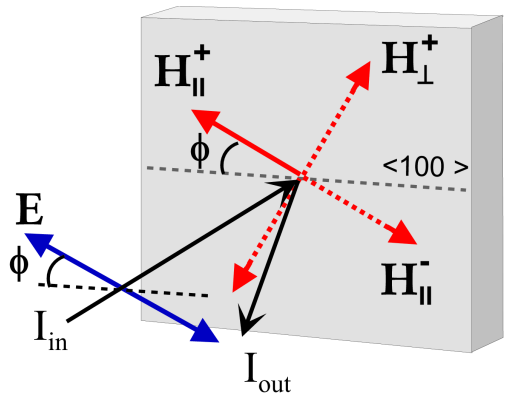
3nmAu/x nm Fe/150nm Ag/GaAs

$$A_{TMOKE} = \frac{R_{pp}(M_+) - R_{pp}(M_-)}{R_{pp}(M_+) + R_{pp}(M_-)},$$

$$A_{XMLD} = \frac{R(H_{\parallel}) - R(H_{\perp})}{R(H_{\parallel}) + R(H_{\perp})}, \qquad R(H_{\parallel,\perp}) = \frac{R(H^{+}_{\parallel,\perp}) + R(H^{-}_{\parallel,\perp})}{2}$$

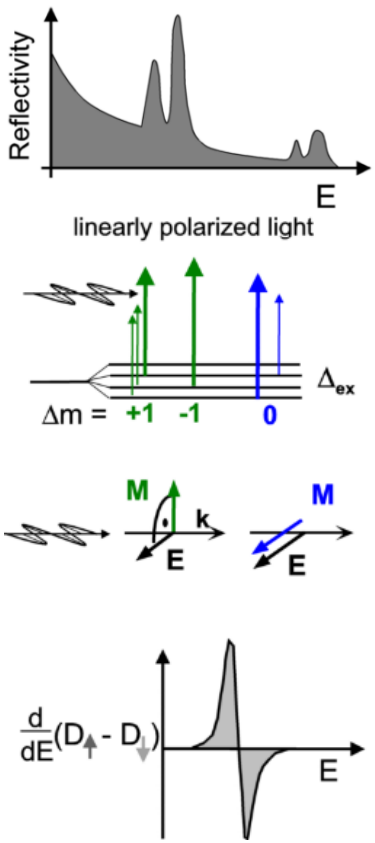


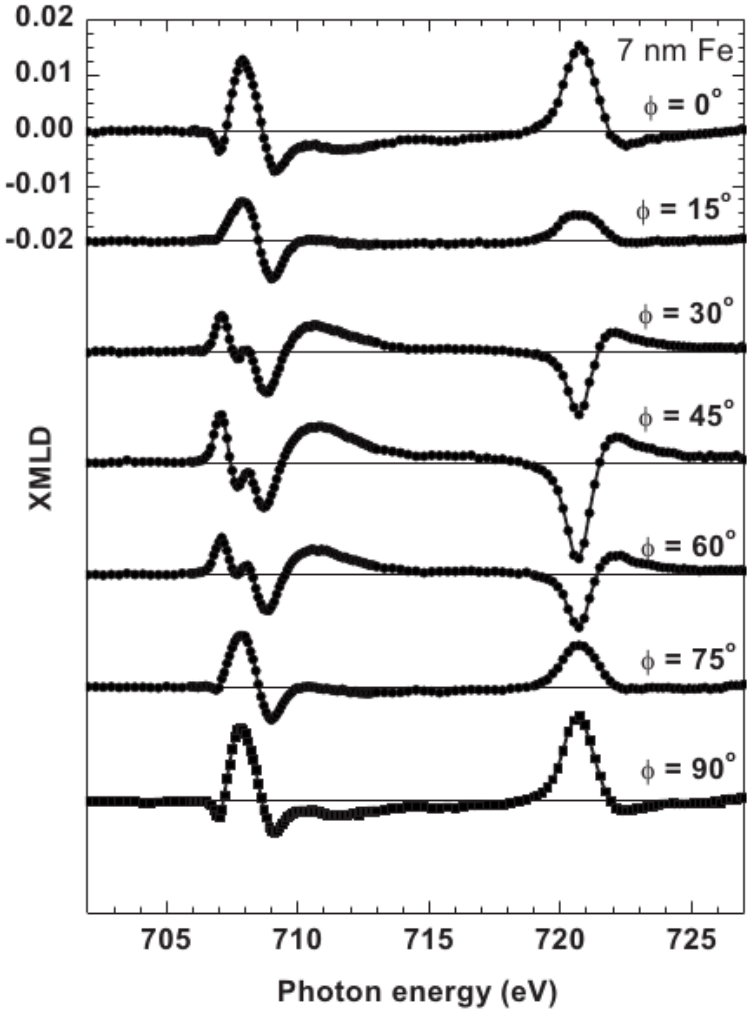
TMOKE setup

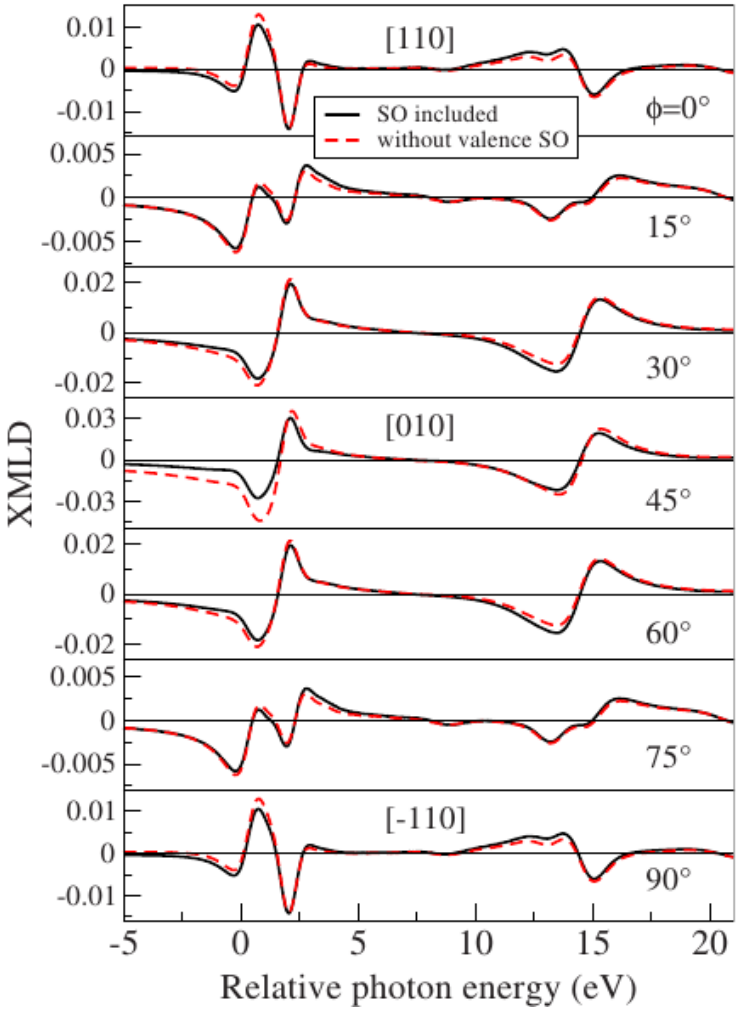


XMLD setup

XMLD at 2p edges in transmission - angular dependence





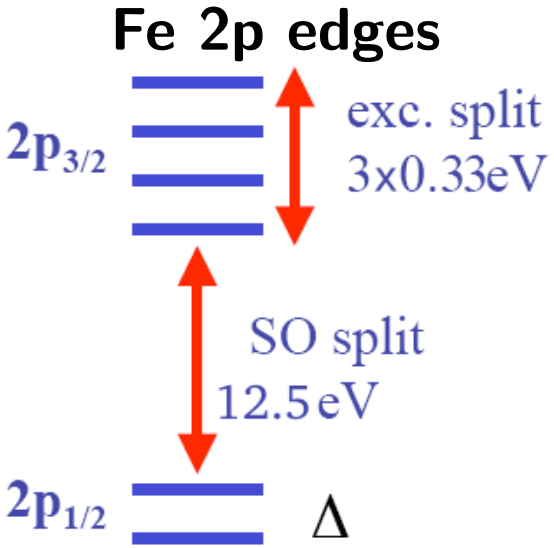


