Magneto-optical Kerr effect (MOKE) and Xray Circular Dichroism (XMCD) - microscopic mechanism and experimental measurement

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Magneto-optical Kerr effect:

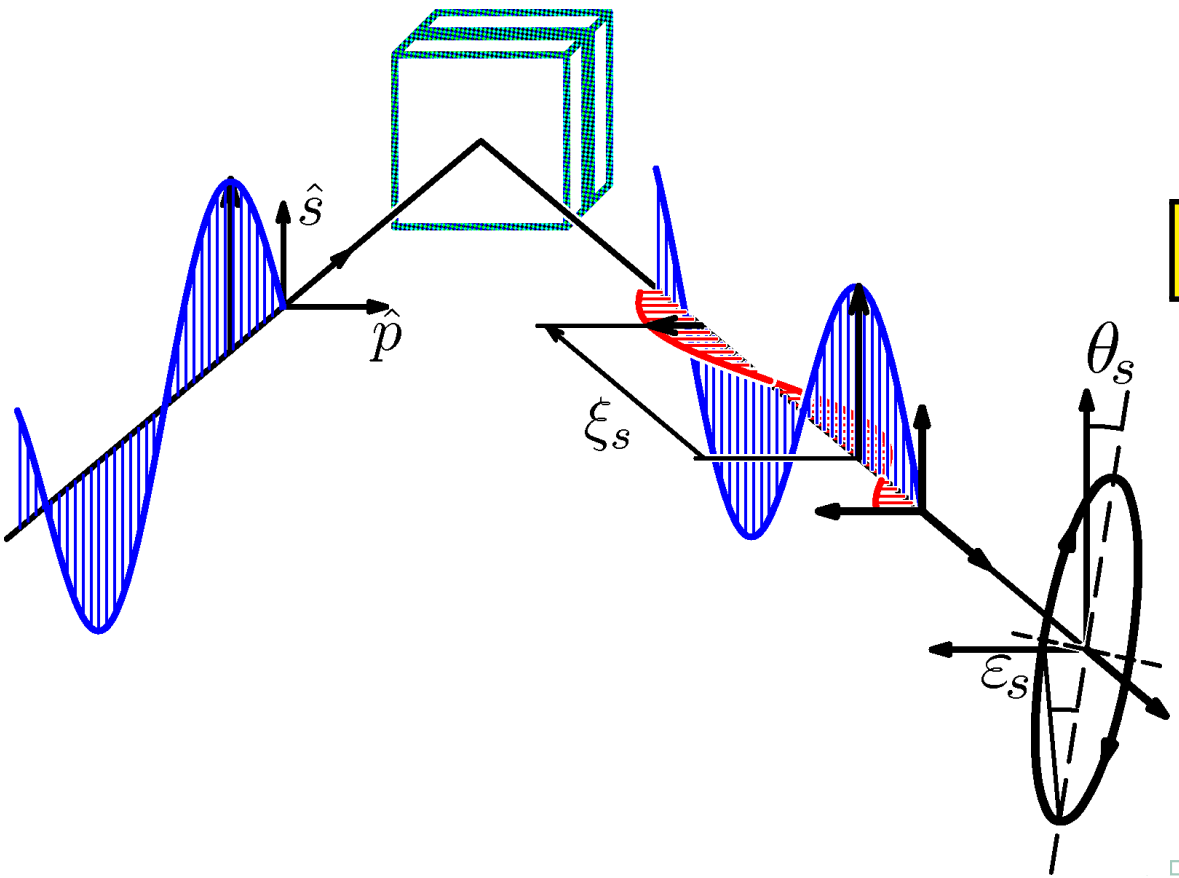
- Change of optical properties (polarization state, reflectivity) by presence/change of magnetization of the sample.

One can separate usage of magneto-optical (MO) effects to:

- MO as a metrology tool to study magnetism:
  - MO magnetometry (study of magnetization reversal).
  - MO microscopy (study of domain wall and its propagation).
  - Magnetization dynamic studies (precession etc.)
  - MO as a tool for ultrafast magnetization processes.
- MO spectroscopy to study optical properties of the MO effect:
  - Magnetism is understand as a perturbation, reducing symmetry of the solids and hence introducing new optical features.
  - Study of spin-orbit interaction.
  - Interaction between light and magnetism – a very fundamental interaction.

MO effect I: Magneto-optical Kerr effect (MOKE):

- For example: incident s-polarized wave.
- Magnetized sample
  $\Rightarrow$ hence: also p-polarized wave appears on the reflection.



