# Light – Matter Interactions (II)

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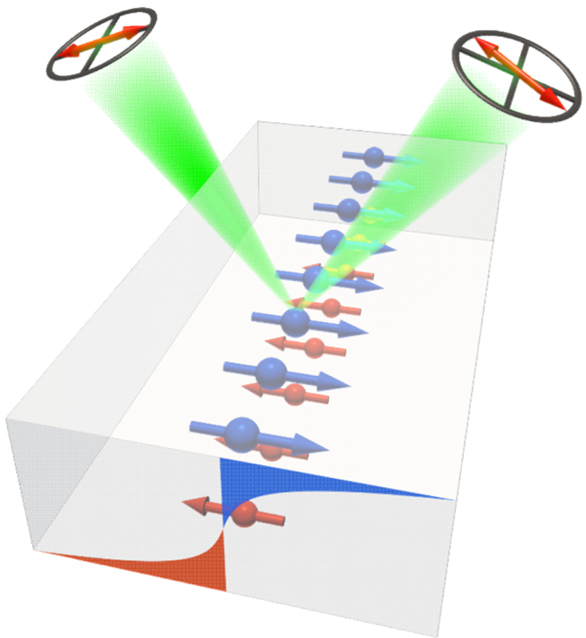
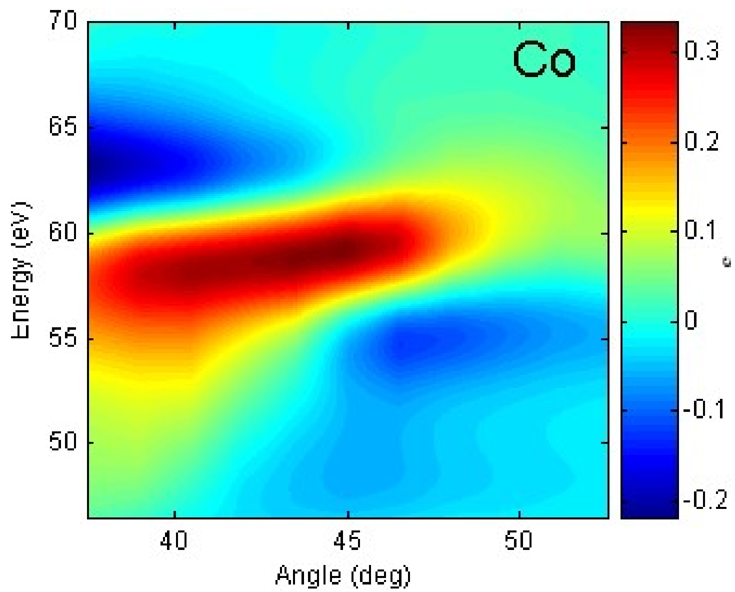
# Outline – Lecture II – Light-magnetism interaction

- Phenomenology of magnetic spectroscopies
- Electronic structure theory, linear-response theory
- Theory/understanding of magnetic spectroscopies
    - Optical regime
    - Ultraviolet and soft X-ray regime

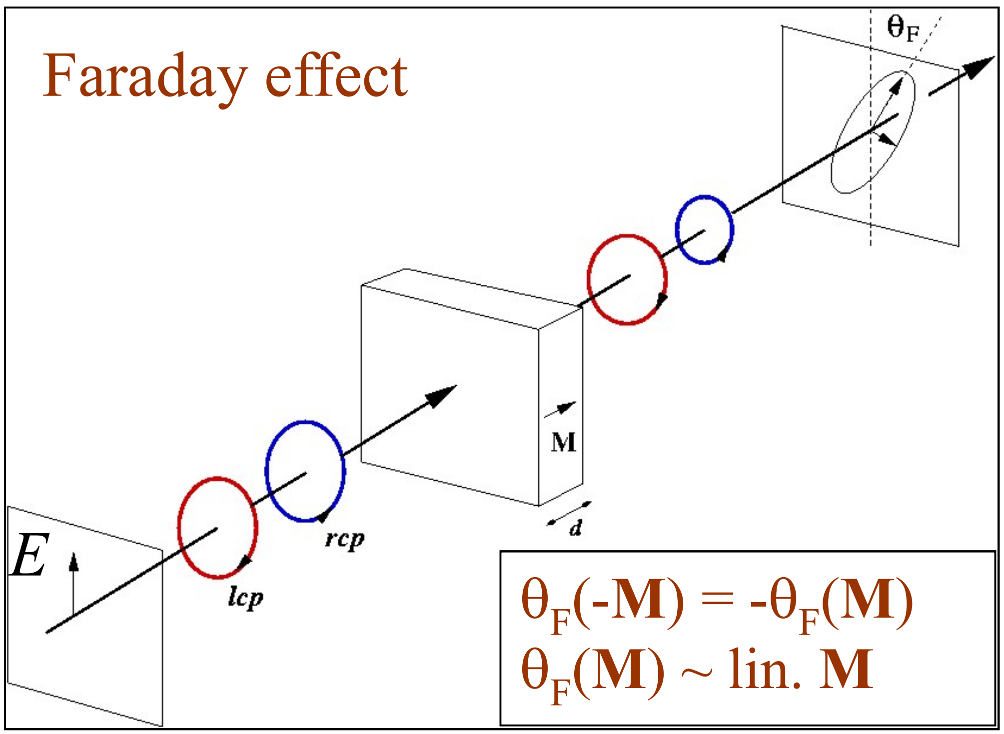




# In the beginning … First observation of magneto-optics

## Magneto-optical Faraday effect (1845)

- Observation of interaction light-magnetism
  enormous impact on development of science!




Michael Faraday
(1791 – 1868)

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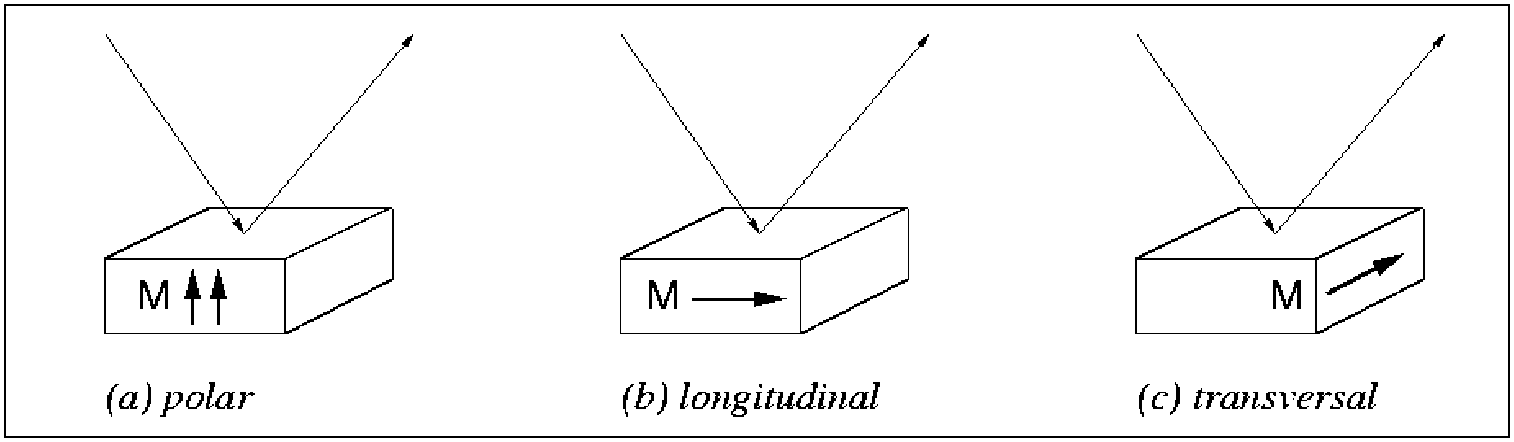
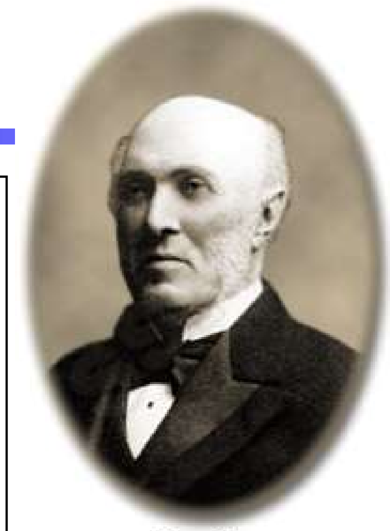
# Magneto-optical Kerr effect

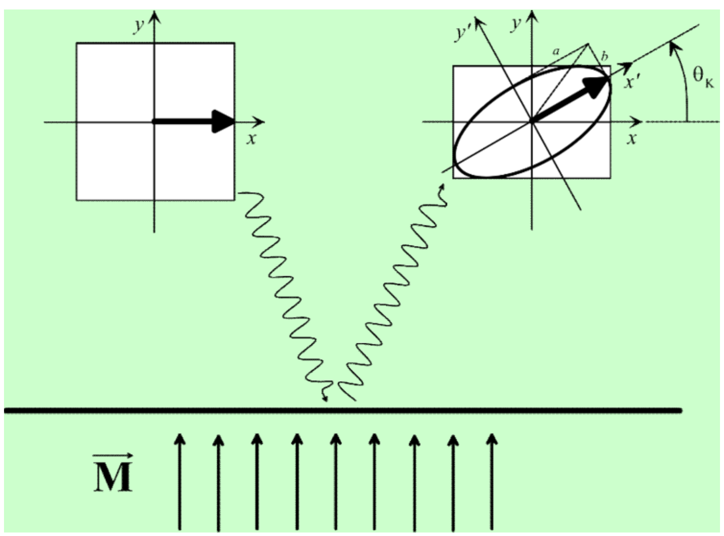




John Kerr (1824-1907)

Kerr (1876)
pol. analysis

Kerr (1878)
pol. analysis

Zeeman (1896)
intensity measurement



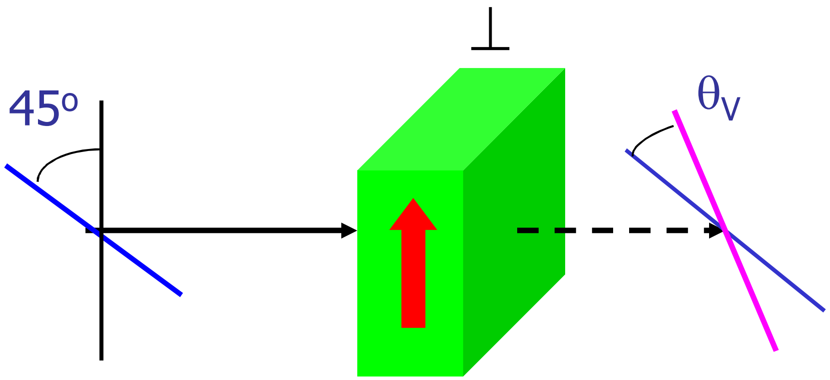

$$\tan \varepsilon_K = \frac{b}{a} = \frac{|r_+| - |r_-|}{|r_+| + |r_-|}$$

Rotation of the polarization plane & ellipticity: Completely a magnetic effect!

→ One of the best tools in magnetism research!

# Magneto-optical Voigt effect

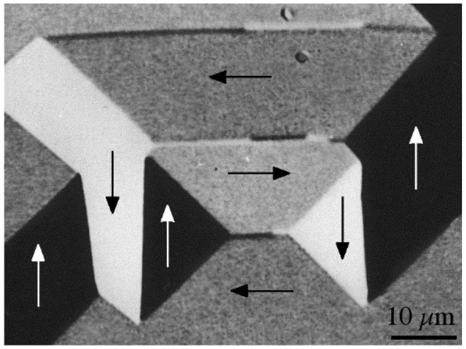
Voigt effect (1899)




Woldemar Voigt
(1850 – 1919)

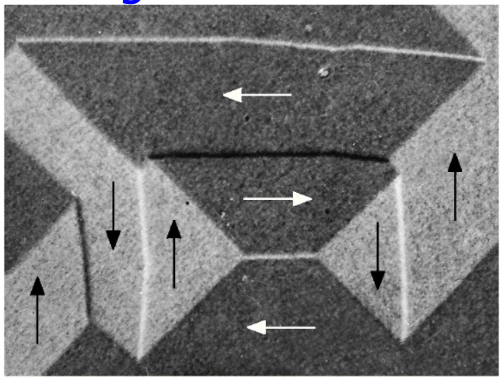
Very different from Faraday effect;
Voigt effect is "quadratic" (even) in M

Kerr effect



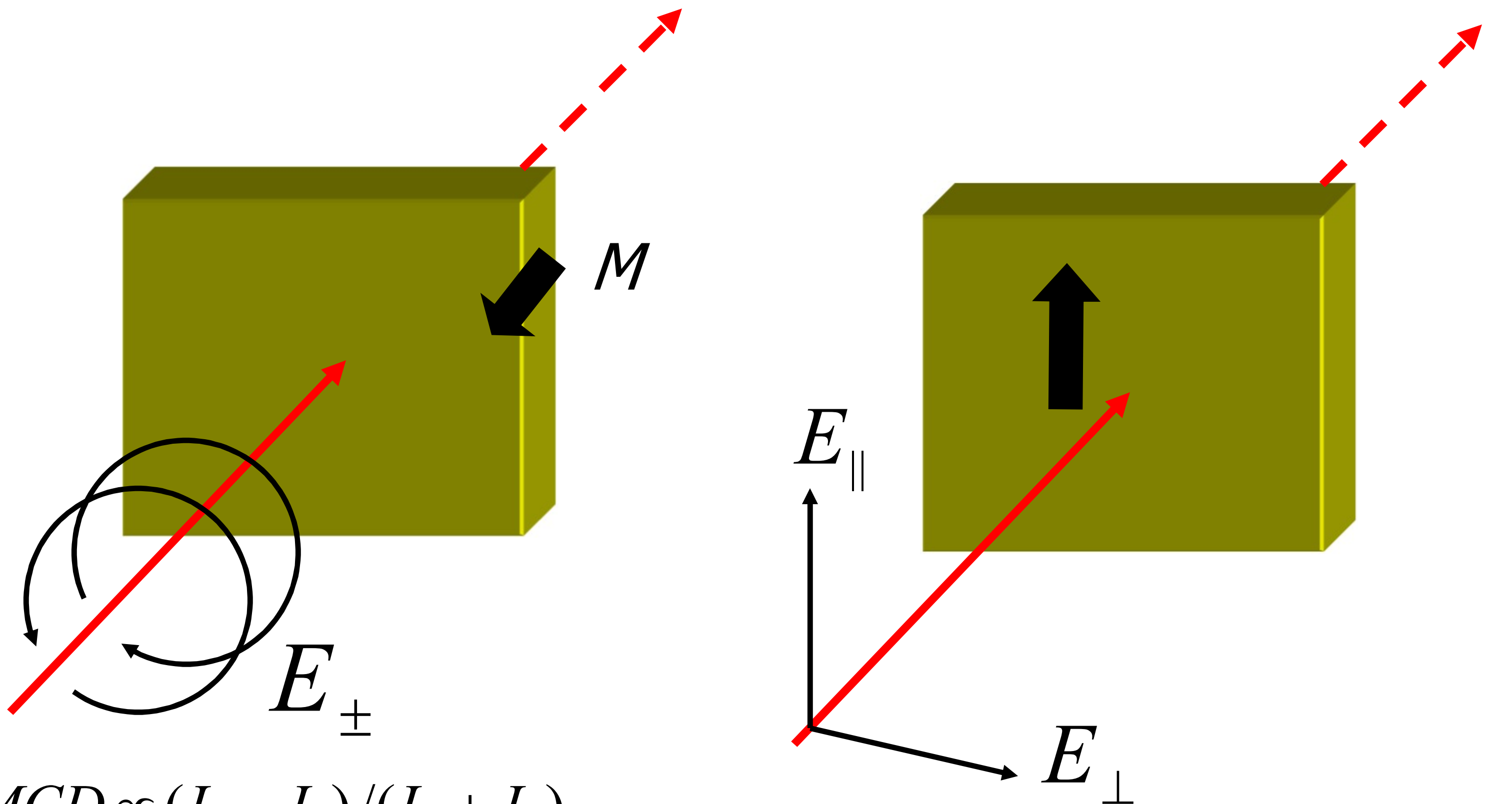

Imaging of magnetic domains using Voigt and Kerr effect in reflection

Voigt effect



(Courtesy R. Schäfer)

# Magnetic circular and linear dichroism




$$MCD \propto (I_+ - I_-)/(I_+ + I_-)$$

Magnetic Circular Dichroism

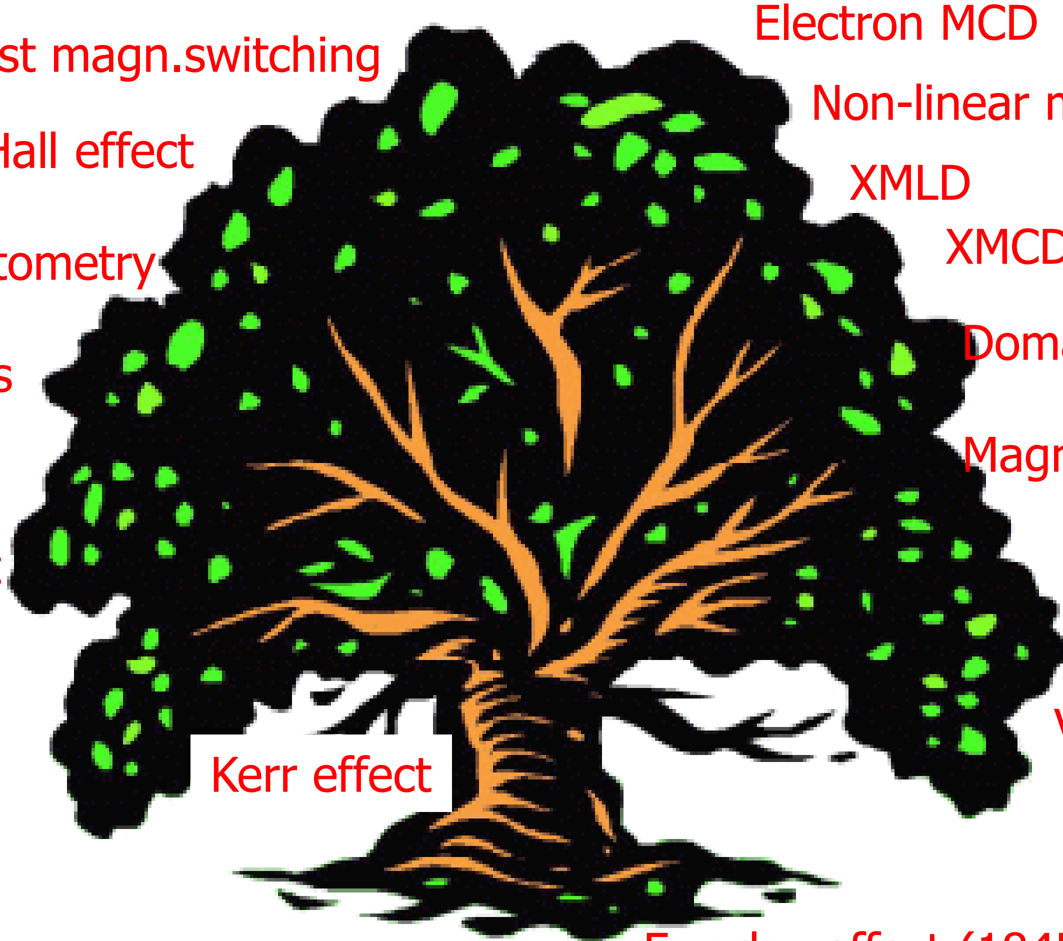
"odd in M"

$$MLD \propto (I_{||} - I_{\perp})/(I_{||} + I_{\perp})$$

Magnetic Linear Dichroism

"even in M"