Nolting et al. PRB (2010), XMLD for high symmetry crystal directions at 2p edges,

theory by Kuneš and Oppeneer PRB (2003)

2p vs. 3p edges

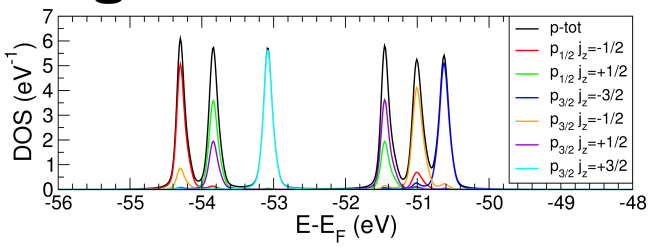
2p edges: larger energy separation, no **k**-dependence
3p edges: smaller energy separation ≈ hybridization, **k**-dependence

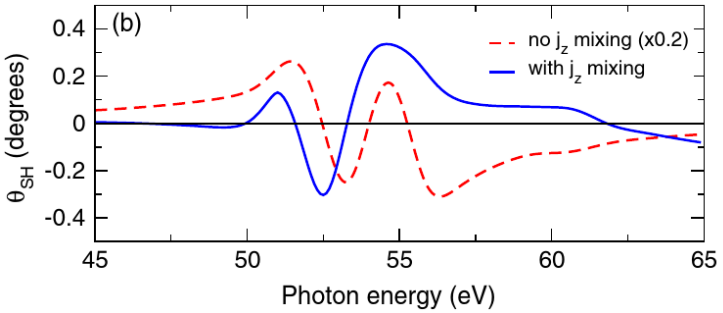


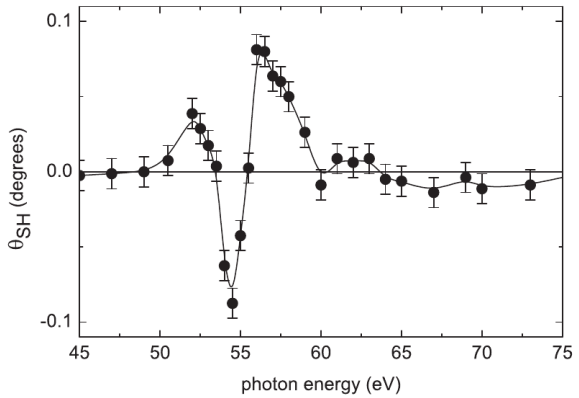
MO ≈ M^2 (XMLD in reflection, Schaeffer-Hubert effect, etc.)

Valencia et al., Phys. Rev. B **104**, 187401 (2010)

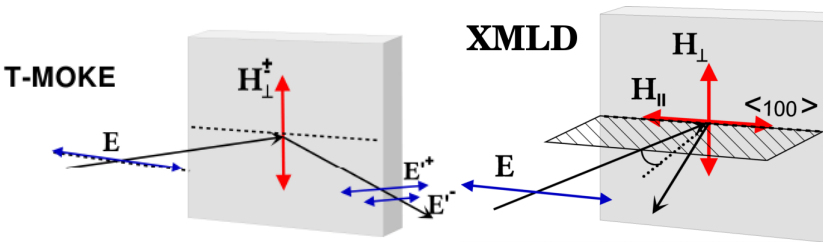
3p edges J_z splitting of 3*p*-states in bcc Fe