MOKE linear in $\vec{M}$

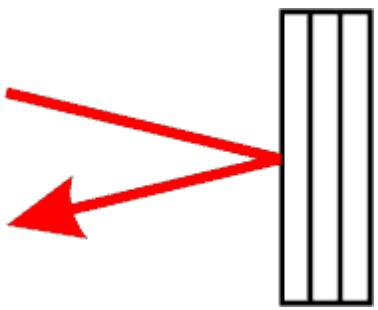
- θ_s: Kerr rotation.
- ϵ_s: Kerr ellipticity.

MO effect I: Kerr and Faraday MO effect:

Due to historical reasons, there are different names for MO effects measured in reflection and transmission.

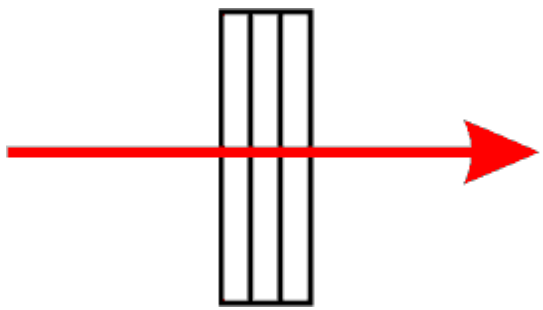
Kerr effect:

- measured in reflection.
- discovered 1876.



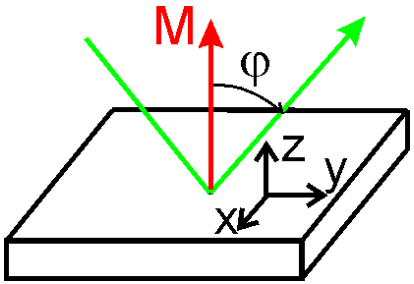
Faraday effect:

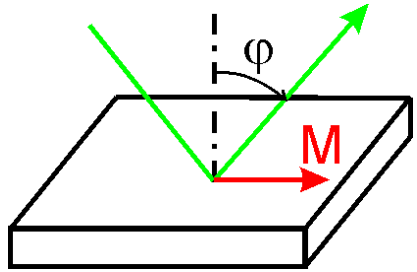
- measured in transmission.
- discovered 1845.

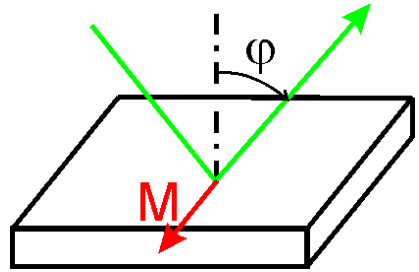


MOKE configurations and permittivity tensor:

Polar MOKE	Longitudinal MOKE	Transversal MOKE
$M \perp$ sample surface	$M \parallel$ plane of incidence	$M \perp$ plane of incidence





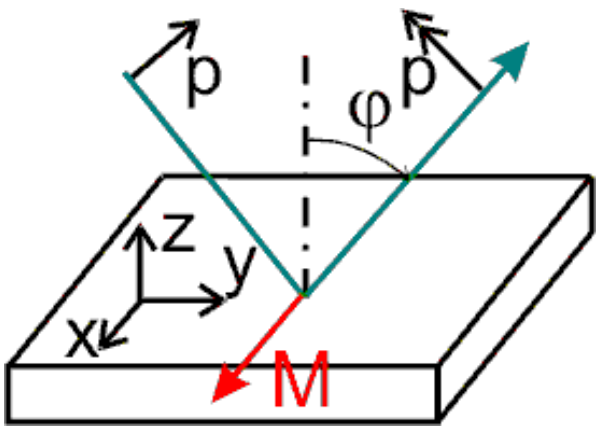


Polarization induced by magnetization: $\Delta\vec{P}_M = \varepsilon_1(\vec{M} \times \vec{E})$

$\begin{bmatrix} \varepsilon_0 & -\varepsilon_1 m_z & 0 \\ \varepsilon_1 m_z & \varepsilon_0 & 0 \\ 0 & 0 & \varepsilon_0 \end{bmatrix}$	$\begin{bmatrix} \varepsilon_0 & 0 & \varepsilon_1 m_y \\ 0 & \varepsilon_0 & 0 \\ -\varepsilon_1 m_y & 0 & \varepsilon_0 \end{bmatrix}$	$\begin{bmatrix} \varepsilon_0 & 0 & 0 \\ 0 & \varepsilon_0 & -\varepsilon_1 m_x \\ 0 & \varepsilon_1 m_x & \varepsilon_0 \end{bmatrix}$
$\Phi_{s/p}(m_z)$	$\Phi_{s/p}(m_y)$	$\Delta r_{pp}(m_x)$

MO effect II: transversal MOKE:

- Incident p-polarized wave.
- Magnetization in-plane and perpendicular to the incident plane (so-called transversal magnetization direction).
- Change of the reflected p-polarized intensity due to magnetization in the sample (in this particular case, no change in polarization of the reflected light appears).