# Development of light-magnetic material interaction

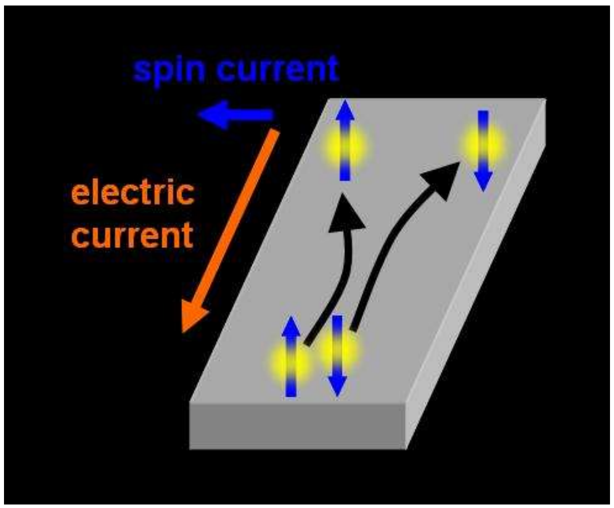


# Recent Examples of Light – Magnetism Interactions

## Observation of the spin Hall effect

Kato, Myers, Gossard & Awschalom,
Science **306**, 1910 (2004)

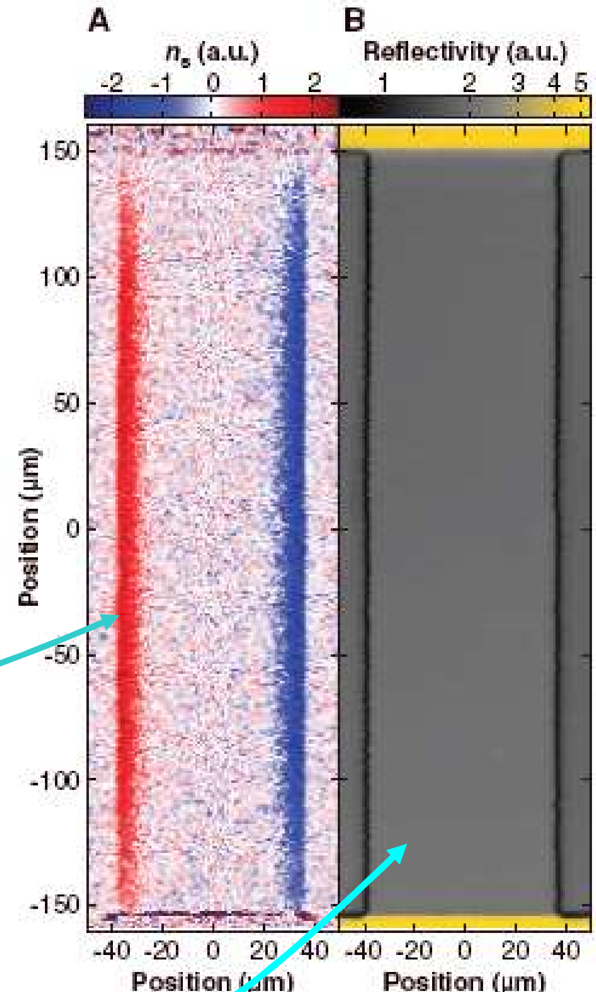
Spin Hall effect




Dyakonov & Perel, JETP Lett.
**13**, 467 (1971)

Kerr rotation image

Reflection image

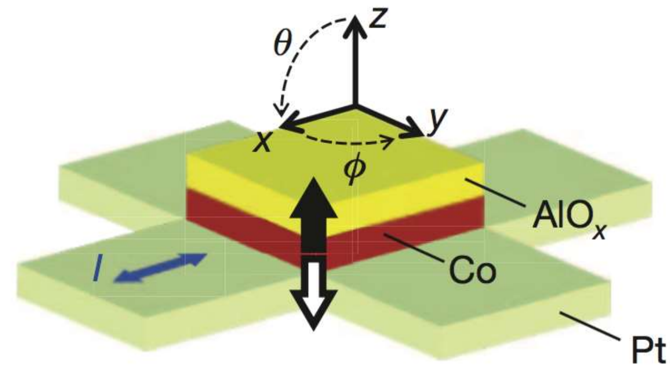



Material: non-magnetic n-GaAs [110]
MO Kerr rotation detection ~ $10^{-5}$ deg.

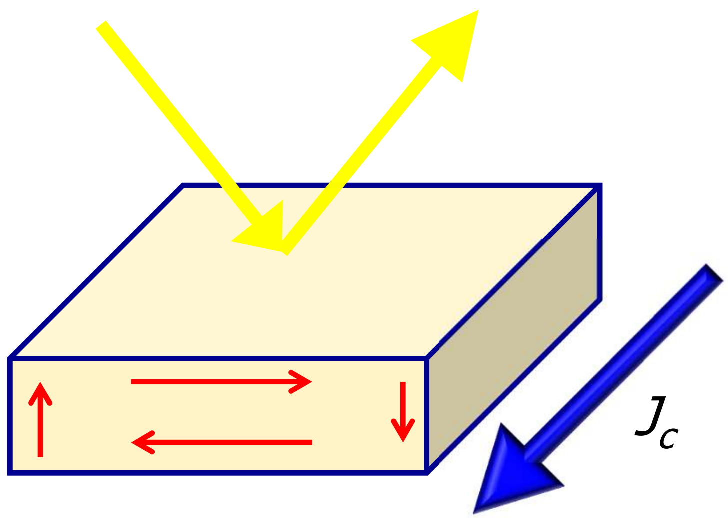
# Spin Hall effect in heavy metals

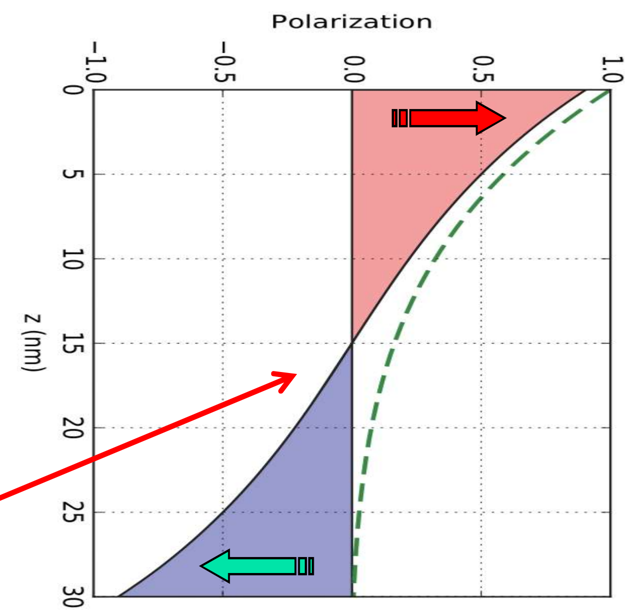
## Gives rise to spin-orbit torque

Direct observation of SHE in pure heavy-metal difficult because of short spin lifetime and spin diffusion length



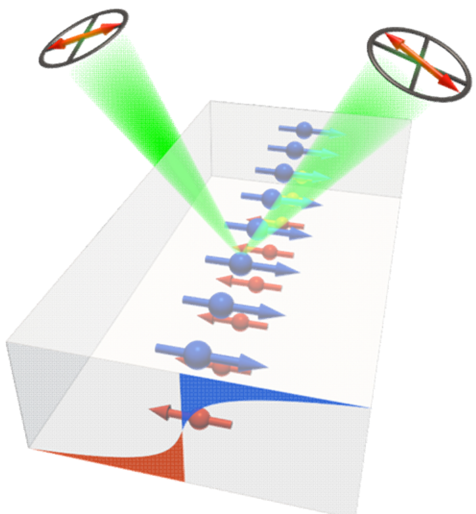

Miron et al, Nature **476**, 189 (2011)
Liu et al, Science **336**, 555 (2012)



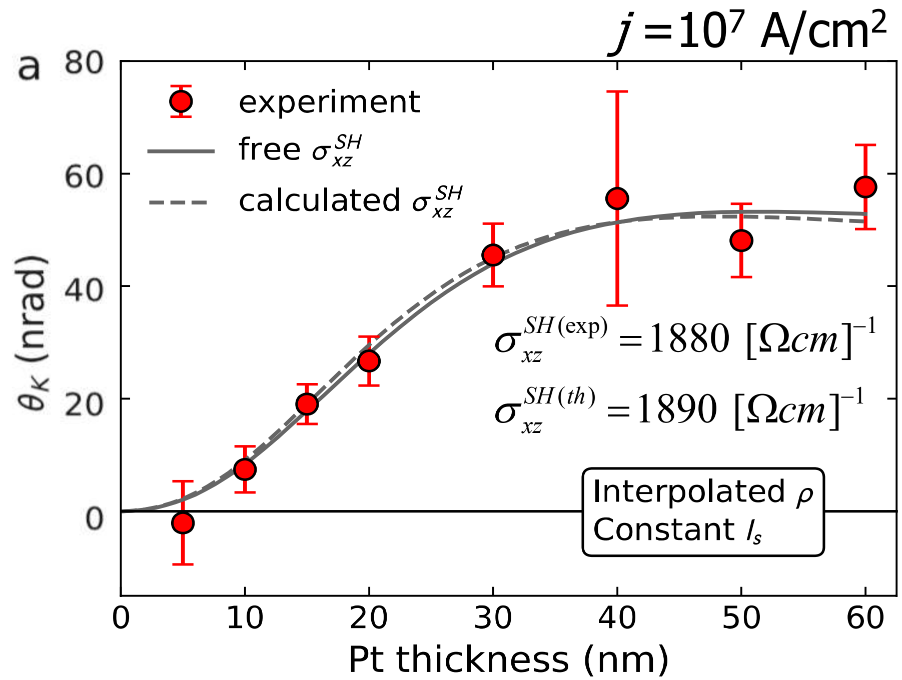




MOKE detection could be possible due to penetration depth

# Experimental direct observation of spin Hall effect Pt



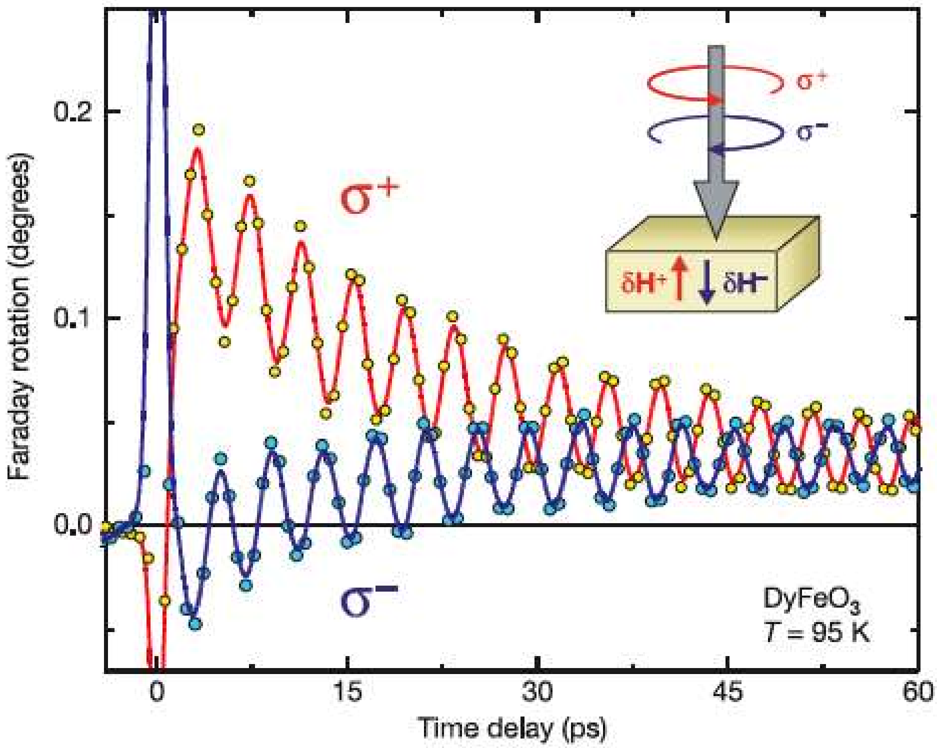
Stamm, Murer, Berritta, Feng, Gabureac, Oppeneer & Gambardella, PRL **119**, 087203 (2017)




Excellent agreement with experiment
Estimated $l_s$=11.4±2 nm for pure Pt

- Accurate MOKE measurements of SH conductivity in heavy metals feasible with nrad sensitivity

# Optically induced magnetization




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

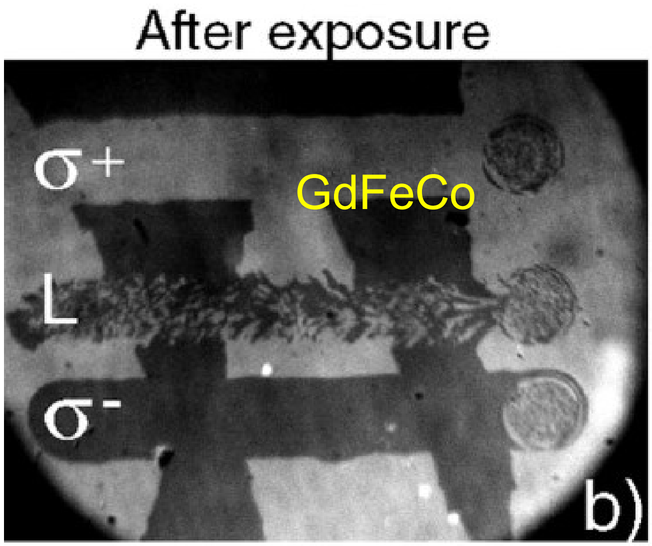
Due to nonlinear "opto-magnetic" effect, the inverse Faraday effect:

Induces magnetization δM

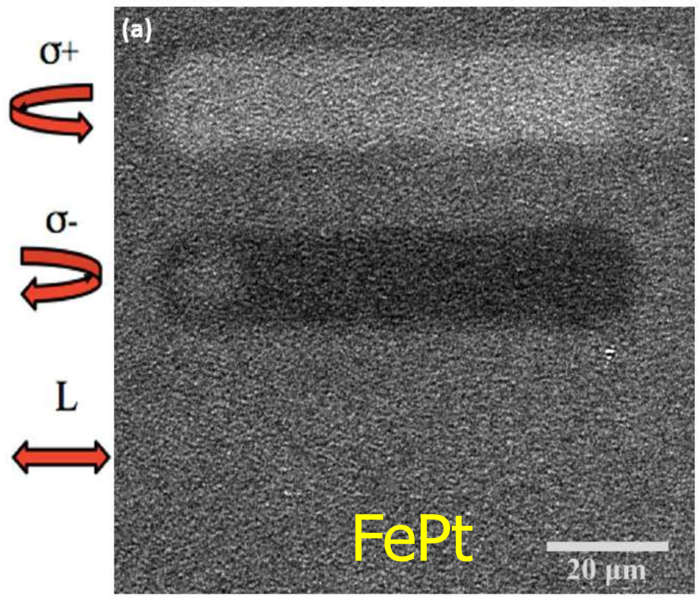
$$\vec{M}^{ind} \propto \nu_{IFE} \cdot E_i E_j^*$$

Could potentially lead to a fast, optically driven magnetization reversal

# All-optical writing of magnetic domains



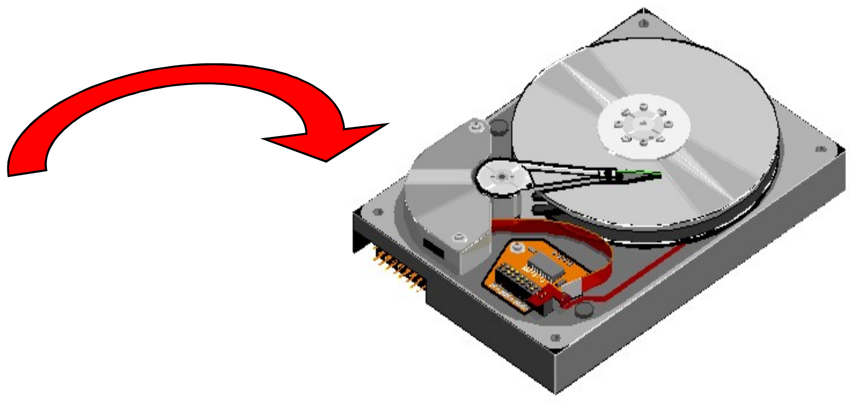

Stanciu et al, PRL **99**, 047601 (2007)




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

- Due to *inverse Faraday effect?*
- Background all-optical magn. recording
- Erasing & writing with fs-laser pulses
- Approx. $10^3$ times faster recording?