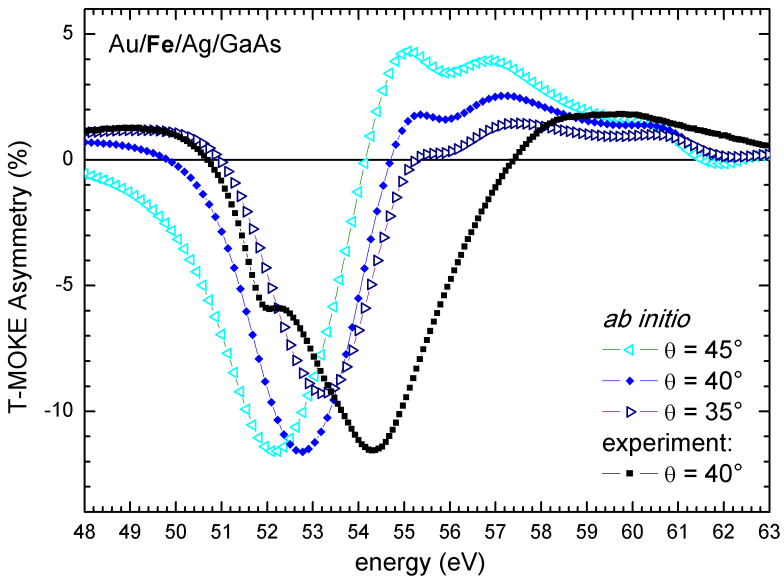


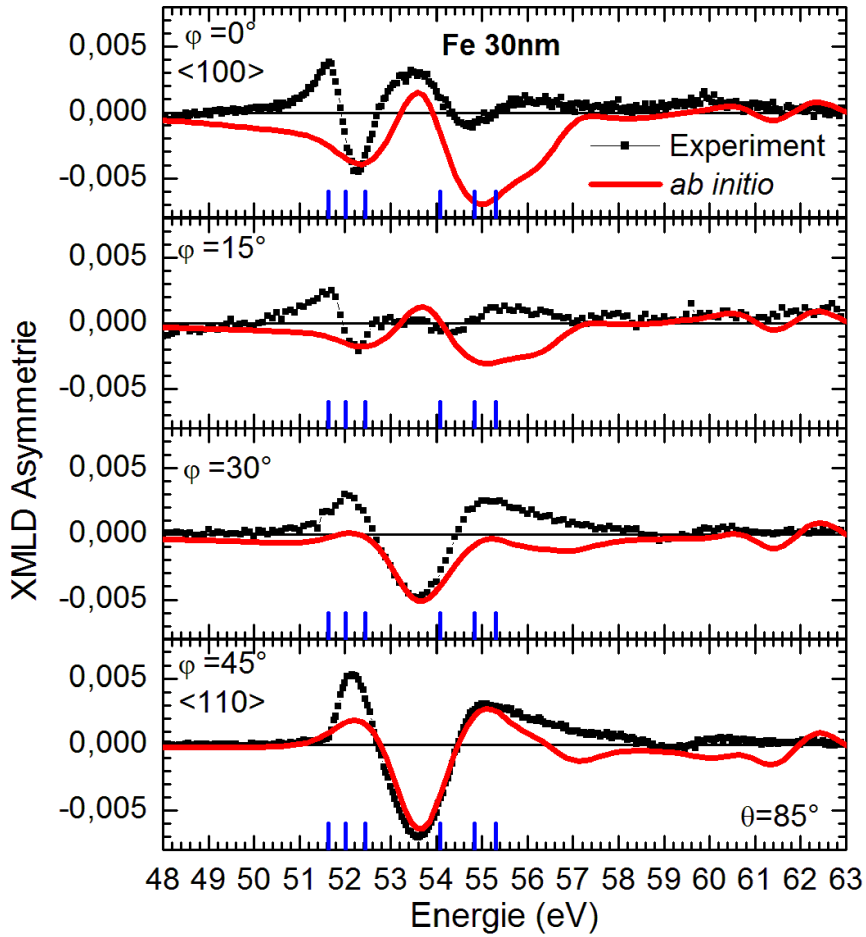


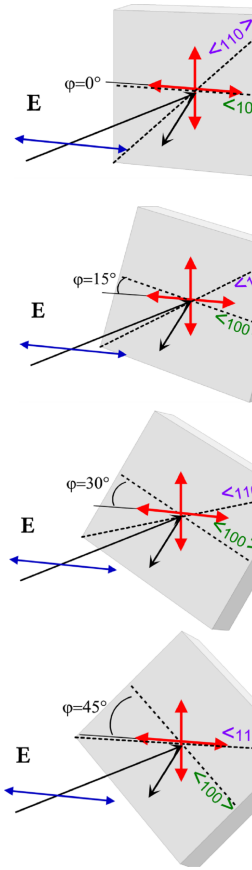


TMOKE and XMLD of 3p Fe edge: Exp. vs. Theory







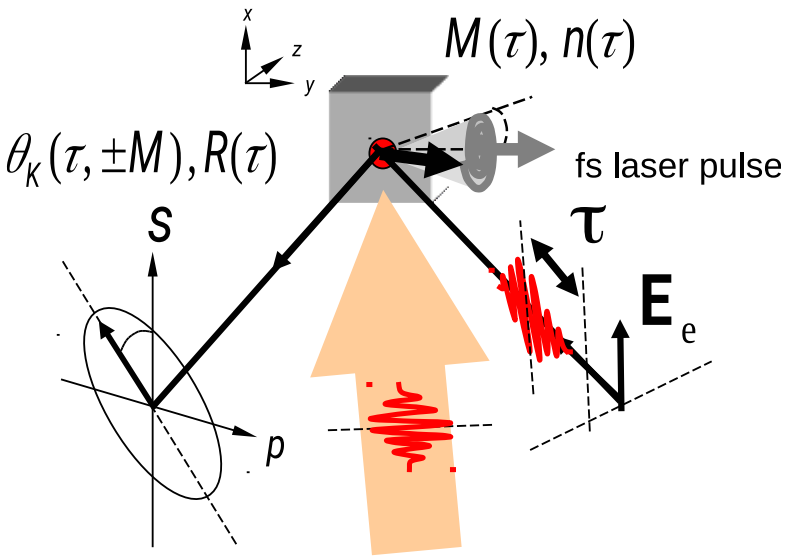


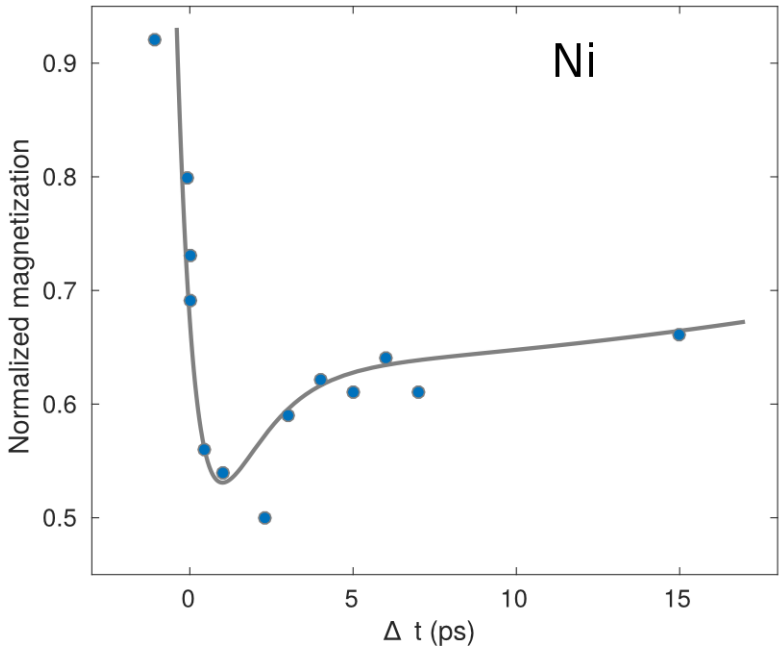
MO 3p edge is much susceptible to interference effects

Acta Phys. Pol. A **127**, 466 (2015), IEEE Trans.Magn., **50**, 2104704 (2014) J. Appl. Phys. **115**, 17E132 (2014), Phys. Rev. B **89**, 140404(R) (2014).

Start of the ultrafast demagnetization