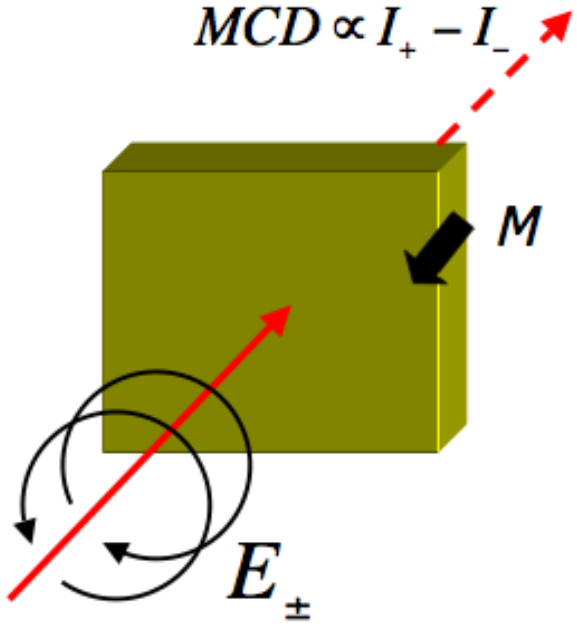
MO effect III: Magnetic dichroism and birefringence:

Dichroism: different damping of both light's eigen-modes.

Birefringence: different propagation speed of both light's eigen-modes.

Magnetic circular dichroism (MCD):

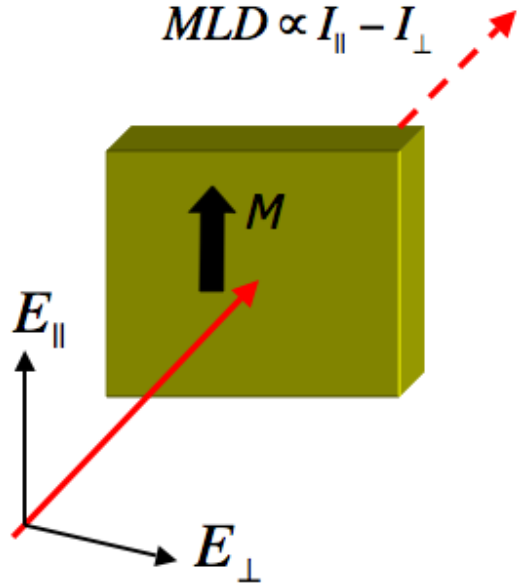
- Different absorption for circularly left and right polarized light.
- MCD linear in $\vec{M}$.
- MOKE and MCD has the same microscopic origin, they just manifest in different ways.



$$MCD \propto \mathrm{Im}[\varepsilon_+ - \varepsilon_-] \propto \mathrm{Im}[\sigma_{xy}(\omega)]$$

MO effect IV: Voigt effect:

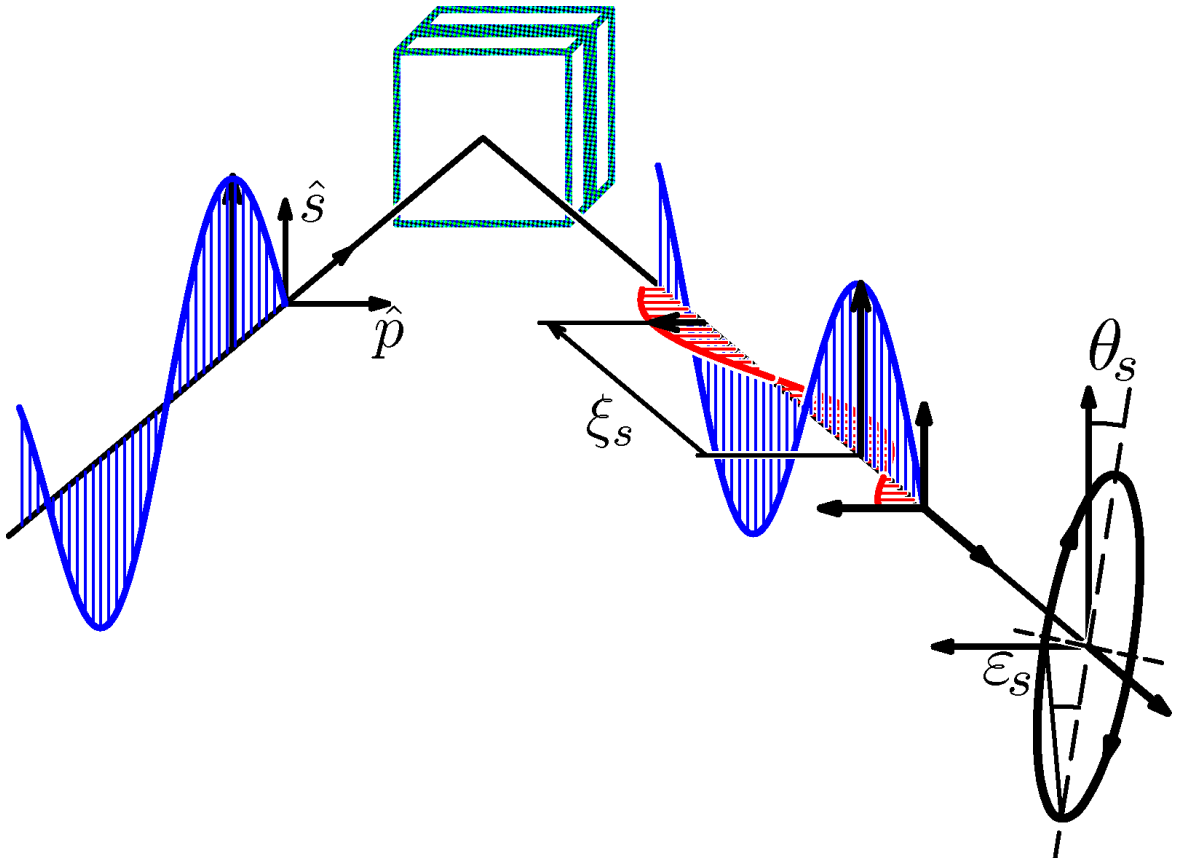
- Discovered 1899.
- Different absorption or phase shift for linear polarization parallel and perpendicular with the magnetization.
- Quadratic in $\vec{M}$ ($\sim M^2$).
- Also called Cotton-Mouton effect or linear magnetic dichroism/birefringence (LMD/LMB)
- The same microscopic origin as quadratic MOKE (QMOKE) (more precisely, Voigt effect is simplest case of QMOKE).



$$MLD \propto \mathrm{Im}[\varepsilon_{\parallel} - \varepsilon_{\perp}] \propto \mathrm{Re}[\sigma_{\parallel} - \sigma_{\perp}]$$

Classification of the MO effects:

- Even / odd effect in magnetization.
- Measured in transmission / reflection.
- Detected change of intensity / polarization.
- Probing light is linearly / circularly polarized.



Family of magneto-optical effects:

Linear pol.	Detected: Polariz.	Detected: Intensity
Linear in M	MOKE, (Kerr and Faraday effect) [Hall effect]	Transversal-MOKE
Quadratic in M	QMOKE, Voigt effect, Linear Magnetic Birefringence (LMB)	Linear Magnetic Dichroism (LMD) [AMR]
Circular pol.	Detected: Polariz.	Detected: Intensity
Linear in M	Mag. Circular Birefringence (MCB)	Magnetic Circular Dichroism (MCD)
Quadratic in M	?	quadratic-MCD (?)

[...] denotes nomenclature in research of conductivity.

Origin of MO effect (microscopic):

Electronic structure of the FM material
[microscopic description]

$\Downarrow$

Permittivity tensor of each layer
[phenomenological description]

$$\varepsilon = \begin{bmatrix} \varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} \\ \varepsilon_{yx} & \varepsilon_{yy} & \varepsilon_{yz} \\ \varepsilon_{zx} & \varepsilon_{zy} & \varepsilon_{zz} \end{bmatrix}$$

$\Downarrow$

Reflectivity matrix of whole sample
[maximal accessible optical information]

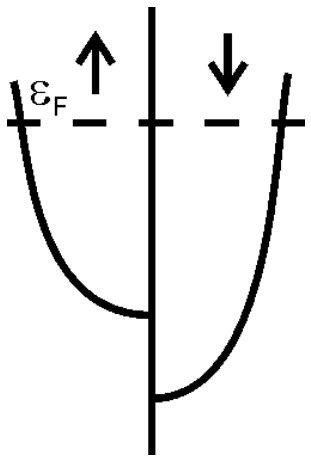
$$R = \begin{bmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{bmatrix}$$