(symposium Th. Rasing, A. Kirilyuk)



# Ultrafast magnetism

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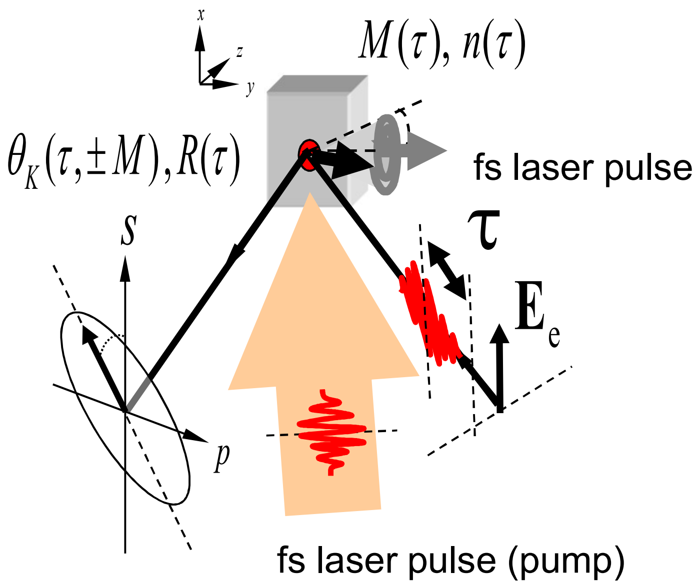
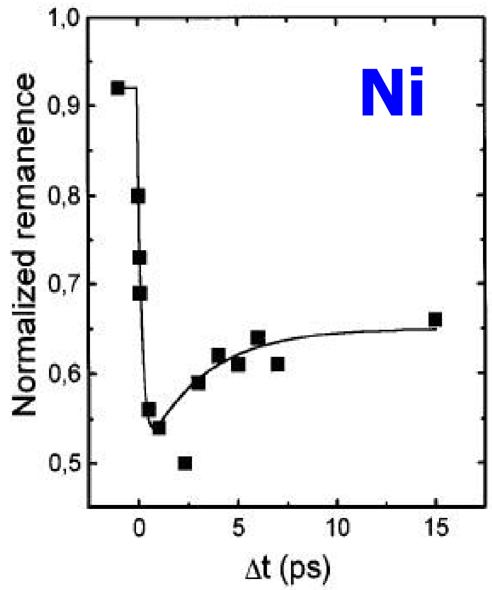
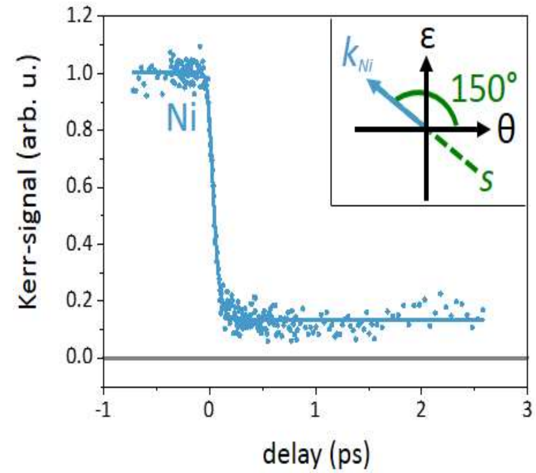
Measurement of ultrafast magnetic response with time-resolved magneto-optics




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




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

- Magnetization decay in <250 fs
- Very fast decay ~40 fs

# Theoretical description of light – magnetism interaction

Use 2nd level: Combination of Maxwell-Fresnel theory and *ab initio* quantum theory

Fresnel equation for modes in material:

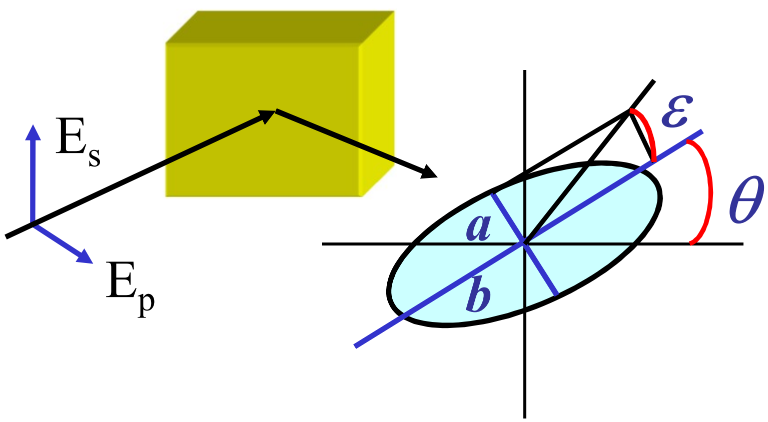
$$\left[n^2\mathbf{1} - \boldsymbol{\epsilon} - \boldsymbol{n} : \boldsymbol{n}\right] \cdot \boldsymbol{E} = 0$$

Geometry & materials´ boundary conditions:

$$\begin{pmatrix} E_s^r \\ E_p^r \end{pmatrix} = \begin{pmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{pmatrix} \bullet \begin{pmatrix} E_s^i \\ E_p^i \end{pmatrix}$$

$$\frac{E_s^r}{E_s^i} \equiv r_{ss} = \frac{n_i \cos\theta_i - n_t \cos\theta_t}{n_i \cos\theta_i + n_t \cos\theta_t}$$

$$\frac{E_s^t}{E_s^i} \equiv t_{ss} = \frac{2 n_i \cos\theta_i}{n_i \cos\theta_i + n_t \cos\theta_t}$$




Polarization analysis *or* intensity measurement

And: *ab initio* theory for calculation of $\varepsilon(\omega)$

# Dependence of the dielectric tensor on fields

The dielectric tensor depends on external fields $\overleftrightarrow{\varepsilon} \rightarrow \overleftrightarrow{\varepsilon}(\vec{k}, \omega, \vec{B}, \vec{E})$

Use a Taylor expansion for effects to lowest order: $(\vec{\mu} \rightarrow 1)$

$$\varepsilon(\vec{k}, \vec{B}, \vec{E}, \omega) \approx$$

$$\varepsilon_0 + O(\vec{k}) + O(\vec{B}) + O(\vec{E}) + O(B_i E_j) + O(B_i B_j) + O(E_i E_j) + \ldots$$

Natural optical activity

Magneto-optical effects

Electro-optical effects

Magneto-electric effects

Magneto-optical effects

Optomagnetic effects

All the (linear) phenomena can be described, using the Fresnel formalism

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# Magnetic effects in Fresnel equations

**Typical ε tensor:**

$$\overleftrightarrow{\varepsilon} = \begin{pmatrix} \varepsilon_{\perp} & \varepsilon_{xy} & 0 \\ \varepsilon_{yx} & \varepsilon_{\perp} & 0 \\ 0 & 0 & \varepsilon_{\parallel} \end{pmatrix}$$

**Onsager relations** → Magnetic parity

$$\varepsilon_{xy}(-H,\omega) = -\varepsilon_{xy}(H,\omega)$$ → odd

$$\varepsilon_{\alpha\alpha}(-H,\omega) = +\varepsilon_{\alpha\alpha}(H,\omega)$$ → even

→ Magn. effects probe always $\sim M$ or $\sim M^2$ (to lowest order)

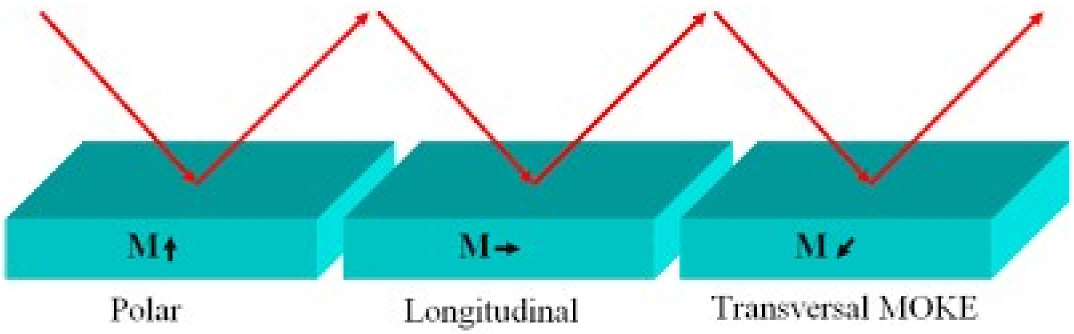
Examples:

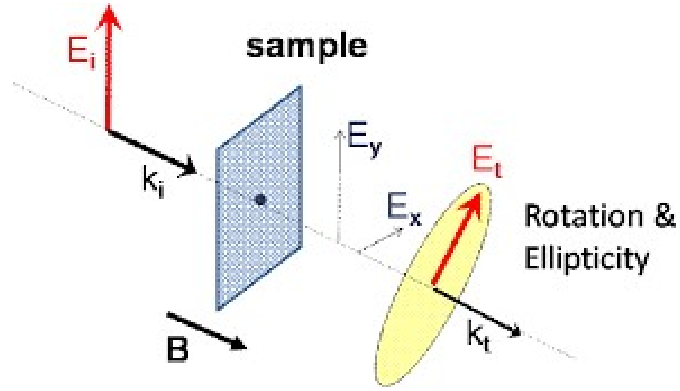
$$\mathrm{MCD} = \frac{\omega d}{2c}\mathrm{Re}\left[\frac{\varepsilon_{xy}}{n}\right]$$ Odd in M;

$$\mathrm{MLD} = \frac{\omega d}{2cn}\mathrm{Im}\left[\varepsilon_{\parallel} - \varepsilon_{\perp} - \frac{\varepsilon_{xy}^2}{\varepsilon_{\perp}}\right]$$ even

$$\varepsilon_{\alpha\alpha} \approx \varepsilon_{\alpha\alpha}^0 + \varsigma_{\alpha} M_{\alpha}^2$$

# Magneto-optical Kerr and Faraday effects




polar Kerr effect, normal incidence

$$\left(\frac{1+\tan\epsilon_K}{1-\tan\epsilon_K}\right)e^{-2i\theta_K} = \frac{1+n_+}{1-n_+}\,\frac{1-n_-}{1+n_-}$$

Faraday effect, normal incidence

$$\left(\frac{1-\tan\varepsilon_F}{1+\tan\varepsilon_F}\right)e^{2i\theta_F} = e^{i\omega d_t(n_+ - n_-)/c}$$

Use $n_\pm^2 = \varepsilon_{xx} \pm i\varepsilon_{xy}$ Assume $|\varepsilon_{xy}| << |\varepsilon_{xx}|$ or $|\sigma_{xy}| << |\sigma_{xx}|$

$$\theta_K + i\varepsilon_K = \frac{-\sigma_{xy}(\omega)}{\sigma_{xx}(1+4\pi i\sigma_{xx}/\omega)^{1/2}}$$

$$\theta_F + i\varepsilon_F = \frac{2\pi d}{c}\,\frac{\sigma_{xy}(\omega)}{(1+4\pi i\sigma_{xx}/\omega)^{1/2}}$$

# Classification of magnetic spectroscopies

## Linear (odd) in M  spectroscopies:

| | Polarization analysis | Intensity | |
|---|---|---|---|
| Transmission | Faraday L | MCD | C |
| Reflection | P-MOKE<br>L-MOKE L | T-MOKE<br>RMS | C L |
| | 2 quantities | 1 quant. | |

**Classification criteria:**

1. Magnetic parity
2. Transmission or reflection
3. Polarization or intensity
4. Linearly or circ. polarized light

➡ Suitable for (element-selective) study of ferro-, ferrimagnets

# Even-in-*M* magnetic spectroscopies

## Quadratic (even) in M spectroscopies:

| | Polarization analysis | Intensity |
|---|---|---|
| Transmission | Voigt L | MLD L |
| Reflection | birefringence R-Voigt L | R-MLD L |
| | 2 quantities | 1 quant. |

Suitable for (element-selective) study of antiferromagnets (and ferromagnets as well)

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# Linear-response theory

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

Lifetime broadening,
$1/\tau \sim 0.4$ eV

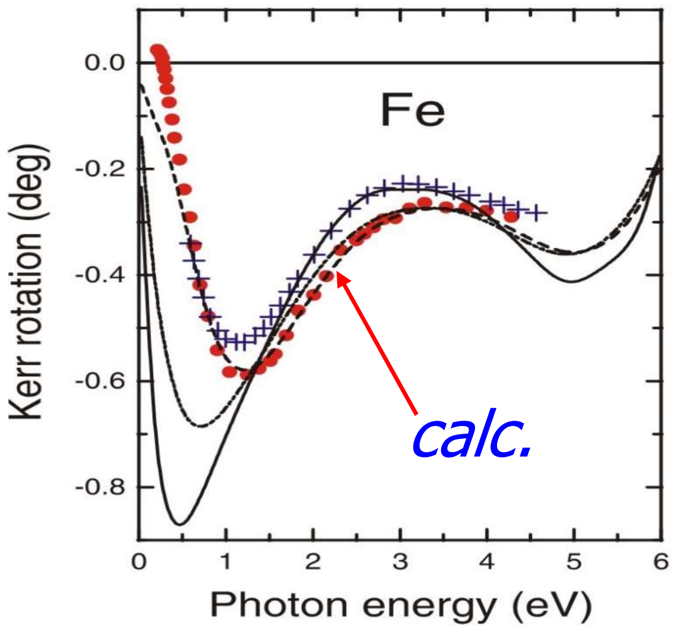
$$\mathrm{Im}[\varepsilon_{xx}(\omega)] = \frac{4\pi^2 e^2}{m^2 V\hbar\omega^2}\sum_{\substack{n \\ un.}}\sum_{\substack{n' \\ occ.}}\mathrm{Re}\{\Pi^{x}_{n'n}\Pi^{x}_{nn'}\}\delta(\omega-\omega_{nn'})$$

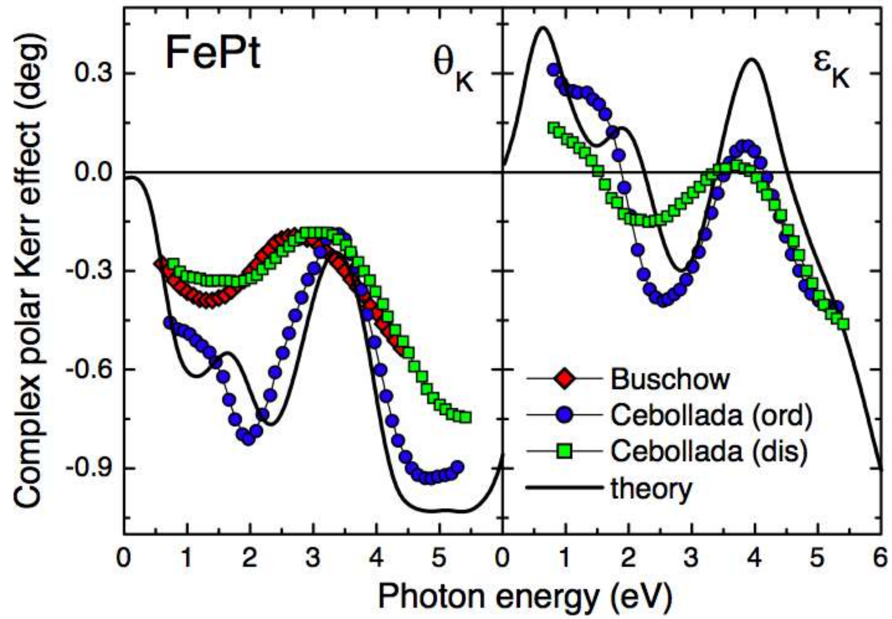
$$\mathrm{Re}[\varepsilon_{xy}(\omega)] = \frac{4\pi^2 e^2}{m^2 V\hbar\omega^2}\sum_{\substack{n \\ un.}}\sum_{\substack{n' \\ occ.}}\mathrm{Im}\{\Pi^{x}_{n'n}\Pi^{y}_{nn'}\}\delta(\omega-\omega_{nn'})$$

(for $1/\tau$ -> 0)

Lifetime broadening happens and needs to be taken into account

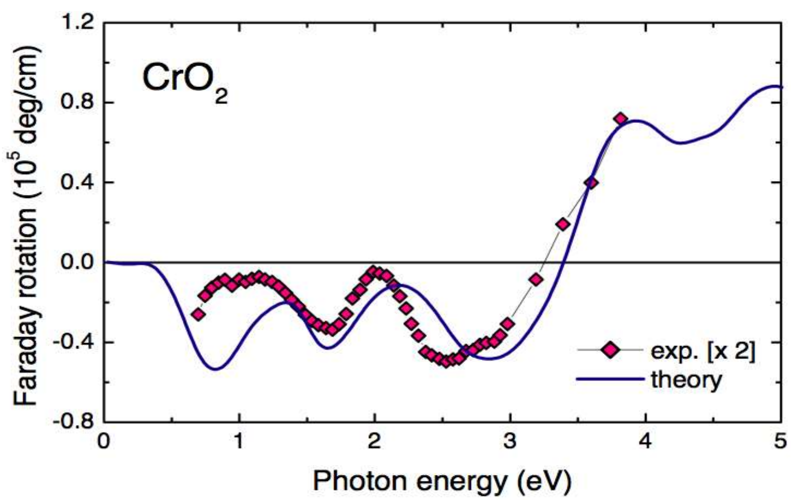
# Lifetime broadening – linear magneto-optics




Optical frequencies:
lifetime $\Gamma=1/\tau$ ≈ 0.03 Ry

Oppeneer, Handbook of Magnetic Materials, Vol. 13 (2001)



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# Origin of magneto-optical effects

Effective Kohn-Sham Hamiltonian:

Exchange field

$$\hat{H} = \left[ -\frac{\nabla^2}{2m} + V_{e,N}(\vec{r}) + V_0(\vec{r}) \right] 1 + \vec{B}_{xc}(\vec{r}) \cdot \hat{\vec{\sigma}} + \xi \hat{\vec{\ell}} \cdot \hat{\vec{\sigma}}$$

Spin-orbit coupling

Spin-density (2x2):

$$n(\vec{r}) = \{ n_0(\vec{r}) 1 + \vec{m}(\vec{r}) \cdot \vec{\sigma} \} / 2 \qquad \begin{cases} n(\vec{r}) = n_\uparrow(\vec{r}) + n_\downarrow(\vec{r}) \\ m(\vec{r}) = \mu_B \{ n_\uparrow(\vec{r}) - n_\downarrow(\vec{r}) \} \end{cases}$$

Vary the two magnetic interactions (exchange & spin-orbit) to deduce how magnetic spectra depend on these.