Experiment opening the field of ultrafast demagnetization





Beaurepaire et al.(1996), magnetization quenched under 250fs.

Theories of ultrafast demagnetization

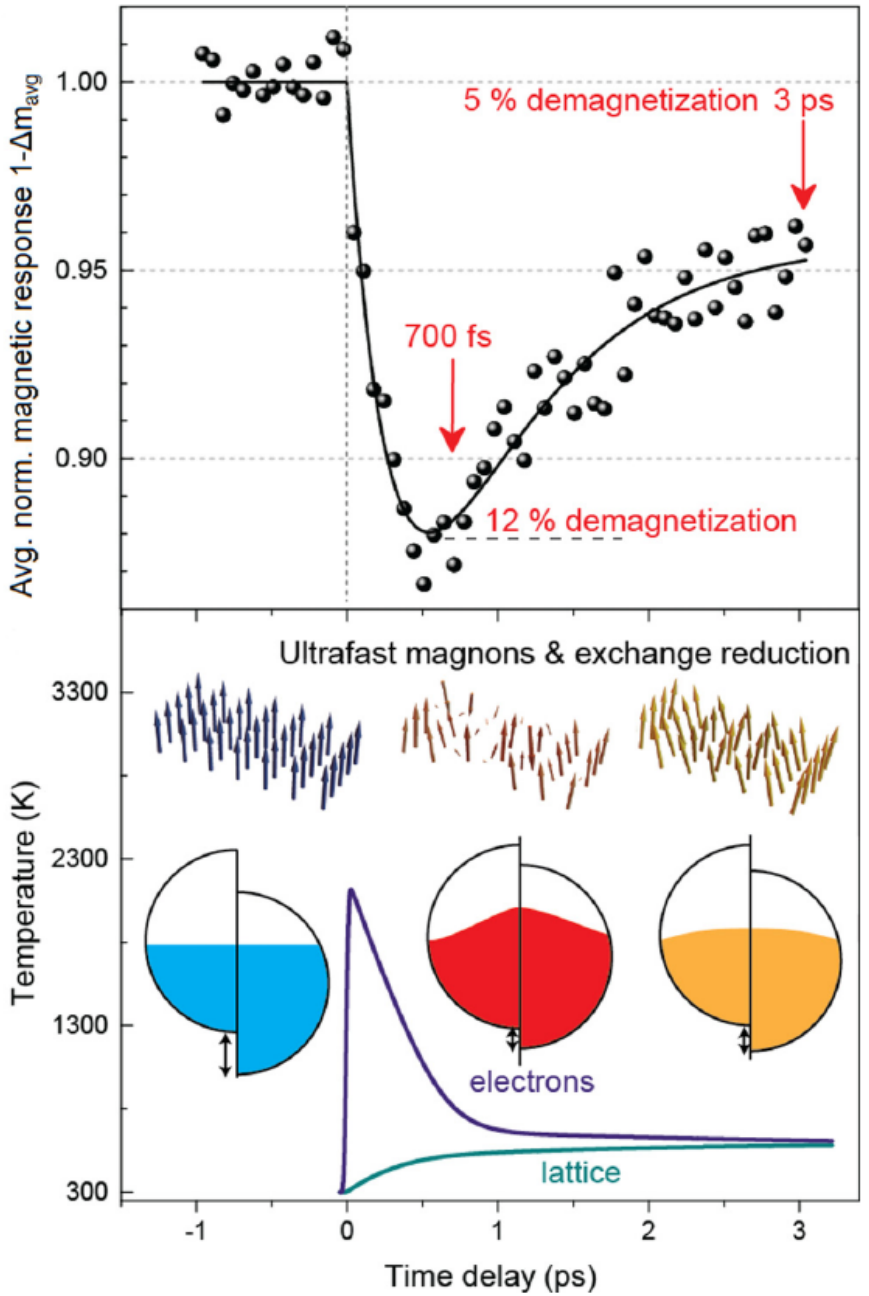
- Elliott-Yafet el.-ph. scattering, Koopmans et al., PRL 95 (2005)
- Spin-orbital interaction S→L, Zhang & Hübner, PRL 85 (2000)
- Fast magnon excitation, Carpene et al., PRB 78 (2008)
- Coulomb-exchange e-e scattering, Krauss et al., PRB 80 (2009)
- Chem. potential adjustment, Mueller et al., PRL 111 (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)