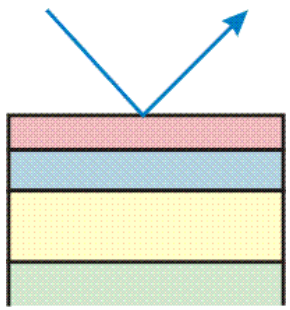
$\Downarrow$

Measured Kerr effect: $\Phi_s = \frac{r_{ps}}{r_{ss}}$

$\Downarrow$

Signal measured by MO setup





MO effects and permittivity tensors

[Note: tensors on this slide are only illustrative.]

$\Rightarrow$ Linear MOKE: PMOKE, LMOKE, TMOKE, MCD, MCB, [Hall]

$$\begin{bmatrix} \varepsilon_0 & -\varepsilon_1 m_z & \varepsilon_1 m_y \\ \varepsilon_1 m_z & \varepsilon_0 & -\varepsilon_1 m_x \\ -\varepsilon_1 m_y & \varepsilon_1 m_x & \varepsilon_0 \end{bmatrix} \quad \text{MO signal} \sim \varepsilon_1(m_i)$$

$\Rightarrow$ Quadratic MOKE:

$$\begin{bmatrix} \varepsilon_0 & \varepsilon_1(m_i m_j) & 0 \\ \varepsilon_1(m_i m_j) & \varepsilon_0 & 0 \\ 0 & 0 & \varepsilon_0 \end{bmatrix} \quad \text{MO signal} \sim \varepsilon_1(m_i m_j)$$

$\Rightarrow$ Voigt effect: MLD, MLD, [AMR]

$$\begin{bmatrix} \varepsilon_{xx}(m_i m_j) & 0 & 0 \\ 0 & \varepsilon_{yy}(m_i m_j) & 0 \\ 0 & 0 & \varepsilon_{zz}(m_i m_j) \end{bmatrix} \quad \text{MO signal} \sim \sqrt{\varepsilon_{zz}(m_i m_j) - \varepsilon_{yy}(m_i m_j)}$$

Photon absorption: electric-dipole approximation:

- The largest contribution to the absorption is given by so-called electric-dipole approximation (valid for $\lambda \gg a$), providing so-called electric-dipole transitions.
- Hence, whole vast energy range can be described by so-called Kubo formula, determining conductivity (absorption) for a given photon energy (shown later).

Selection rules of electric-dipole transitions:

Electric dipole transition is allowed when following conditions are fulfilled:

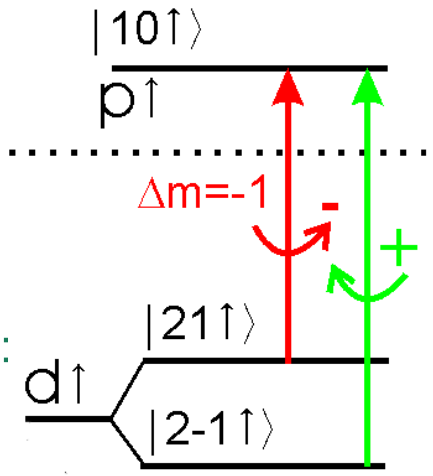
Energy: $E_f - E_i = \hbar\omega$ (absorbed photon energy is difference between energies of the final and initial electron states)

Momentum: $\hbar\omega/c \approx 0$ (photon has negligible momentum compared to one of the electron. I.e. the momentum of the electron is kept between initial and final state (vertical transitions)).

Electron spin : $\Delta s = 0$ (as photon has no spin, spin of electron is preserved for electric dipole transitions)

Orbital momentum: $\Delta l = \pm 1$ (photon has angular momentum $1\hbar$). Therefore only $s \leftrightarrow p$, $p \leftrightarrow d$ etc. transitions are allowed.

Orbital momentum along z-axis (magnetic number): $\Delta m = \pm 1$ (determines if photon is circularly right or left polarized).



Ab-initio calculation of permittivity tensor

Kubo formula: conductivity determination.