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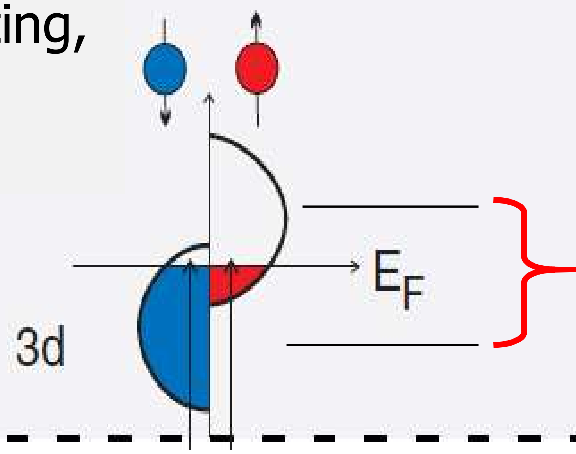
# Effect of SOI and exchange interaction

Full form of SOI: $H_{SO} = \frac{e^2\hbar}{4m^2c^2}\left(\frac{1}{r}\frac{dV}{dr}\right)\vec{L}\cdot\vec{\sigma} \cong \xi(\vec{L}\cdot\vec{S})$

(small relativistic effect)

Exchange splitting,
1 – 2 eV
(3d atom)

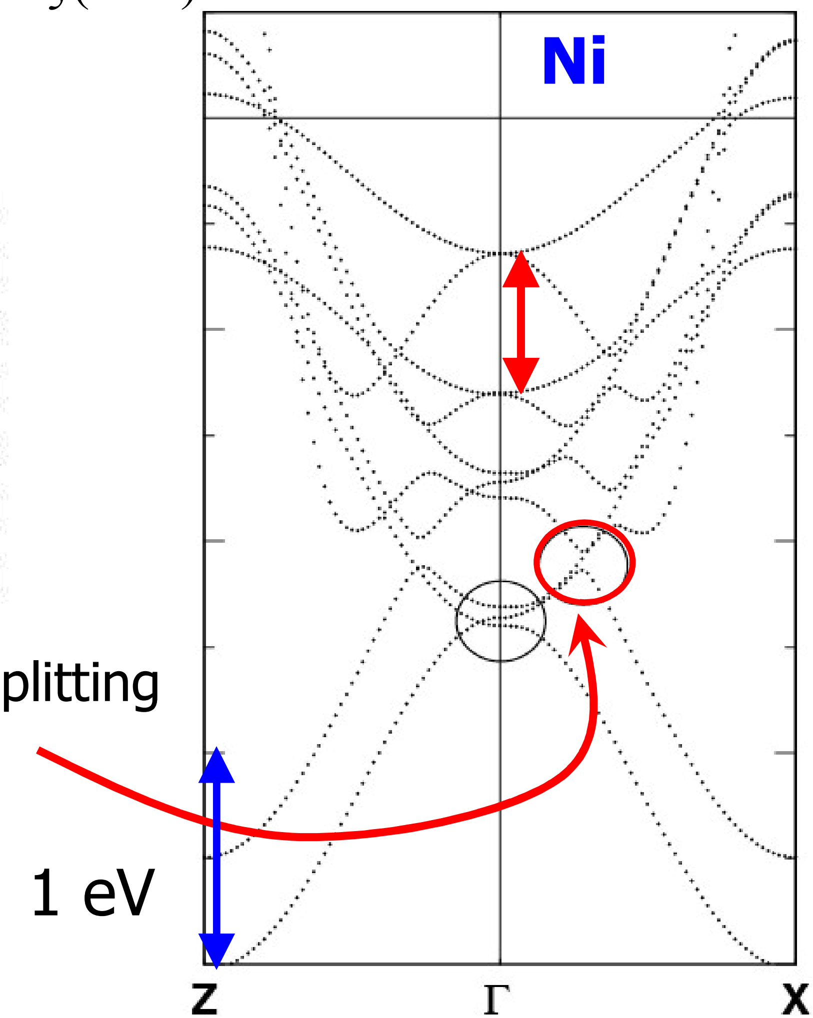



$$\Delta_{ex} >> \Delta_{SOC}$$

Spin-orbit coupling breaks crystal symmetry

Spin-orbit splitting
~20 meV

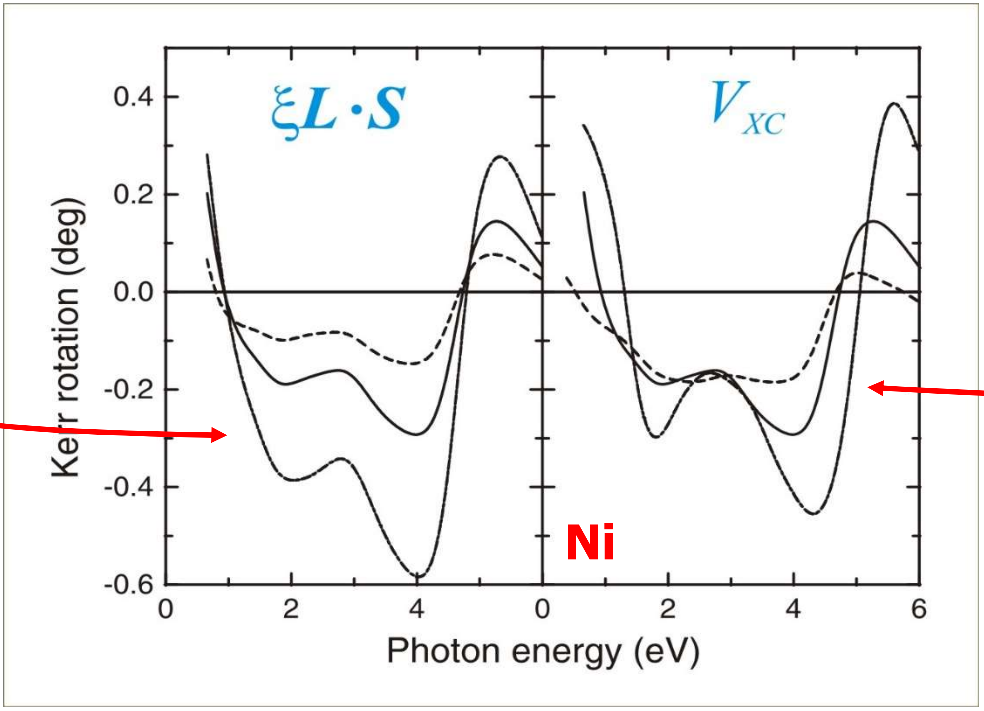
(b) With SOI



# Leading order quantity: spin-orbit coupling

Leading quantity determining the valence band MO effect is spin-orbit coupling $\xi$ (**L.S**)
$\Rightarrow$ Kerr and Faraday effect scale linear in the SOC, not in the exc.-splitting!

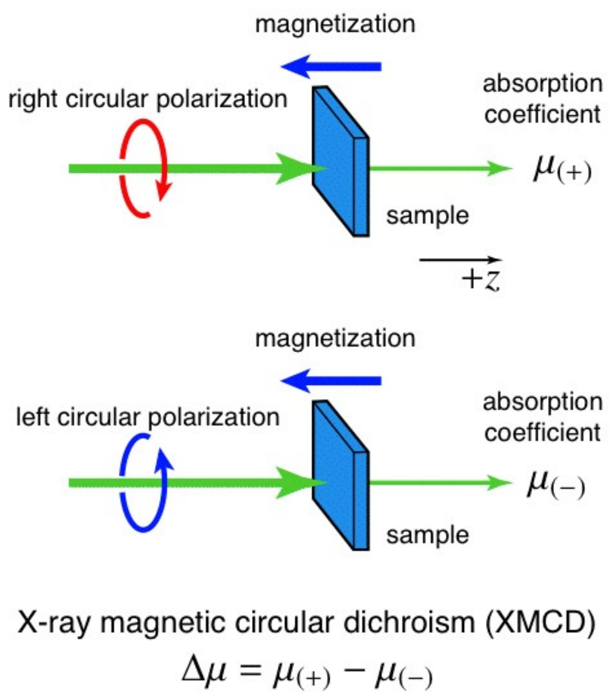
Scaling of SOI

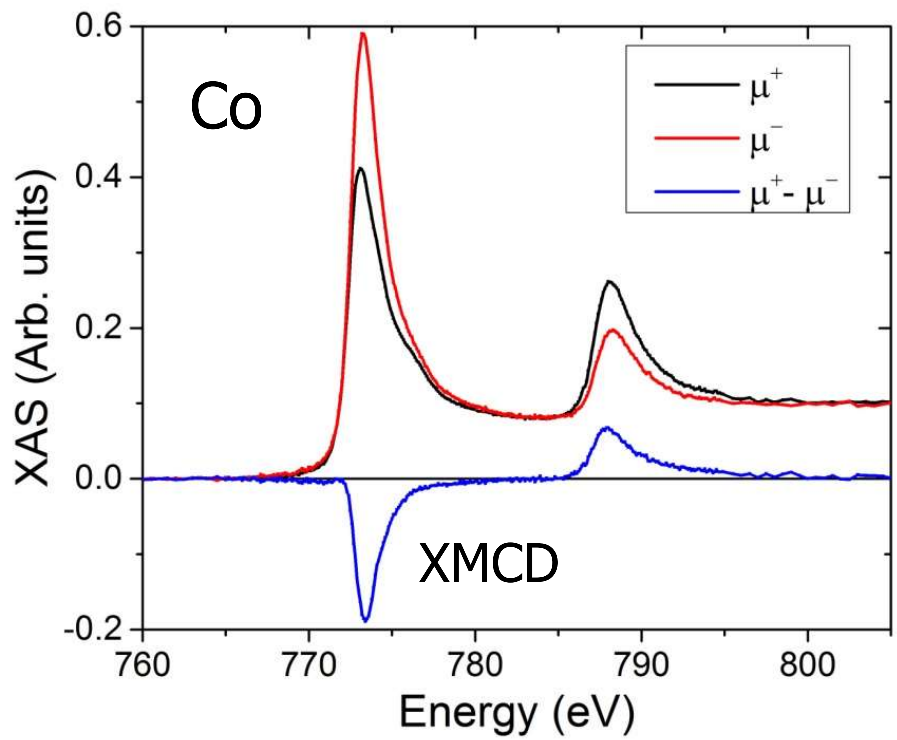



Scaling of exc.int.

# What about the X-ray regime ?

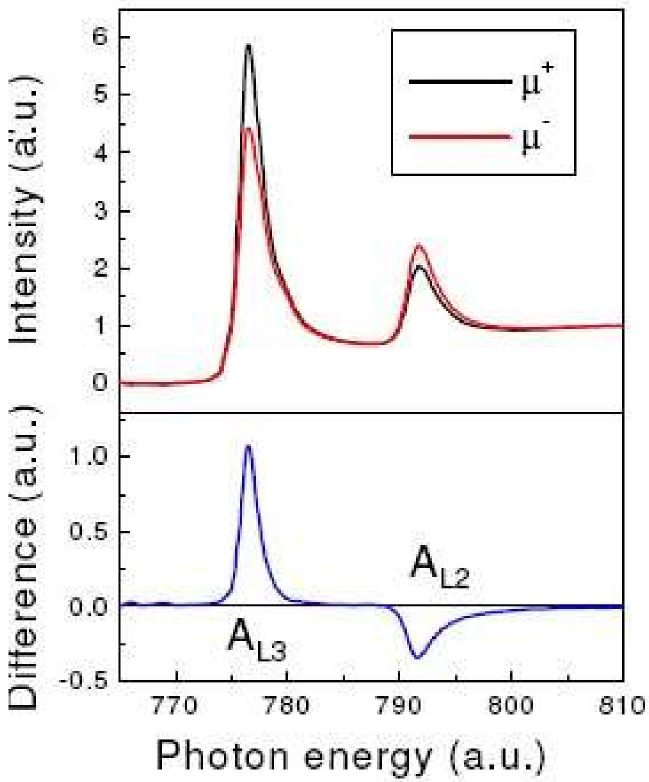
X-ray magnetic circular dichroism $\vec{E}_{\pm}(z,t) \propto (\vec{e}_x \pm i\vec{e}_y) \cdot e^{i\omega/c(n_{\pm}\cdot z)-i\omega t}$




Understand origin of and perform *ab initio* calculations for XMCD & XMLD at L-edges

# Note on importance of XMCD – sum rules




Thole et al, PRL **68**, 1943 (1992)
Carra et al, PRL **70**, 694 (1993)

**Sum rules**

$$m_s/\mu_B \sim \frac{A_{L3} - 2A_{L2}}{A_{iso}}$$ ⇐ Atomic spin moment

$$m_l/\mu_B \sim \frac{A_{L3} + A_{L2}}{A_{iso}}$$ ⇐ Atomic orbital moment

The XMCD sum rules are not exact but are intensively used, because they allow an element-selective determination of the spin & orbital moment on a 3d element in a material.

(Lecture E. Goering)

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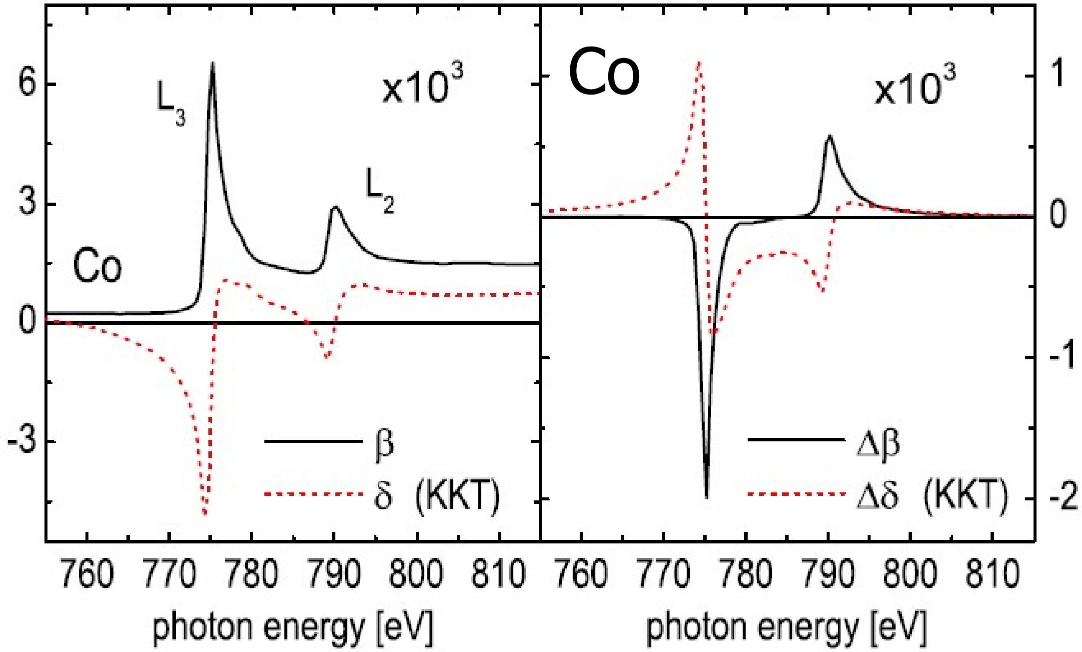
# Definition of refractive index (X-ray regime)

We had: $n_{\pm}^2 = \varepsilon_{xx} \pm i\varepsilon_{xy}$ (nonmagnetic: $n^2 = \varepsilon_{xx}$ )

In x-ray regime: $n_{\pm} = 1 - (\delta \pm \Delta\delta) + i(\beta \pm \Delta\beta)$

Small quantities!




These quantities can be obtained from XAS, XMCD & Faraday effect measurements

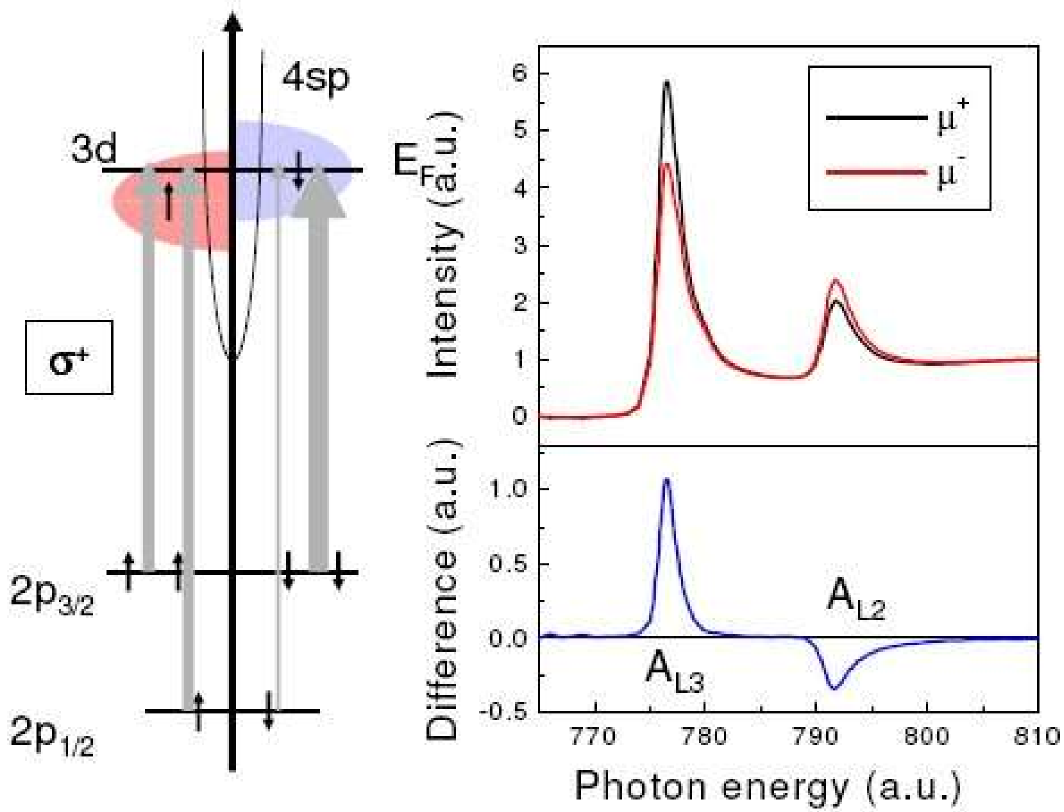
# Basic electronic structure picture

1) Spin-splitting of 3d states due to exchange interaction

2) Helicity-dependent optical selection rules

$$left: \quad \Delta m_\ell = -1, \quad \Delta m_s = 0$$

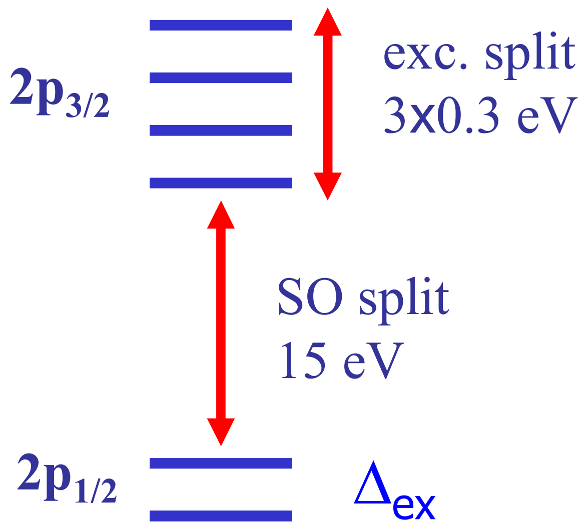
$$right: \Delta m_\ell = +1, \quad \Delta m_s = 0$$