Ultrafast demagnetization in hcp Co

Three possibilities

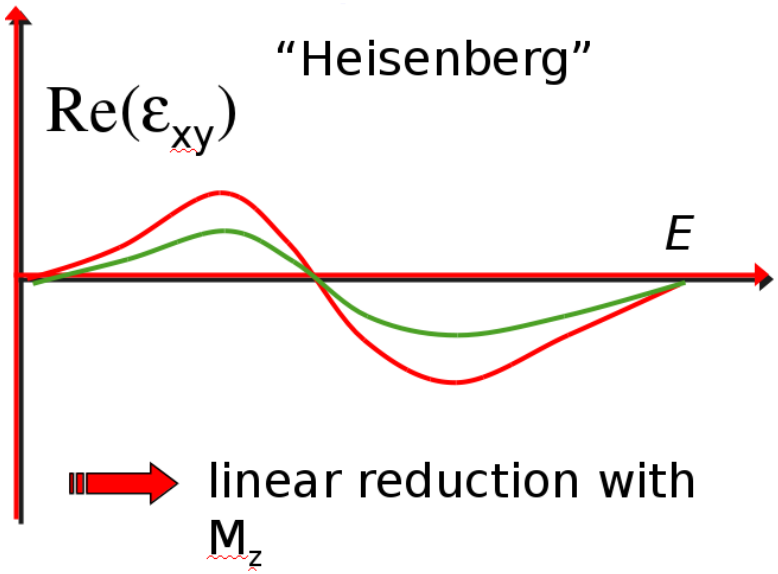
- Magnetic moment disorder (transverse spin excitations)
- Exchange splitting reduction (longitudinal spin excitations)
- Electron temperature increase ("hot" electrons)

- 10nm Co/SiO_2
- No superdiffusion
- Electron thermalized at 400 fs
- Measured at M-edges at ca 50eV



Magnon excitations vs. exchange excitations

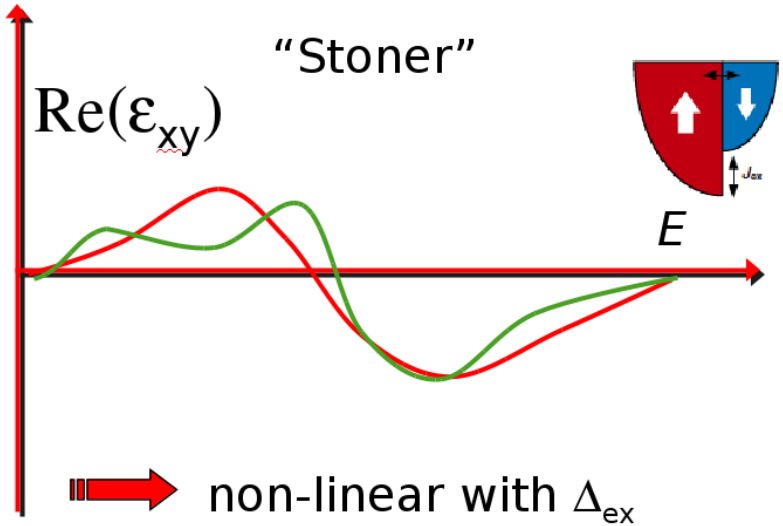
Spin wave



linear reduction with M_z

Transverse excitation

Reduced exchange splitting



non-linear with Δ_{ex}

Longitudinal excitation

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

Can be these effects distinguished by T-MOKE?

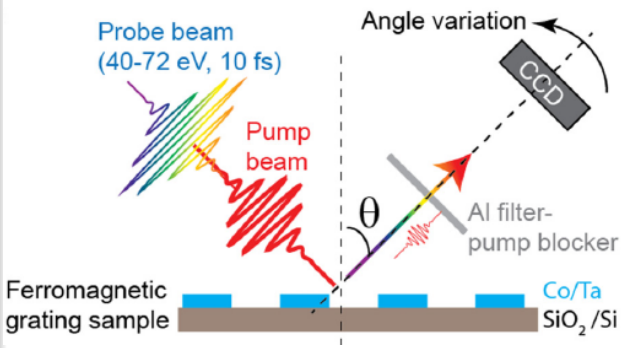
predicted by Erskine Stern, PRB 12, 5016 (1972)

- 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), where A is

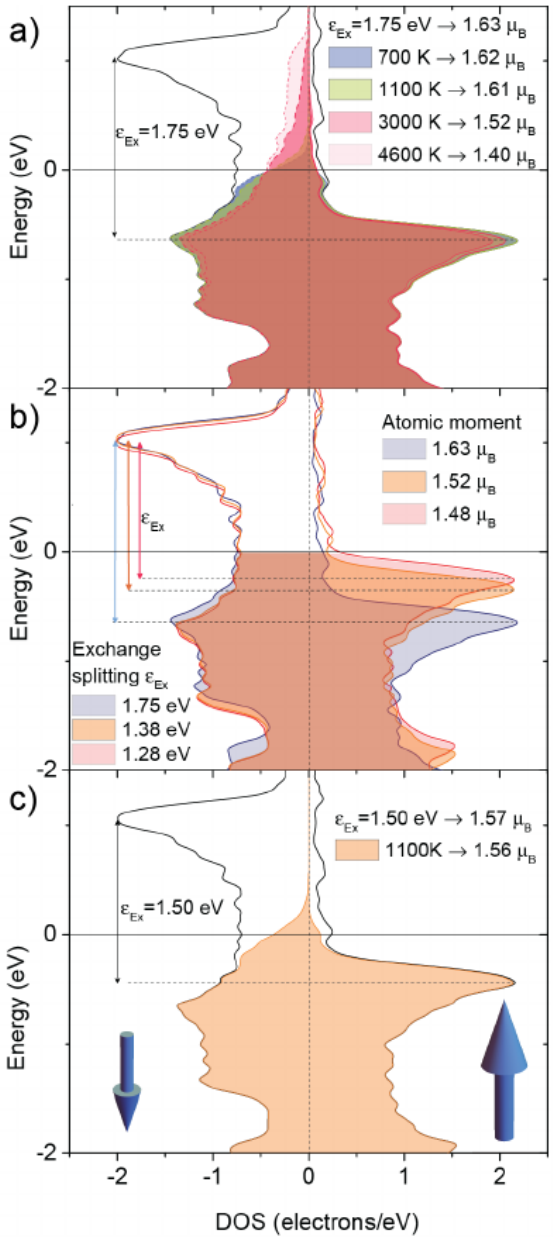
$$A(n,\theta,\varepsilon_{x,y}(t)) = \frac{I_p^+ - I_p^-}{I_p^+ + I_p^-} \approx 2Re\frac{sin2\theta\varepsilon_{x,y}(t)}{n^4 cos^2\theta - n^2 + sin^2\theta}$$