Leads to different absorption of left/right circ. pol. radiation (trans. probabilities)

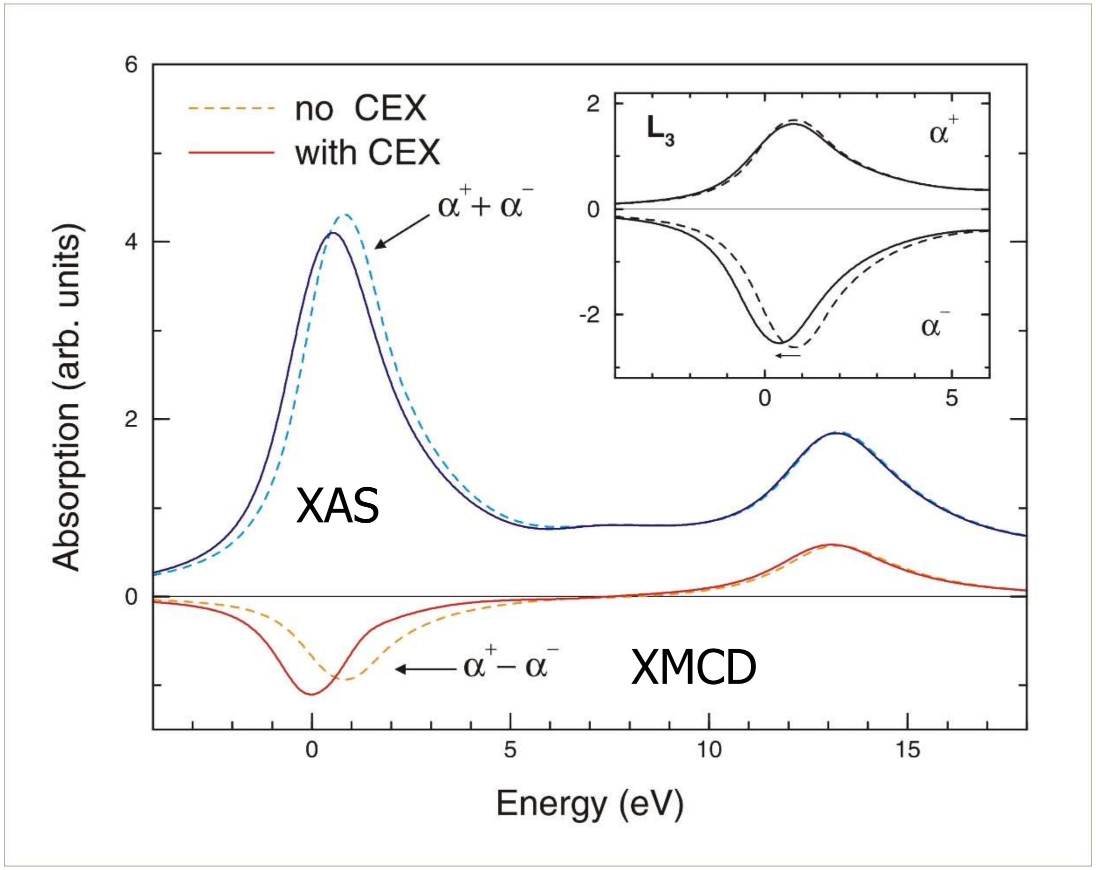
# *Ab initio* calculated XMCD spectra - Effect of $\Delta_{xc}$

Very different size of SOI and $\Delta_{xc}$ !




$$\mathbf{XMCD} \approx \mathrm{Im}[\varepsilon_{+} - \varepsilon_{-}]$$

To lowest order the XMCD does not depend on $\Delta_{ex}$:

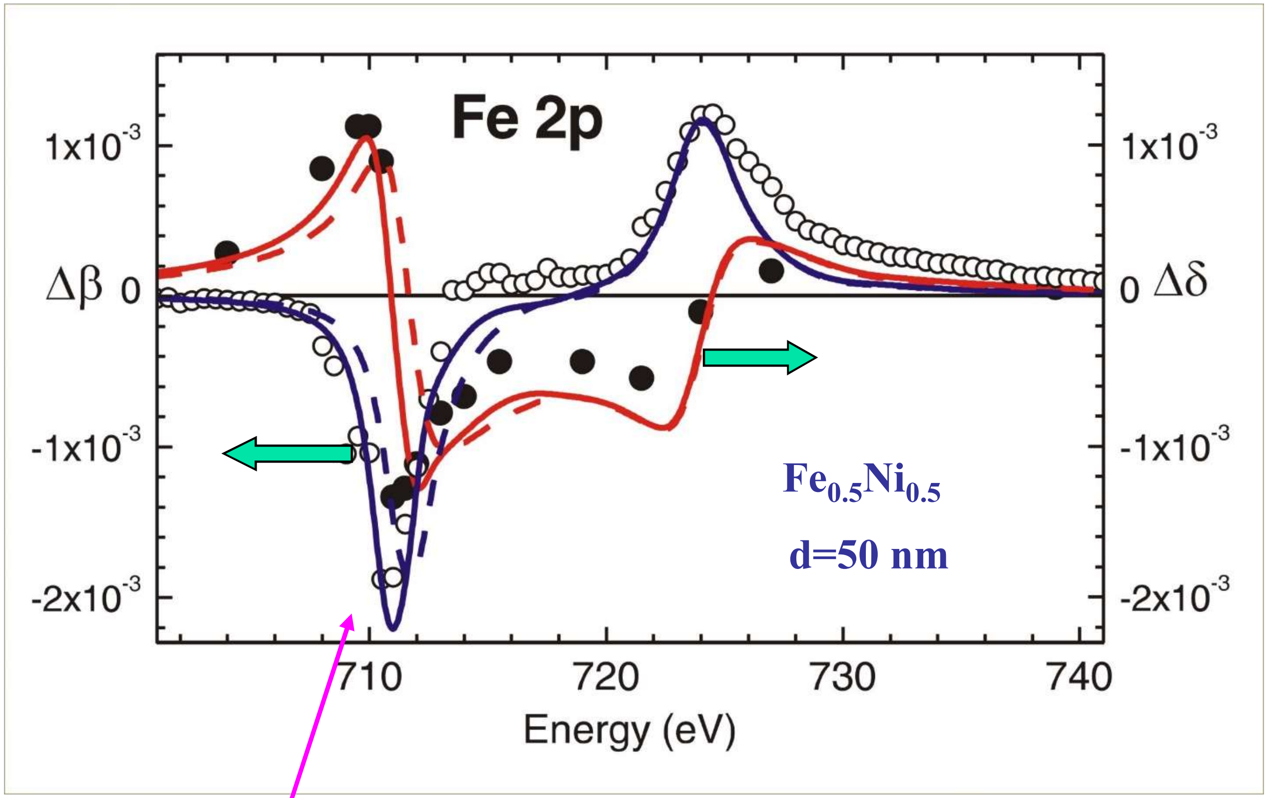



Many calculations ignore $\Delta$, but there is a small effect !

Kunes et al, PRB **64**, 174417 (2001)

# Comparison with experimental XMCD spectra

XMCD




$$n_{\pm} = 1 - (\delta \pm \Delta\delta) + i(\beta \pm \Delta\beta)$$

Exchange-split core states give somewhat better results when compared to experimental spectra !

Kunes et al, PRB **64**, 174417 (2001)

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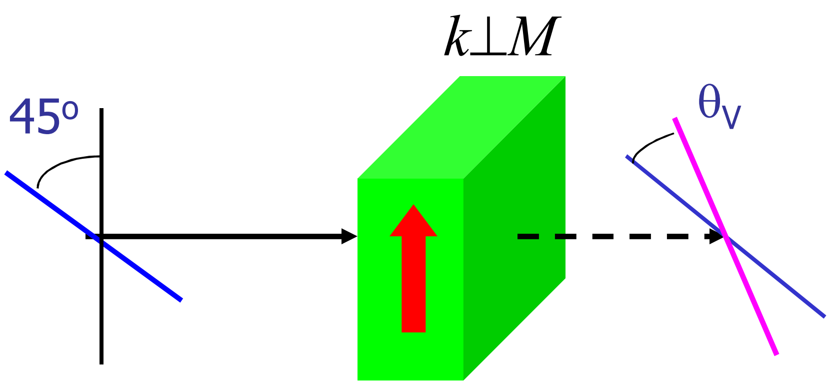
# Quadratic in *M* effects: X-ray Voigt effect or XLMD

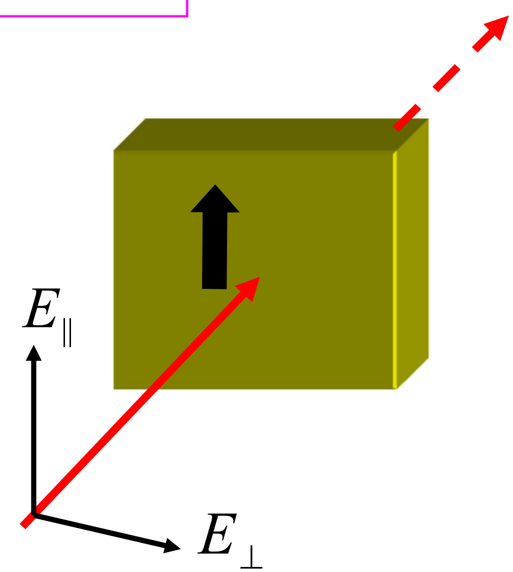
Voigt effect

$$\theta_V + i\varepsilon_V \approx \frac{\omega d}{2ic}\left[n_{\parallel} - n_{\perp}\right] = \frac{\omega d}{2cn}\mathrm{Im}\left[\varepsilon_{\parallel} - \varepsilon_{\perp} - \varepsilon_{xy}^2/\varepsilon_{\perp}\right]$$

$$2\theta_V \cong LMD$$




Voigt effect

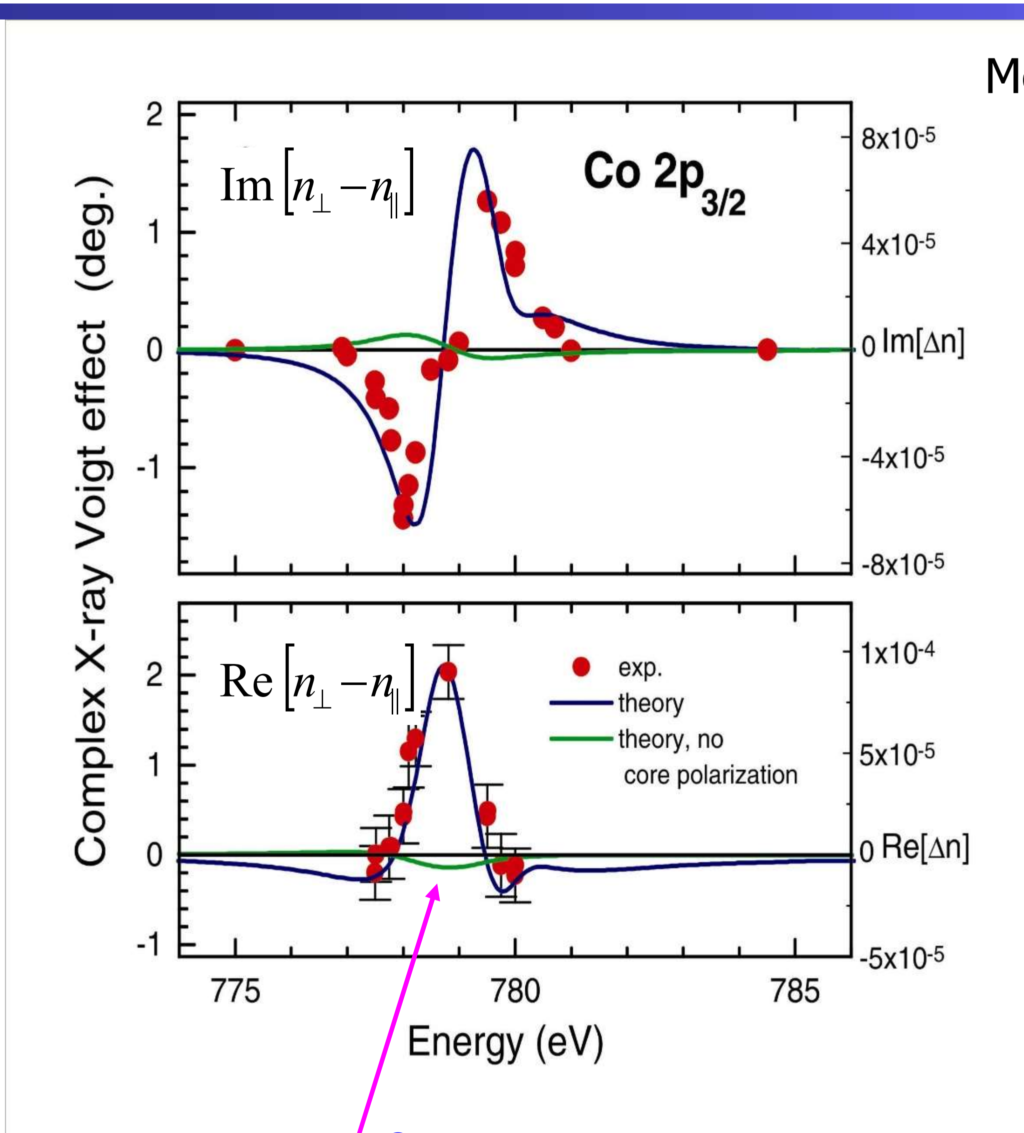



$$MLD \propto (I_{\parallel} - I_{\perp})/(I_{\parallel} + I_{\perp})$$

Origin not understood ...
Magnetovolume effect?
Why much smaller than MCD?
Spin-orbit interaction? $n_{\parallel} \neq n_{\perp}$
Cf. Faraday, Kerr: linear in $\xi_{SO}$

# Measurements & *ab initio* calculations XMLD

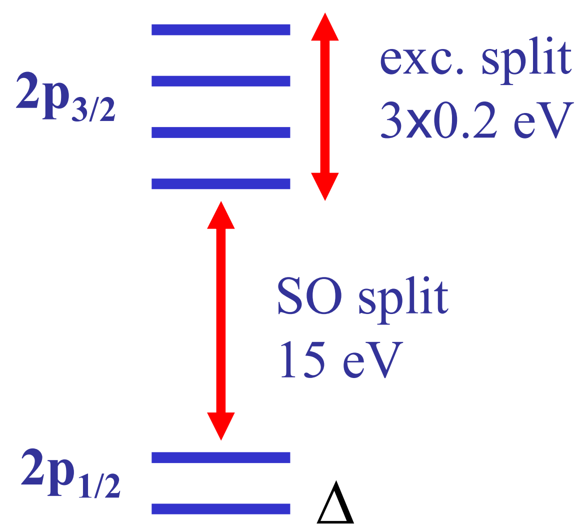
Mertins et al, PRL **87**, 47401 (2001)



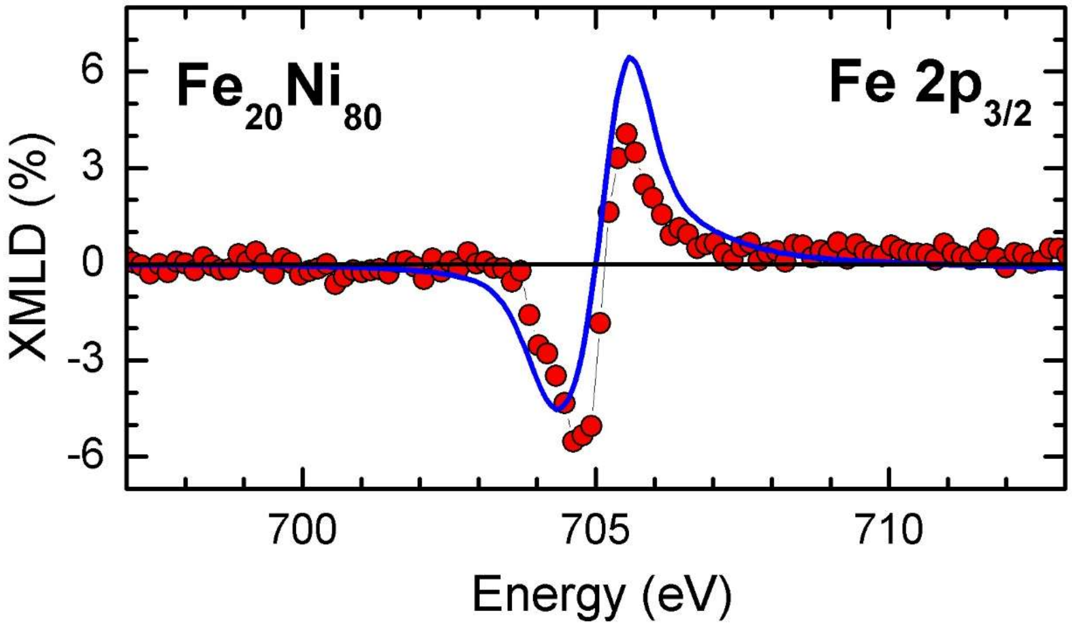

$\Delta_{ex} = 0$

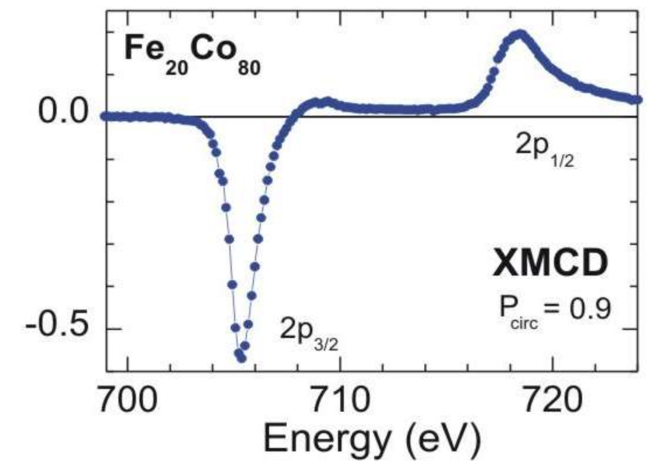
XMLD

XMLD / X-ray Voigt effect would be very small without exchange split core states!



# Further results of *ab initio* calculations XMLD




- small effect ~ 5%
- good *ab initio* theory

$$A_{XMLD} \approx \frac{\omega d}{c}\mathrm{Im}\left[n_{\parallel} - n_{\perp}\right] \approx \frac{\omega d}{2cn}\mathrm{Im}\left[\varepsilon_{\parallel} - \varepsilon_{\perp}\right]$$

Why do we have these spectral structures?

# Simple model for X-ray magnetic spectroscopies

Core-states are ($k$) dispersionless

$$\epsilon_{xy}^{(1)}(\omega) = \frac{4\pi^2 e^2}{\hbar V_{uc}} \mathrm{Im} \sum_{k} \sum_{\substack{c \ \text{occ.} \\ n \ \text{un.}}} r_{nc}^{x}(k) r_{cn}^{y}(k)\, \delta(\omega - \omega_{nc}(k))$$

Expand $\varepsilon_{xy}$ considering 2p -> 3d transitions

Selection rules on m :

$$\begin{cases} \varepsilon_{+} = \varepsilon_{xx} + i\varepsilon_{xy} \Rightarrow \Delta m = +1 \\ \varepsilon_{-} = \varepsilon_{xx} - i\varepsilon_{xy} \Rightarrow \Delta m = -1 \\ \varepsilon_{0} = \varepsilon_{\parallel} \Rightarrow \Delta m = 0 \end{cases}$$

X-ray spectroscopies:

**XAS** $\approx \mathrm{Im}[2\varepsilon_{\perp} + \varepsilon_{\parallel}]/3$

→ **Sum of all transitions Δm**

**XMCD** $\approx \mathrm{Im}[\varepsilon_{+} - \varepsilon_{-}]$

→ **Difference of transitions with Δm=+1 and Δm=-1**

**XMLD**

$\approx \mathrm{Im}[\varepsilon_{\parallel} - \varepsilon_{\perp}] = \mathrm{Im}[\varepsilon_{\parallel} - (\varepsilon_{+} + \varepsilon_{-})/2]$

→ **Difference of transitions with Δm=0 and aver. Δm=+1 & -1**

# Understanding the shape of XMLD and XMCD

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Develop model theory and perform ab initio calculations

**Model theory (2p core):**

- Neglect SO in valence states (~ meV)
- Consider only 2p ->3d transitions
- Expand ε functions with respect to $\Delta_{ex}$

$$\mathrm{Im}[\varepsilon_\mu(\omega)] \propto \sum_{m,s} a^{\mu}_{\gamma,s}(j) D_{ms}\left(\omega \pm \frac{\gamma\Delta}{2}\right), \qquad \gamma/2 = m - \mu + s$$

$D_{ms}$ m- and spin-dependent 3d partial DOS

$\Delta_{SO}$ (15 eV) >> $\Delta_{ex}$ (1-3 eV) >> $\Delta_{ex}$ (0.1-0.3 eV) > $\xi_{so\text{-}v}$ (0.09 eV)

$$\textbf{XMLD} \approx \mathrm{Im}[\varepsilon_{\parallel} - \varepsilon_{\perp}] = \mathrm{Im}[\varepsilon_{\parallel} - (\varepsilon_{+} + \varepsilon_{-})/2]$$

**Difference of transitions with Δm=0 and aver. Δm=+1 & -1**

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# Model theory, results

$D_{ms}$ : m and s-dependent partial DOS of unoccupied 3d states

**XAS**
$$\approx 4\sum_m (D_{m\uparrow} + D_{m\downarrow}) \qquad j = 3/2$$
$$\approx 2\sum_m (D_{m\uparrow} + D_{m\downarrow}) \qquad j = 1/2$$

**XMCD**
$$\approx +2\sum_m (D_{m\uparrow} - D_{m\downarrow}) \qquad j = 3/2$$
$$\approx -2\sum_m (D_{m\uparrow} - D_{m\downarrow}) \qquad j = 1/2$$

**m-orbital degeneracy:**

**XMLD**
$$\approx \Delta \frac{d}{dE}\left(D_{\uparrow} - D_{\downarrow}\right) \qquad j = 1/2,\ 3/2$$
$$\approx \mp\Delta \frac{d}{dE}\left\{XMCD\right\}$$

→ **XAS branching ratio: 2/3**
**no magnetism (M invariant)**

→ **$L_2$ , $L_3$ equal & opposite**
**odd in M, no f.o. effect of Δ**

→

- **Small signal, proportional Δ !**
- **related to energy deriv. XMCD**
- **even in M (Δ is odd, *D* is odd)**

Absence of the crystal field

# Experimental check of XMCD-XMLD relation

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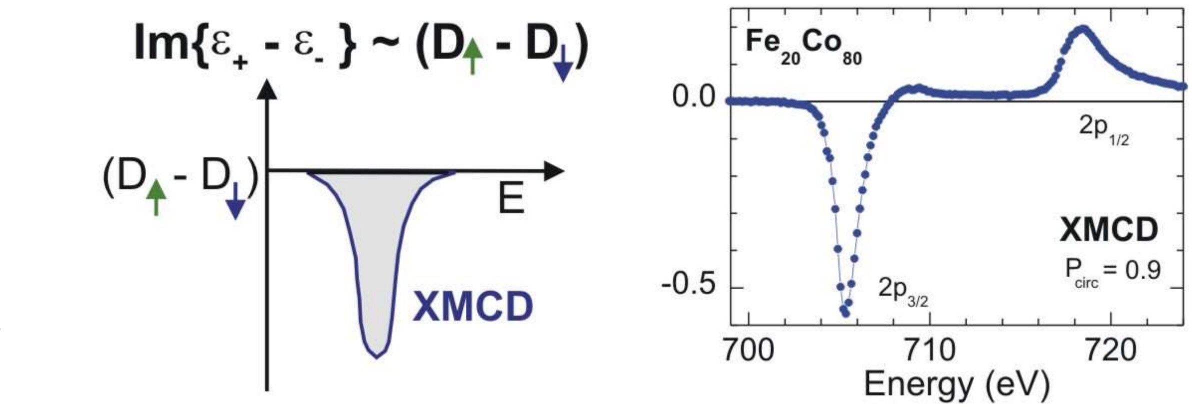
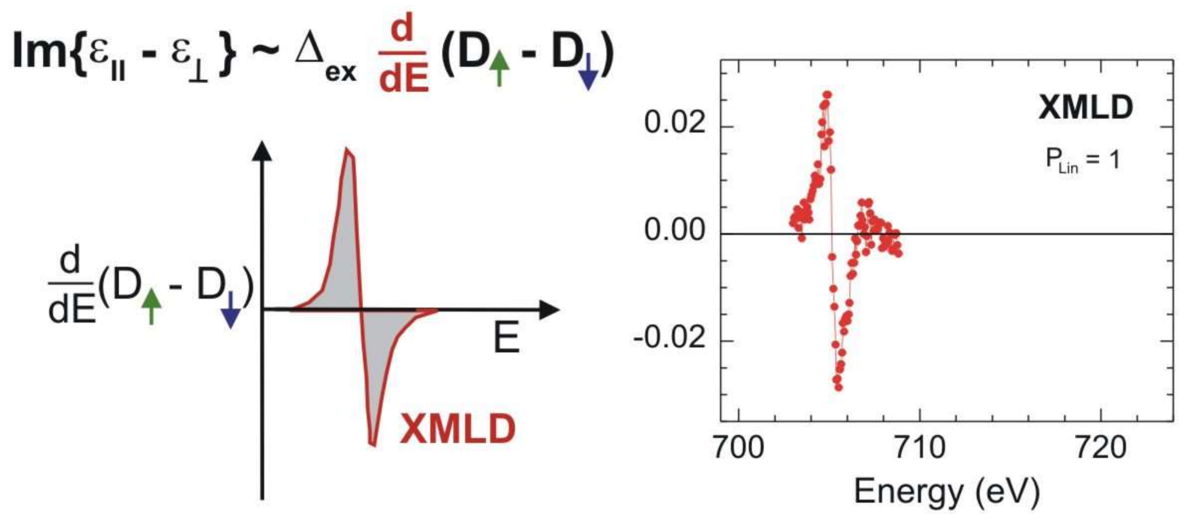
m-orbital degeneracy:
No crystal field,
amorphous FeCo alloy

Spin-pol. unocc. 3d DOS

$$\left(D_{\uparrow} - D_{\downarrow}\right)$$




Leading quantity $\Delta_{ex}$
is very small, yet crucial!

→ relation between
XMCD-XMLD verified

Kunes et al, JMMM **272**, 2146 (2004)

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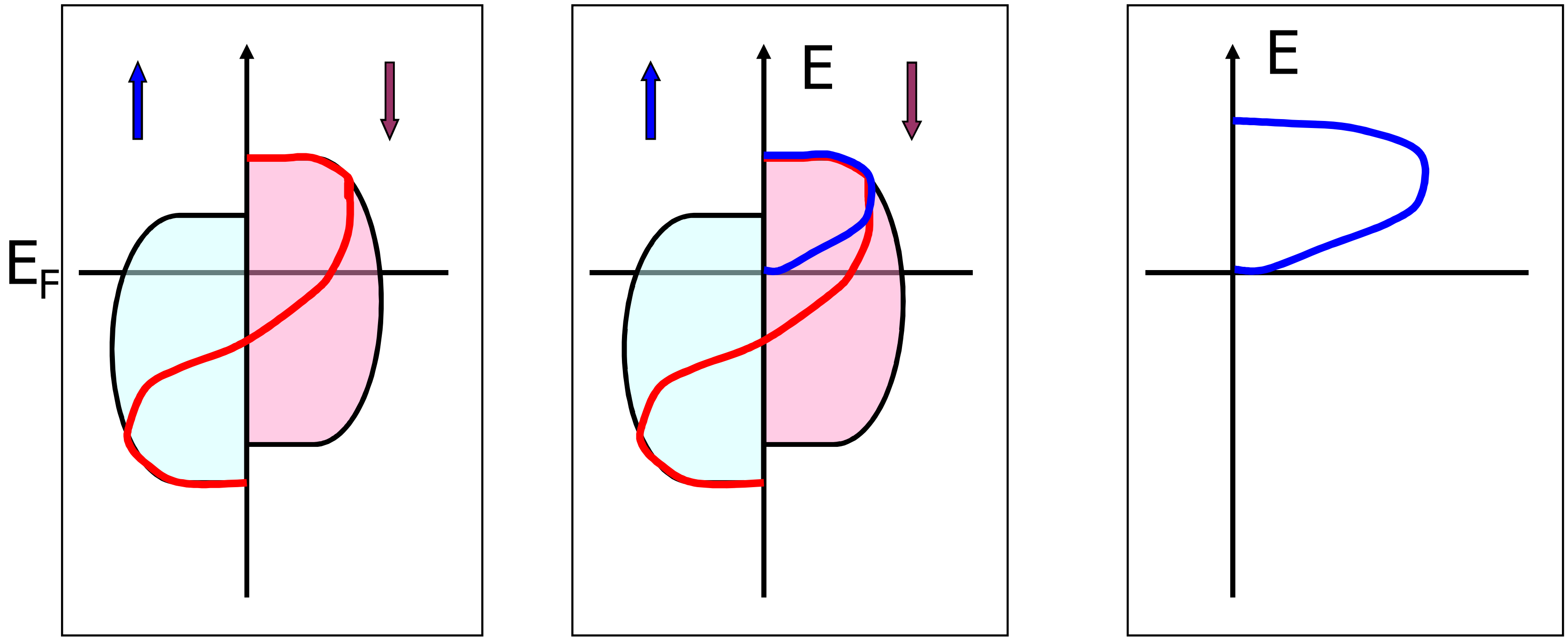
# Explanation of the XMCD shape




$(D_{\uparrow} - D_{\downarrow})(E)$

$f(E) \cdot (D_{\uparrow} - D_{\downarrow})(E)$

XMCD signal at one edge

Leading quantity for XMCD: exchange splitting of 3d DOS, determines XMCD shape

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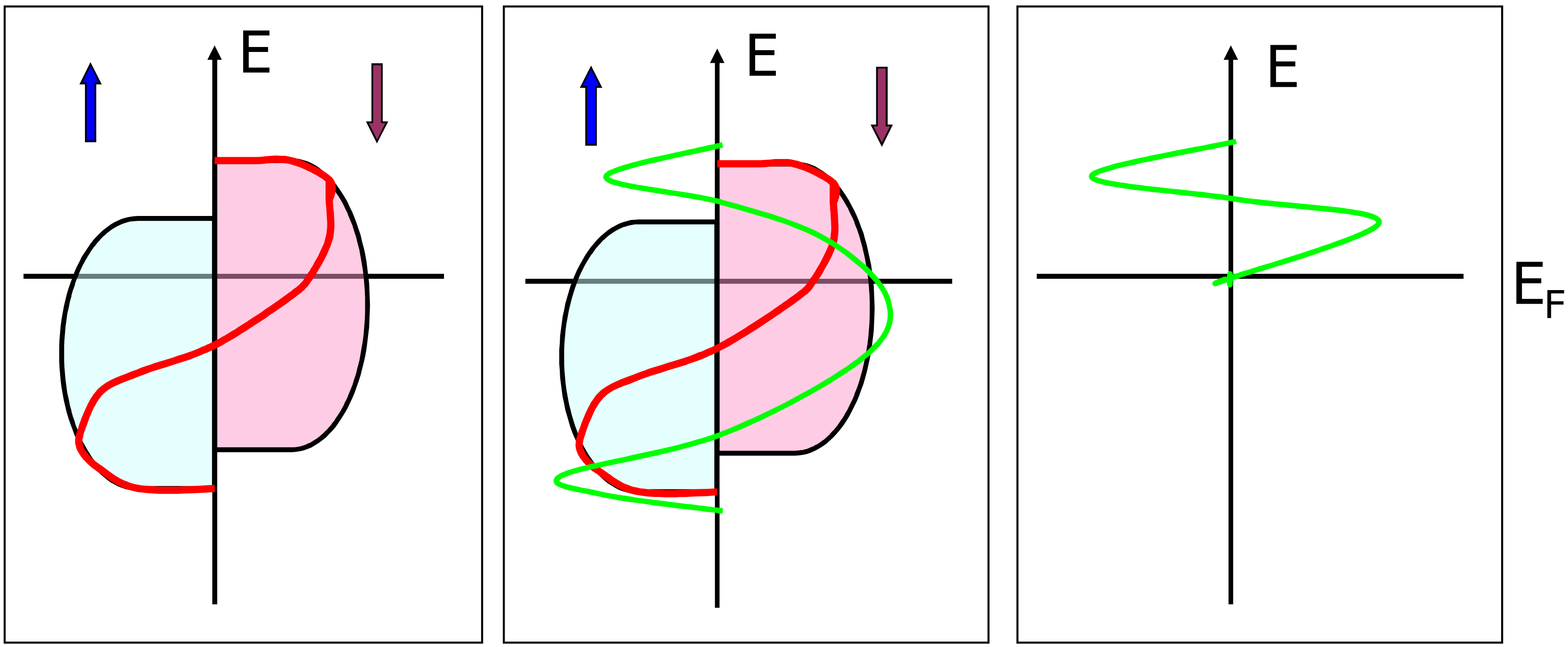
# Explanation of the XMLD shape




$$(D_\uparrow - D_\downarrow)(E) \qquad \Delta \frac{d}{dE}(D_\uparrow - D_\downarrow)(E) \qquad \Delta \cdot f(E) \cdot \frac{d}{dE}(D_\uparrow - D_\downarrow)(E)$$

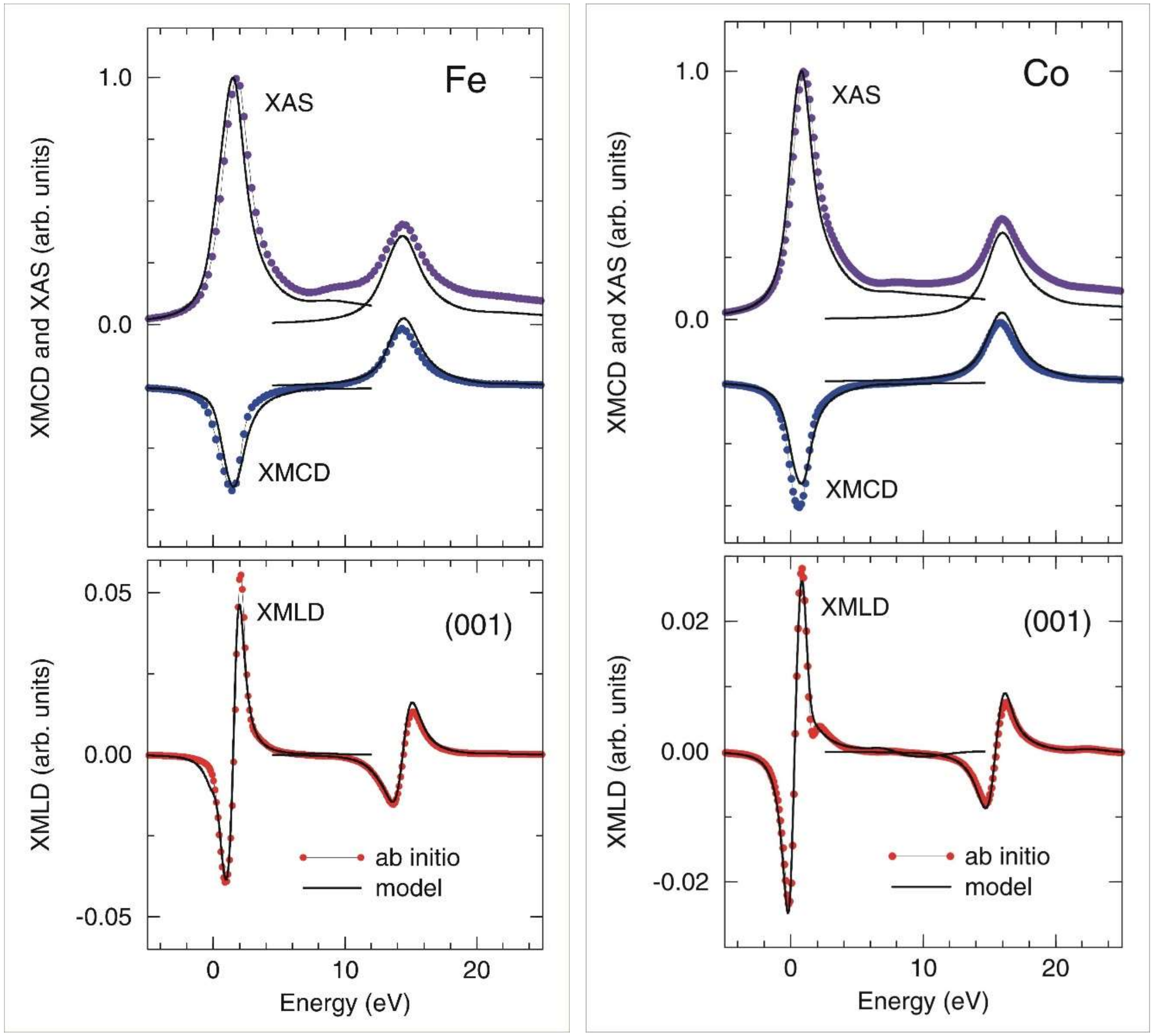
Leading quantity for XMLD: exchange splitting of 2p level