Compute the $\varepsilon_{x,y}$ from first principles (ab initio) for number of cases:

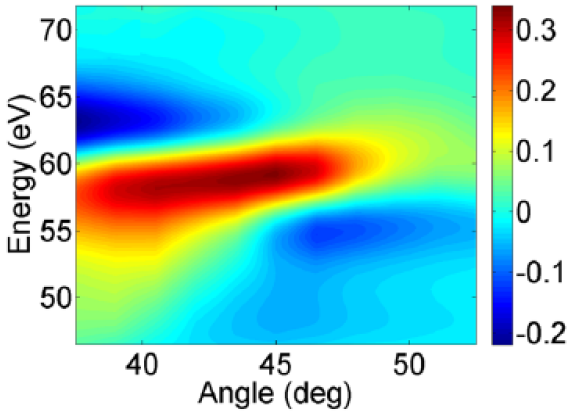
- magnon excitations
- reduced exchange splitting
- increased electron temperature
- construct A(t) wrt A(t=0), i.e. fitting with experiment

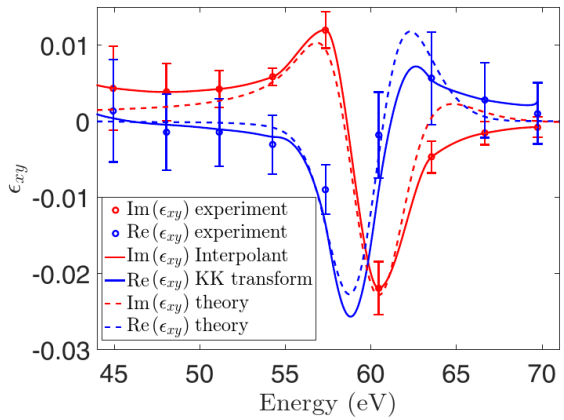


DOS for various excitations by relativistic code WIEN2k



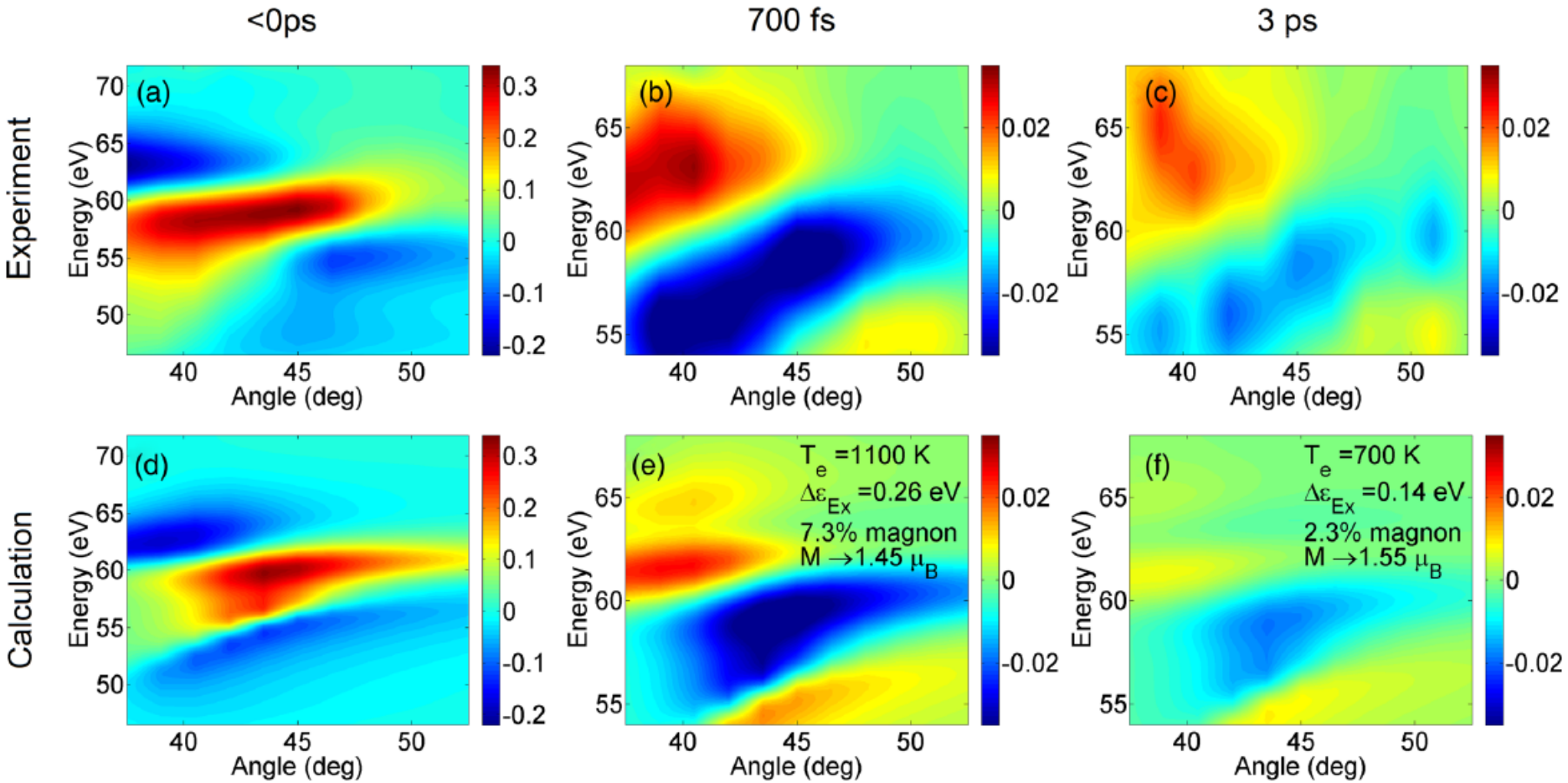
- a) hot electrons, FD multiplication
- b) reduce exchange, fixed spin moment calcs.
- c) combination of both