XMLD is quadratic in M

XMLD signal at one edge

# How good are the assumptions in the model ?

- ➔ It is excellent for XAS, XMCD and XMLD !
- ➔ Useful for studying the origin of XMCD and XMLD




Kunes & Oppeneer, PRB **67**, 024431 (2003)

# Magnetocrystalline anisotropy in XMLD

Effect of crystal field: Cubic symmetry

**Combination of different m-orbital spin-pol. DOS**

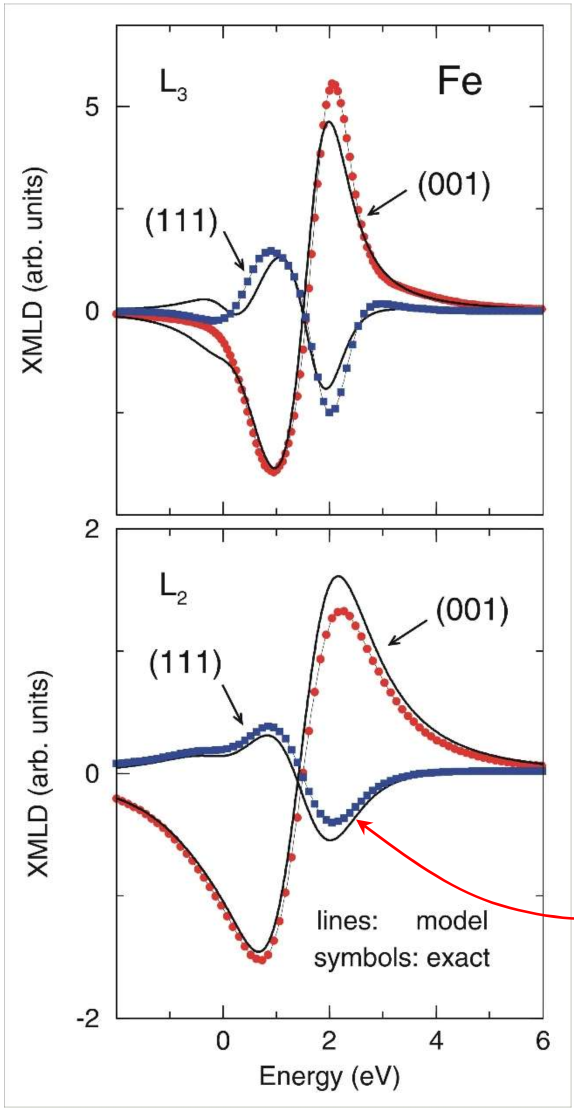
$$\text{XMLD} \approx \Delta \frac{d}{dE}\left[ \alpha(t_{2g\uparrow} - t_{2g\downarrow}) + \beta(e_{g\uparrow} - e_{g\downarrow})\right]$$

$$M(001): \begin{bmatrix} \alpha = -1 \\ \beta = 2 \end{bmatrix} \qquad M(111): \begin{bmatrix} \alpha = 1 \\ \beta = -1 \end{bmatrix}$$

$$\begin{cases} t_{2g} : \frac{xy}{r^2}, \frac{xz}{r^2}, \frac{yz}{r^2} \\ e_g : \frac{x^2 - y^2}{r^2}, \frac{3z^2 - 1}{r^2} \end{cases}$$

➔ different combination of m-partial DOS probed, depending on M axis

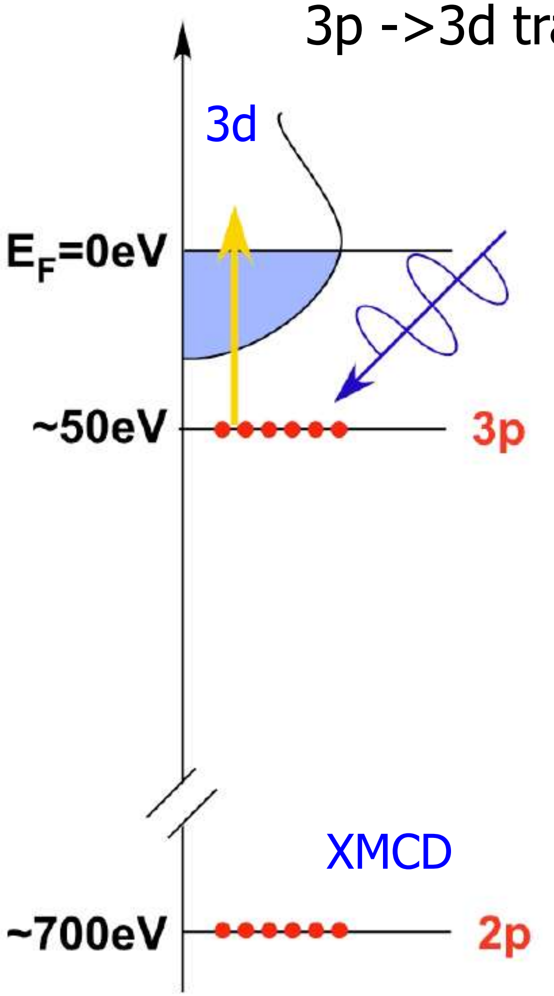
➔ large magnetocrystalline anisotropy appears in XMLD spectra



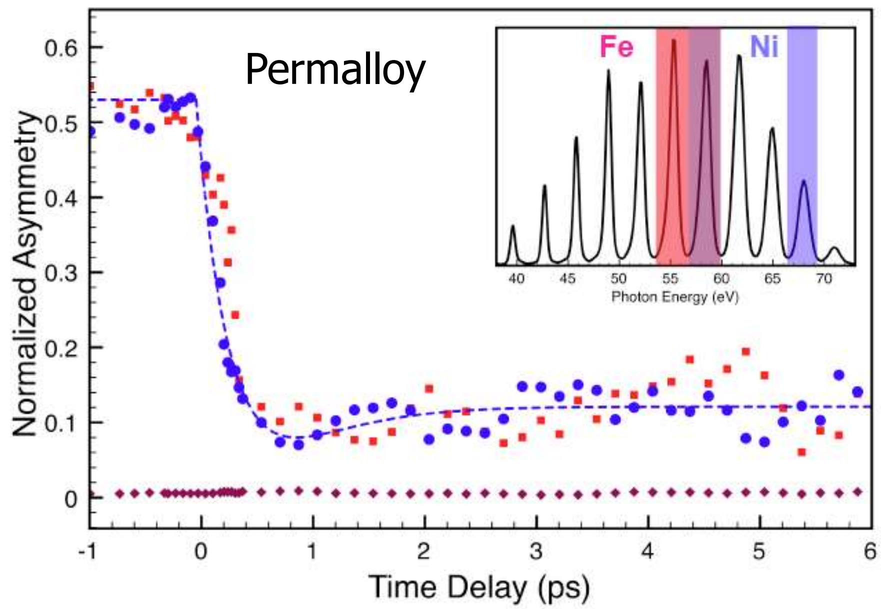

Effect of val.-SOI

Kunes & Oppeneer, PRB **67**, 024431 (2003)

# How about the 3p (M) semi-core edges?




## T-MOKE in XUV




La-O-Vorakiat et al, PRL **103**, 257402 (2009)

- Ultrafast element-selective demagnetization of Fe and Ni in permalloy

# Transversal MOKE at M edges

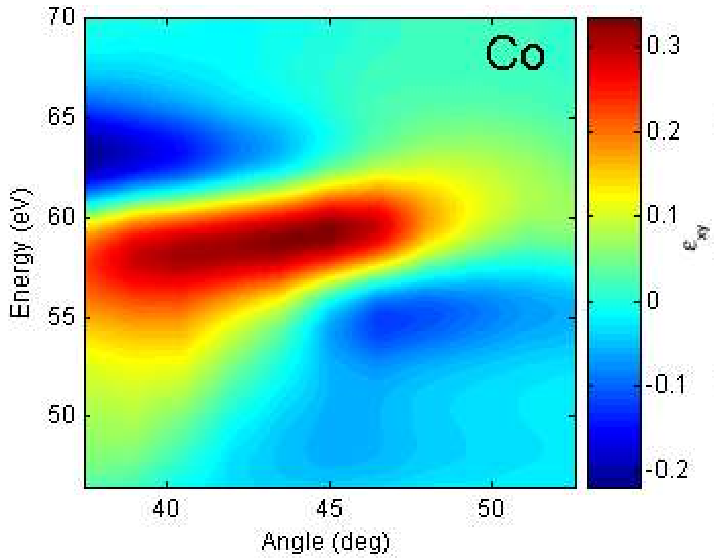
Measured as intensity change of lin. polarized light in reflection (cf. Fresnel theory)

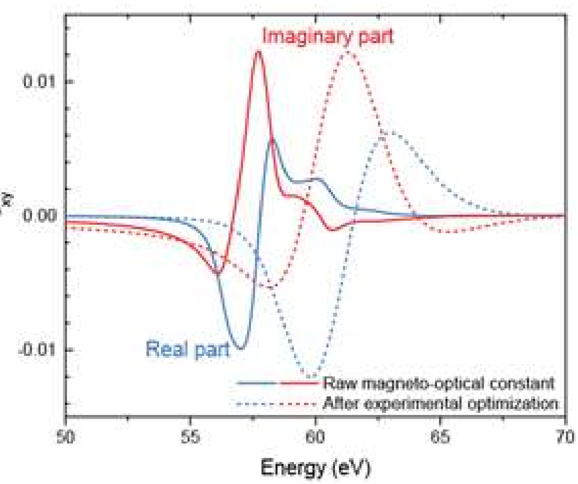
$$A = 2\,Re\left[\frac{\sin 2\theta_i\,\epsilon_{xy}}{n^4\cos\theta_i^2 - n^2 + \sin\theta_i^2}\right] = 2\,\mathrm{Re}[\mathrm{F}(\theta,n)\,\epsilon_{xy}] = 2\,Re[\mathrm{F}(\theta,n)]Re[\epsilon_{xy}] - 2Im[\mathrm{F}(\theta,n)]Im[\epsilon_{xy}]$$

Demagnization mechanism: Compute *ab initio* $\varepsilon_{xy}$ for several cases: 1) frozen magnon excitations, 2) reduced *exchange splitting* (spin-flips), 3) increased electron temperature $T_e$ - construct the <u>change</u> 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)

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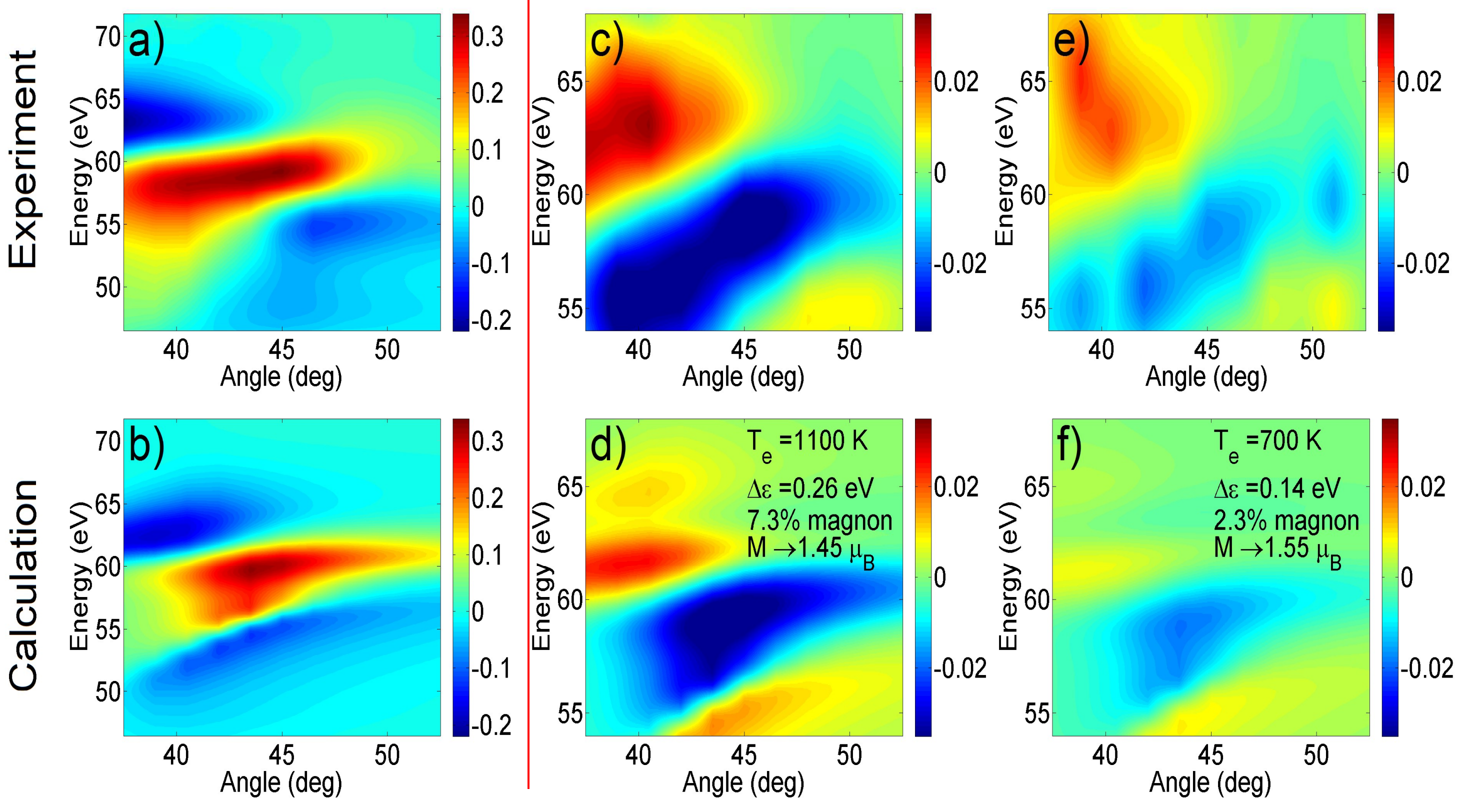
# Comparison of experiment and *ab initio* theory

T-MOKE <0ps 700 fs 3 ps ΔT-MOKE

Exp.

Theory




For Co

- ➢Surprisingly small contribution from spin-flips (exch. split reduction)
- ➢ Larger effect (2/3) is due to fast magnon excitation => reduction of $M_z$

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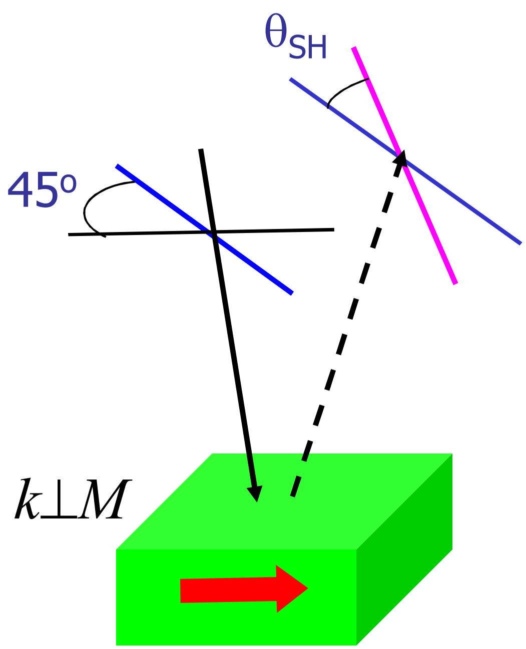
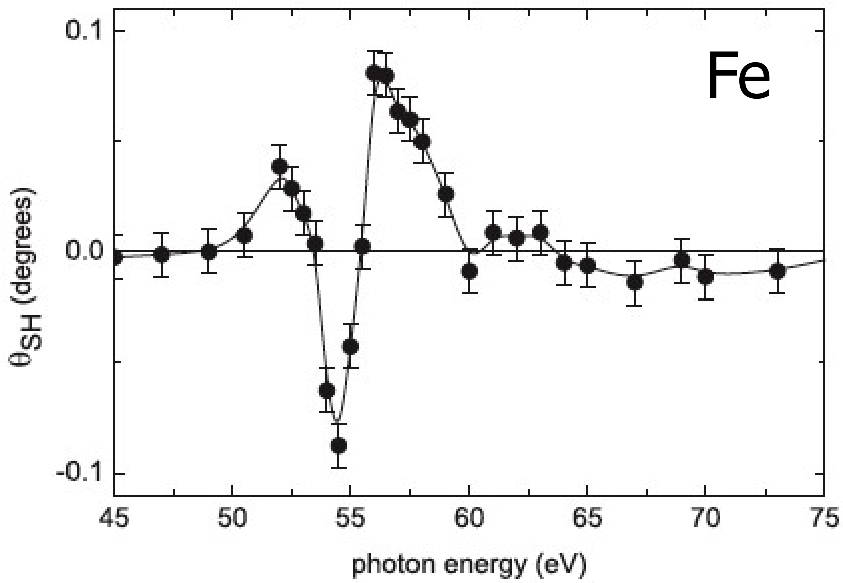
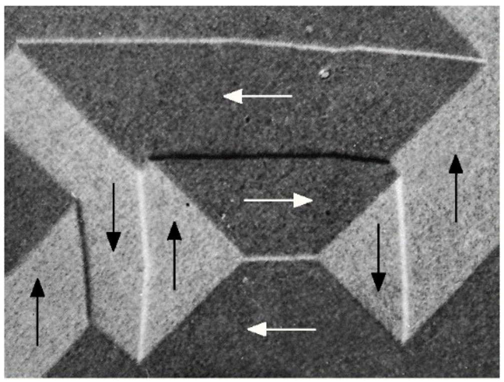
# Quadratic in *M* effect in-near-normal reflection




Schäfer-Hubert effect
(or Voigt effect in reflection)

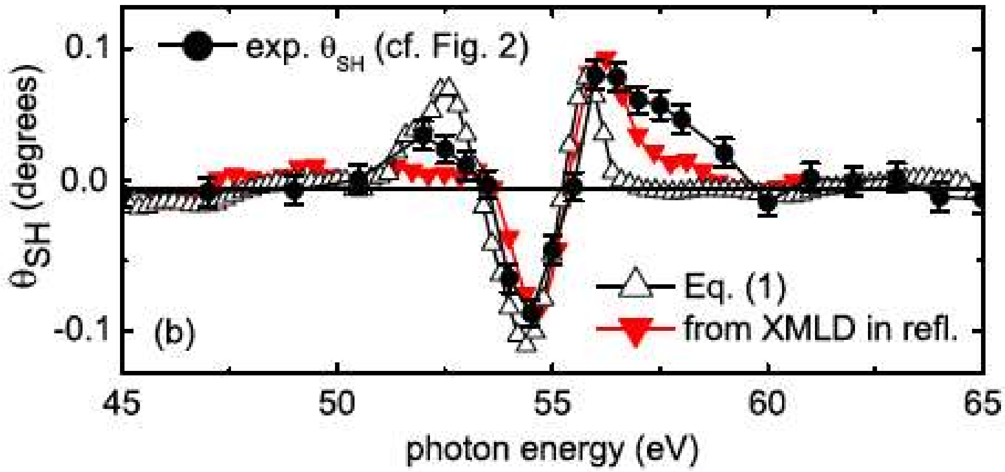


Near-normal incidence detection at Fe 3p edges

$$\theta_{SH} \approx \mathrm{Re}\left[\frac{(n_{\parallel} - n_{\perp})n_0}{n_{\parallel}n_{\perp} - n_0^2}\right] \approx \mathrm{Re}\left[\frac{(\epsilon_{\parallel} - \epsilon_{\perp})n_0}{(n^2 - n_0^2)n}\right]$$

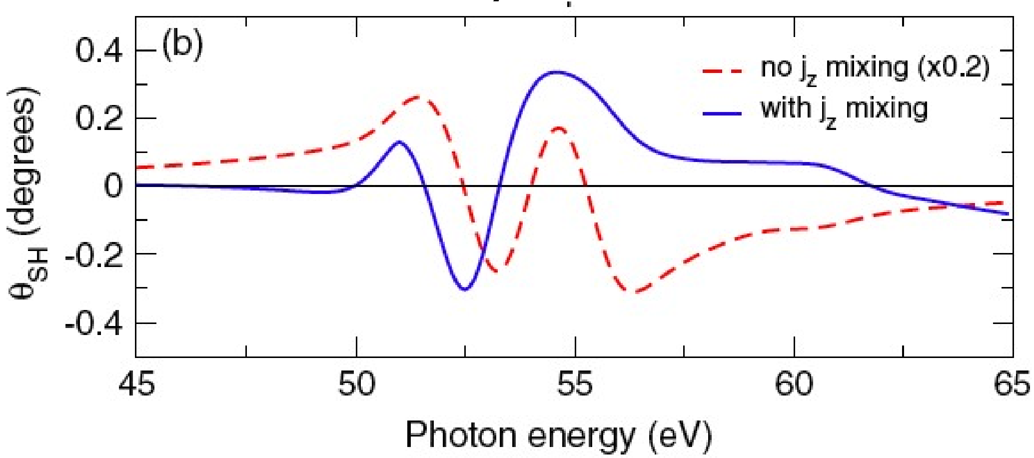
# Comparison to *ab initio* calculations




Importance of treating exchange splitting and spin-orbit interaction on equal footing

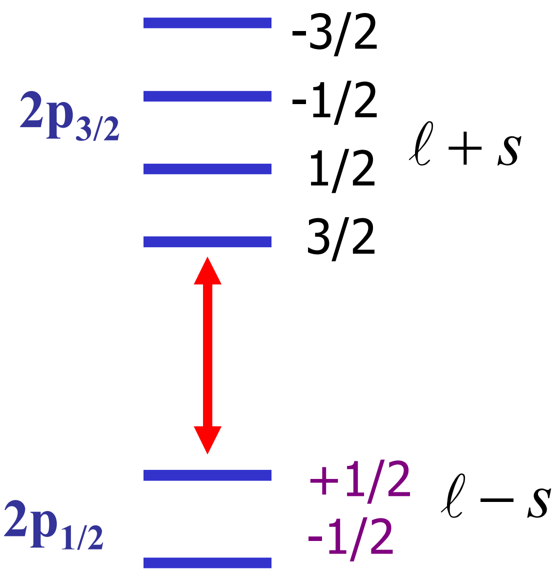
Importance of *hybridization* of $j_z$ states

*Ab initio* theory




Valencia et al, PRL **104**, 187401 (2010)

*2p exchange splitting*



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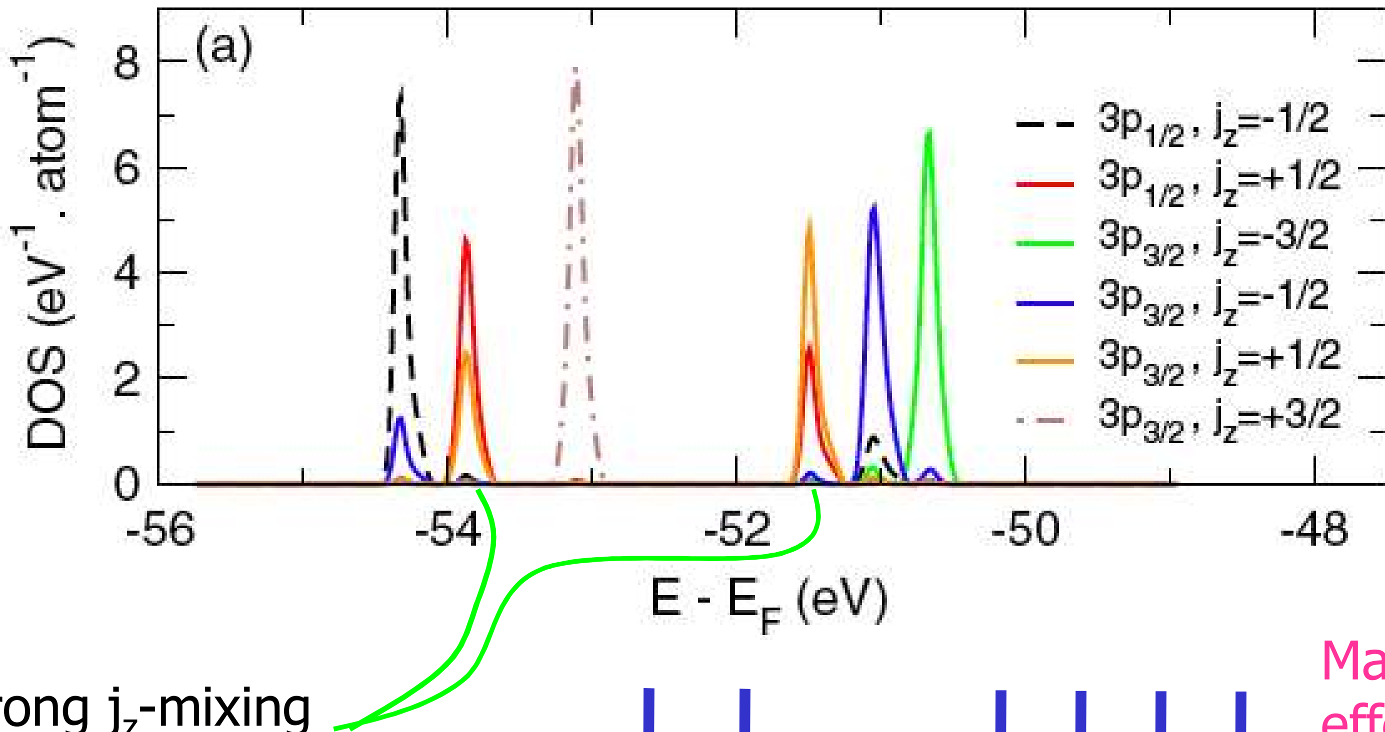
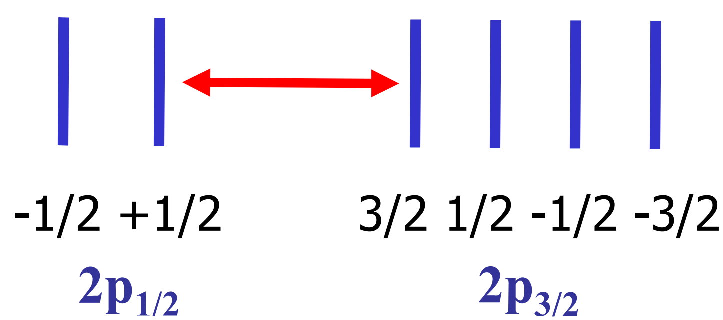
# $j_z$-hybridization 3p semi-core level of Fe

Strong mixing of j, $j_z$ states, SO splitting & exchange splitting equally large




Strong $j_z$-mixing




Magneto-X-ray effects as large as at 2p's!

➢ No expansion in small quantity possible !

# Summarizing light – magnetic matter interaction

Magnetic spectroscopy is a highly sensitive tool that can detect minute magnetizations (spin Hall effect)

Exchange and spin-orbit splitting work together in different ways in valence and X-ray regime to bring about light - magnetic matter interaction

*Ab initio* quantum theory (effective single particle theory) works well but it is needed to know about its limitations

Current frontlines:

1) Ultrasensitive measurements to observe very small spin-orbit related effects (e.g. Inverse spin galvanic effect)
2) Ultrafast limit of modifications & control of magnetization, experiments and suitable theory
3) Nonlinear magneto-optic effects

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# Literature


S. W. Lovesey and S. P. Collins, *X-Ray Scattering and Absorption by Magnetic Materials* (Clarendon Press, Oxford, 1996) .

A.K. Zvezdin and V.A. Kotov, *Modern Magnetooptics and Magnetooptical Materials* (London, Taylor & Francis, 1997).

P.M. Oppeneer, *Magneto-optical Kerr Spectra*, in Handbook of Magnetic Materials, Vol. 13, edited by K.H.J. Buschow (Elsevier, Amsterdam, 2001), pp. 229-422.

W. Kleemann, *Magneto-optical materials*, in Handbook of Magnetism and Advanced Magnetic Materials, Vol. 4, edited by H. Kronmüller and S.S.P. Parkin (Wiley, New York, 2006).

J. Stöhr and H.-C. Siegmann, *Magnetism: From Fundamentals to Nanoscale Dynamics* (Springer, Berlin, 2007).

# Appendix I: Alternative way to describe X-ray spectra

Other way of describing effects – scattering formalism:

$$f \approx (\vec{e}\,'\cdot\vec{e})F_0 - i((\vec{e}\,'\times\vec{e})\cdot\vec{M})F_1 + (\vec{e}\,'\cdot\vec{M})(\vec{e}\cdot\vec{M})F_2$$

Charge scattering (XAS)

1st order magnetic scattering (XMCD)

2nd order magnetic scattering (XMLD)

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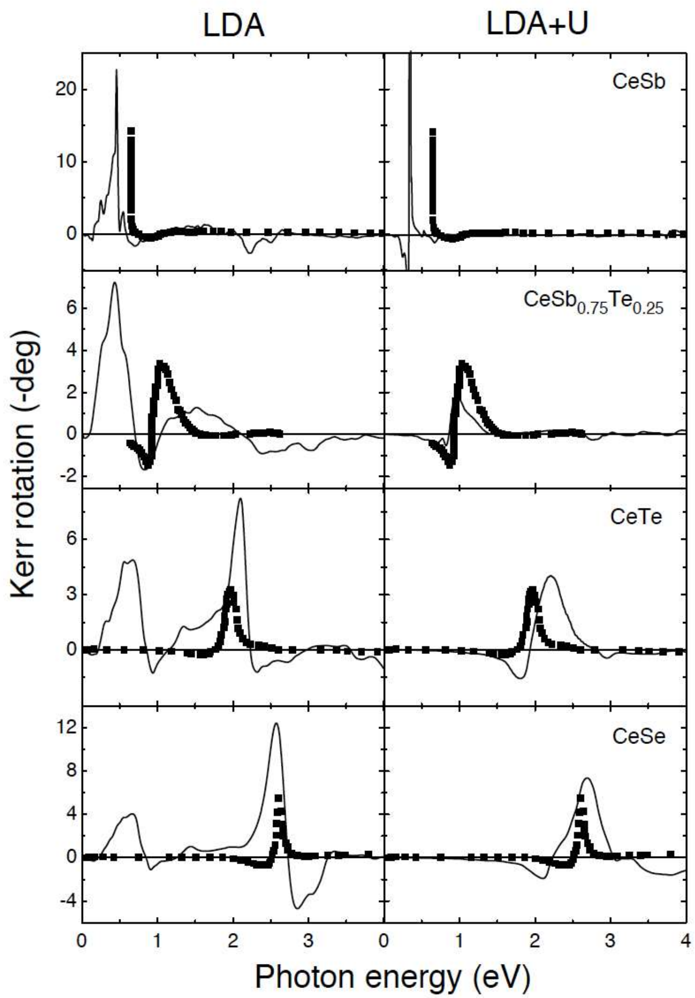
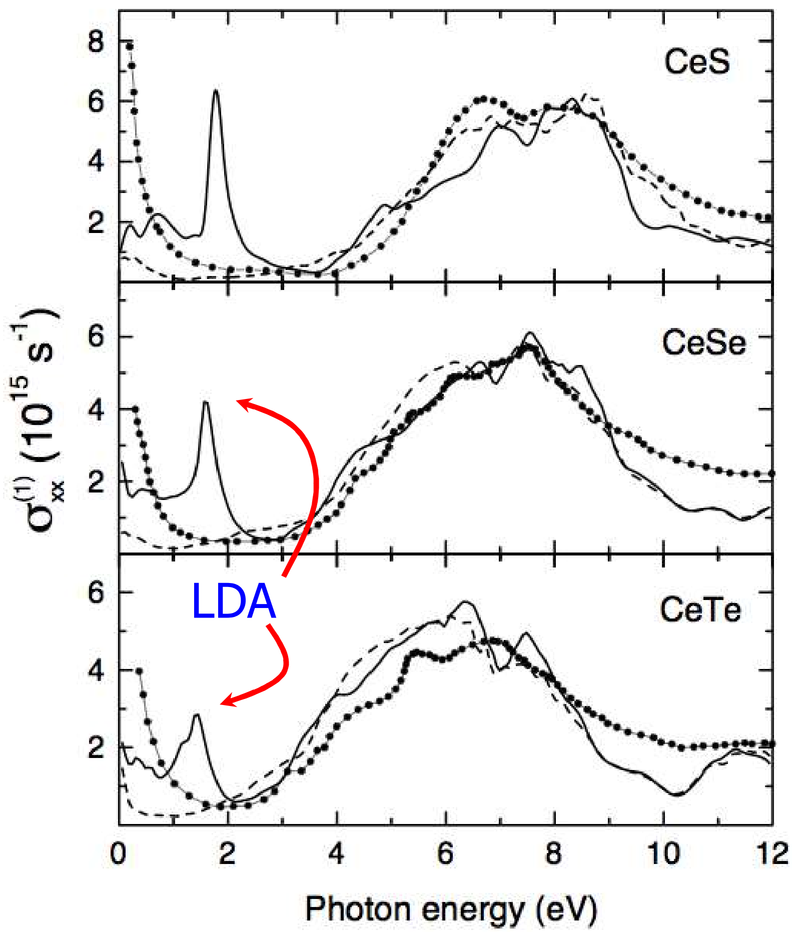
# Deficiency of DFT-LDA for localized 4f states



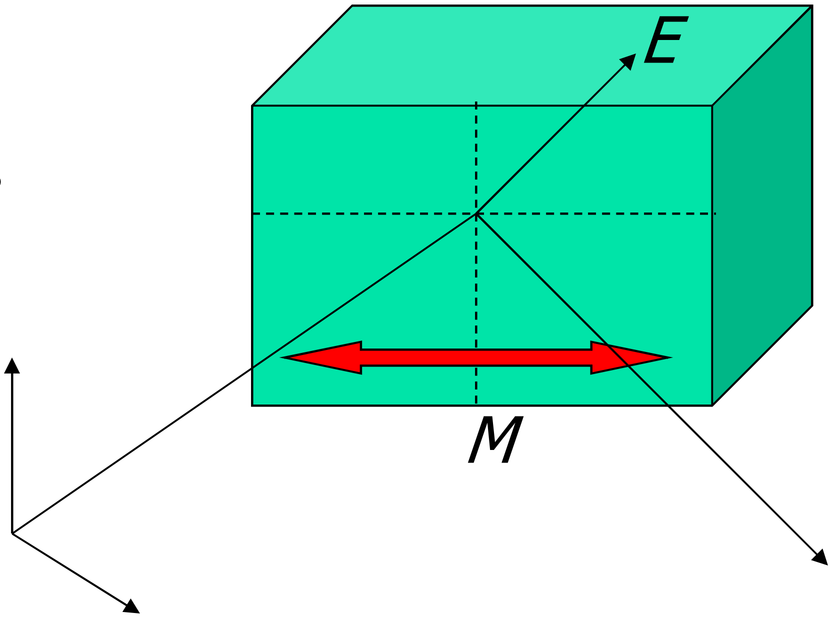




Nearly localized f state

=> LDA+U better

# Practicals' problem:

1) Material with magnetization in the scattering plane
2) Lin. pol. light *E*-vector at 45° to the magnetization
3) Consider *R(+M)-R(-M)*




Use the reflection coefficients to show that *R(+M)-R(-M)* is a measure of the magnetization and derive an expression for the magn. asymmetry:

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