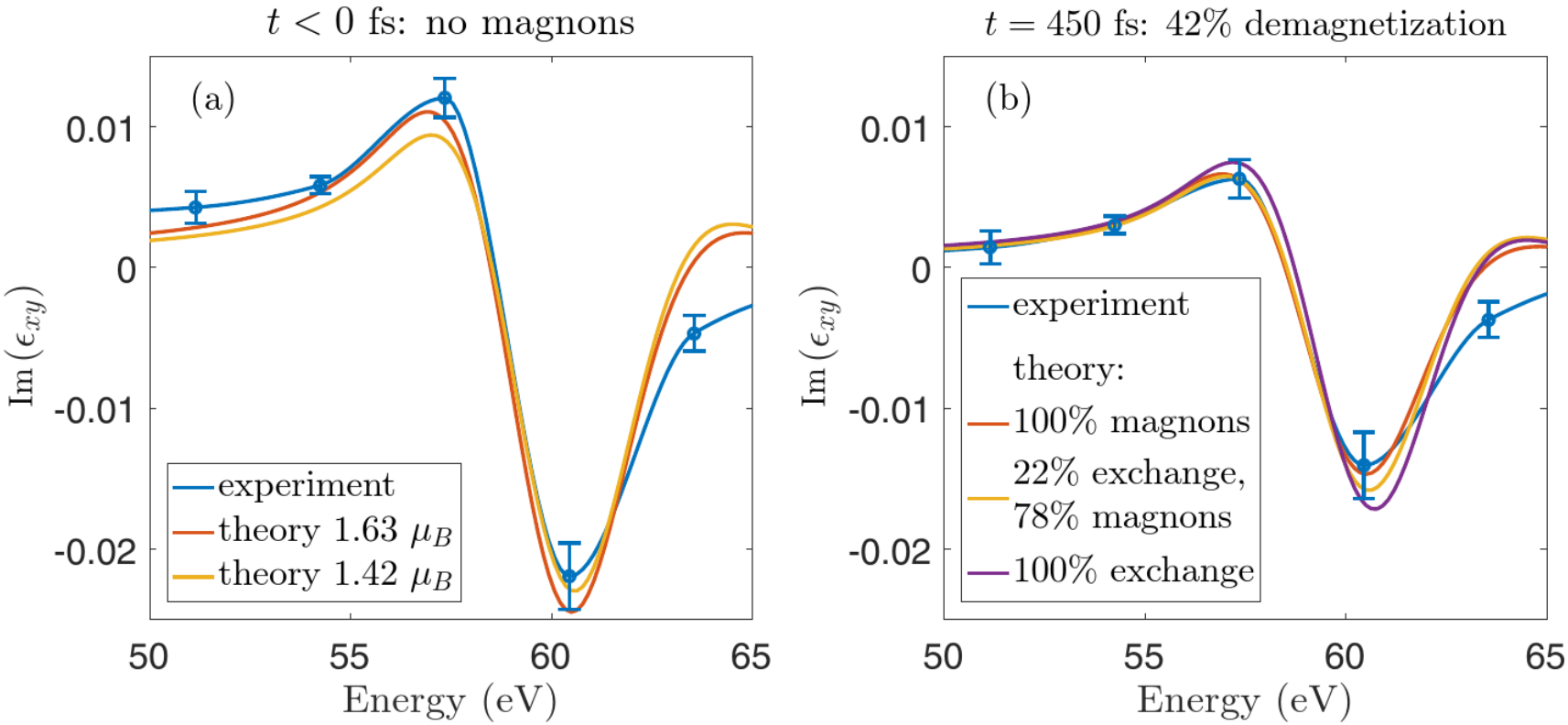
Experiment vs. theory - hcp Co

$$A(n,\theta,\varepsilon_{x,y}(t)) = \frac{I_p^+ - I_p^-}{I_p^+ + I_p^-} \approx 2Re\frac{sin2\theta\varepsilon_{x,y}(t)}{n^4cos^2\theta - n^2 + sin^2\theta}$$



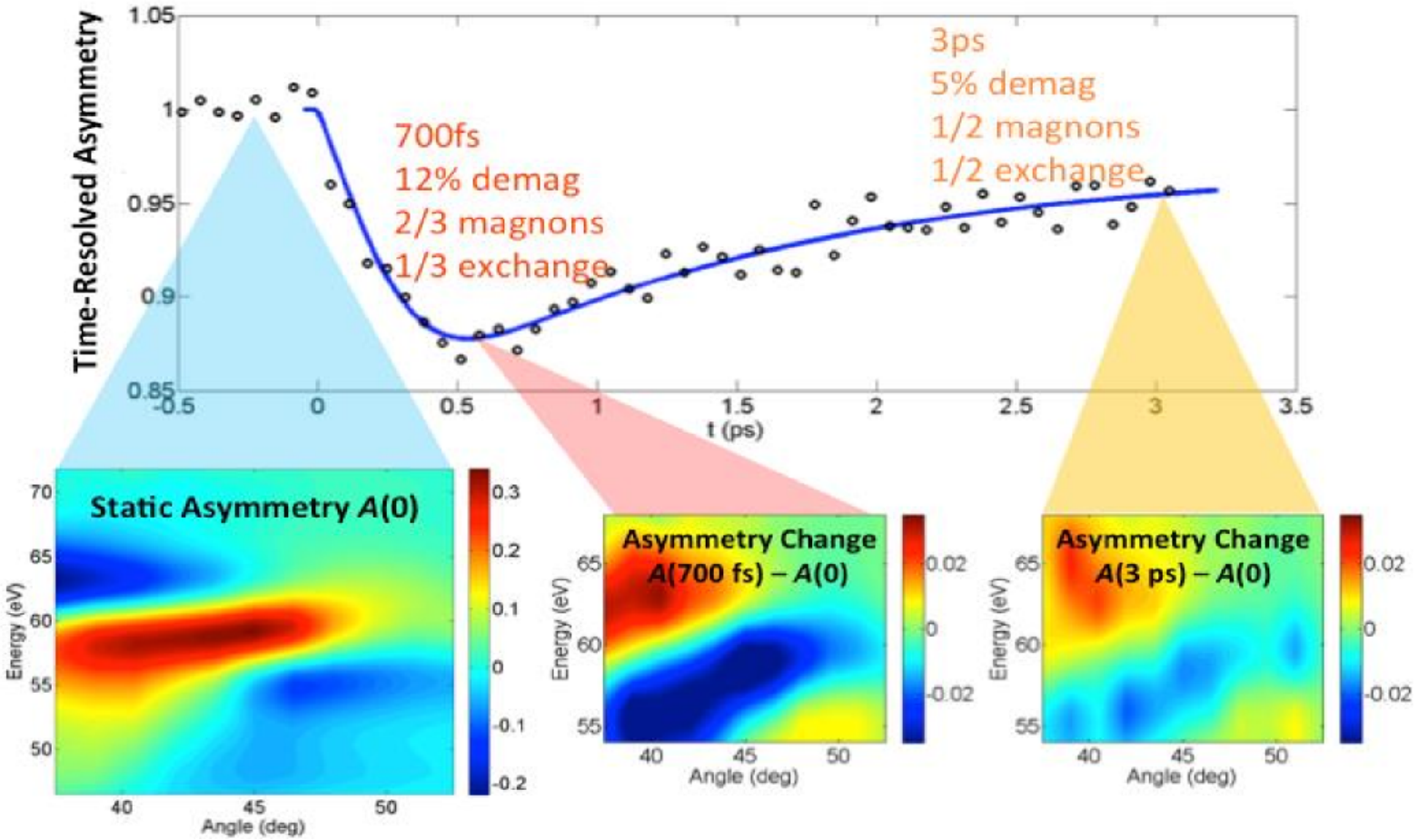
Least square fits to the whole spectrum, contribution from spin flips (exchange reduction) and magnons

$\varepsilon_{x,y}$ of hcp Co



Turgut et al., Phys. Rev. B **94**, 220408(R) (2016), Zusin et al. Phys. Rev. B **97**, 024433 (2018)

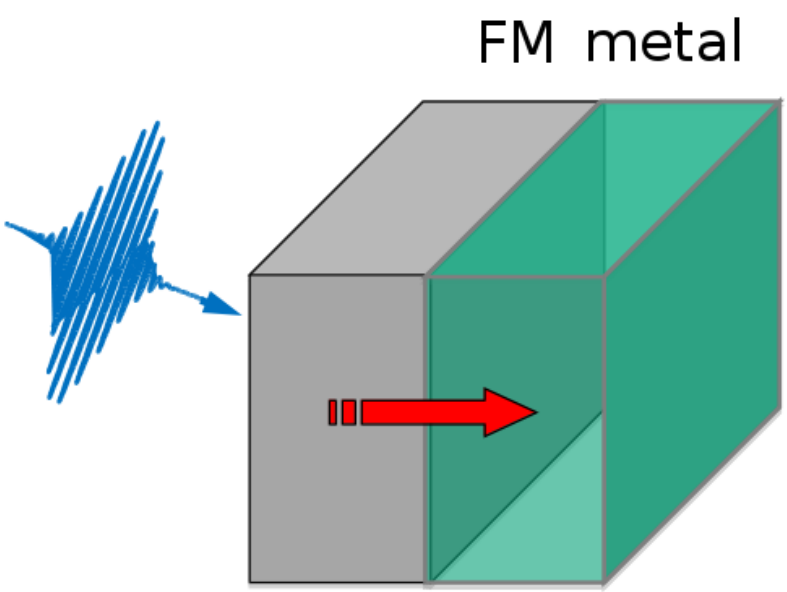
Causes of MOKE time evolution

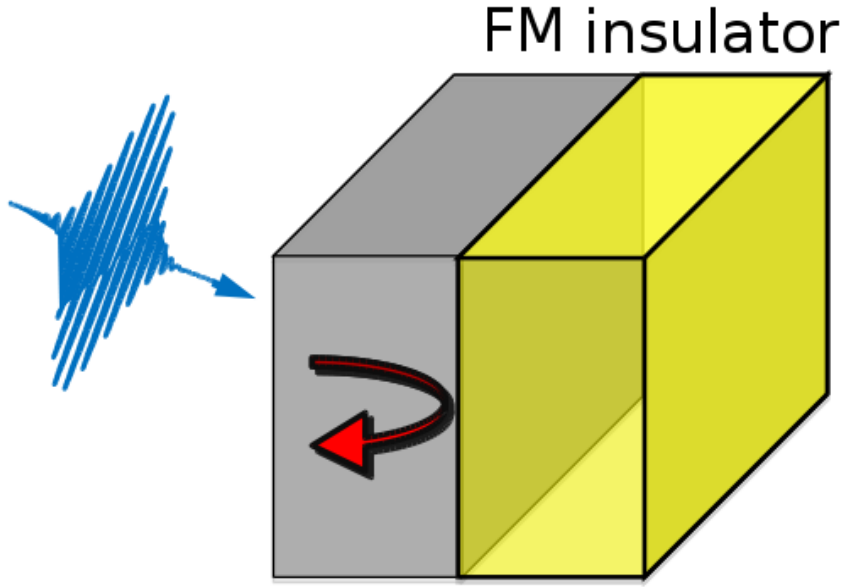


- suprisingly small contribution from spin flips (exchange splitting reduction)
- larger contribution by fast magnon excitations

Turgut et al., Phys. Rev. B **94**, 220408(R) (2016), Zusin et al. Phys. Rev. B **97**, 024433 (2018)

Emerging picture - Ultrafast demag. depends on substrate





- strong and fast demagnetization
- superdiffusive spins currents contribute predominantly

- smaller and slower demagnetization
- superdiffusive currents not possible
- due to spin wave generation and el-phonon spin flips

Conclusions and Outlook

MO effects in transition metals

- MO at 3p edges are accuratelly calculated wrt experiment
- Linear/Quadratic MO effects at 3p edges are one order of magnitude lower than XMLD at 2p edges
- MO asymmetry measurement allows us to disentangle the different contributions to demagnetization
- Both transverse and longitudinal excitations found to contribute, at 700fs Δm_t / $\Delta m_l > 2$, later the contribution from longitudinal component is growing
- Magnon/exchange splitting is not growing from fs into ps time
- Ultrafast demagnetization of hcp Co on insulating (SiO_2) substrate by reduced exchange splitting

Thank you for your attention !

One post-doc and Ph.D. position open at IT4I, VSB - Tech. Univ. Ostrava, Czech Republic

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