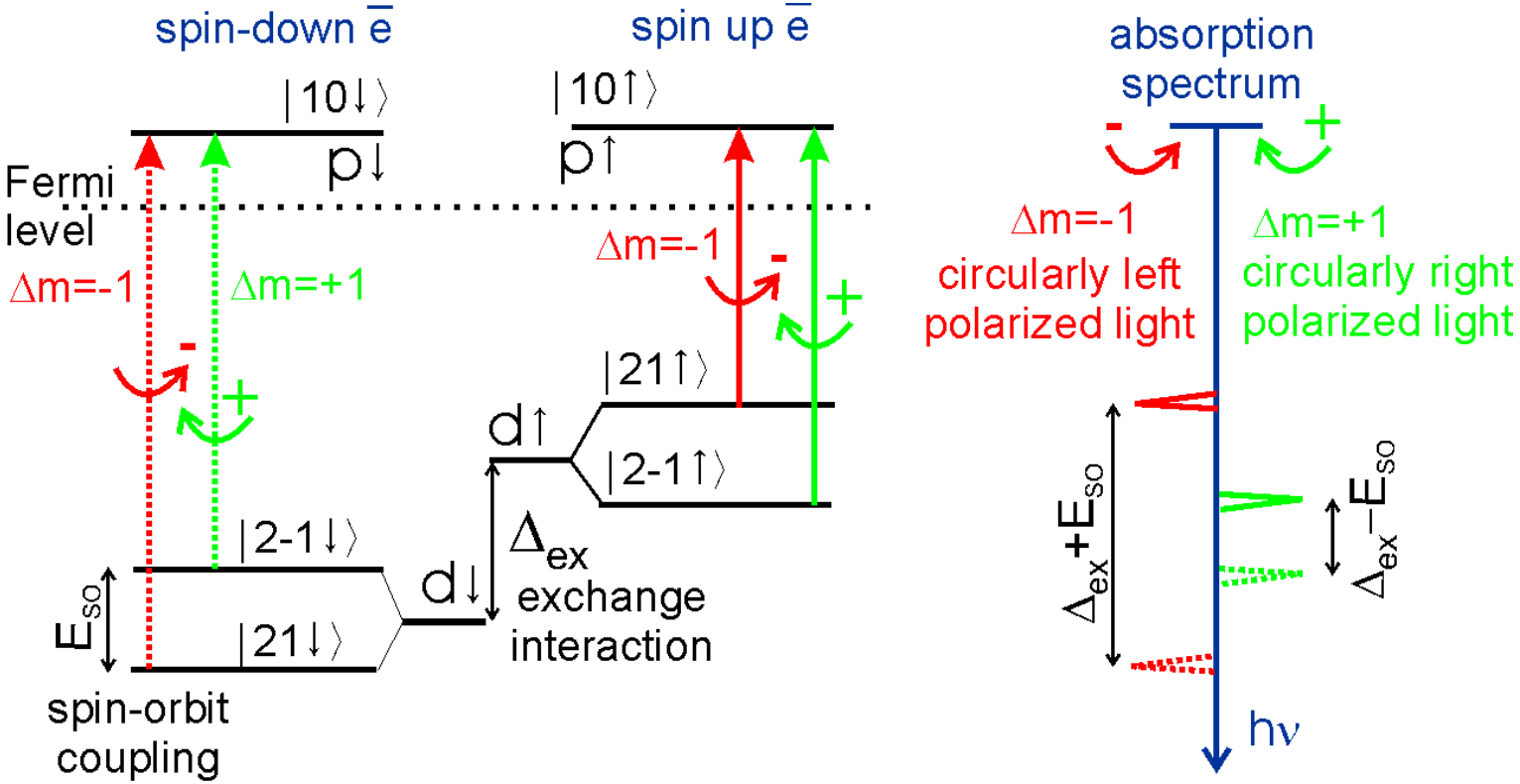
$$\Im[\varepsilon_{xx}] \sim \sum_{i,f} (f(E_i) - f(E_f)) \times [|\langle i|p_+|f\rangle|^2 + |\langle i|p_-|f\rangle|^2] \times \delta(E_f - E_i - \hbar\omega)$$

$$\Re[\varepsilon_{xy}] \sim \sum_{i,f} (f(E_i) - f(E_f)) \times [|\langle i|p_+|f\rangle|^2 - |\langle i|p_-|f\rangle|^2] \times \delta(E_f - E_i - \hbar\omega)$$

where

- $\langle i|$, $|f\rangle$: initial and final states, respectively.
- $p_\pm = p_x \pm i p_y$, $p_x = i\hbar\partial/\partial x$, momentum operator
- terms in the Kubo formula means:
  - summation over all initial and final states, $\langle i|$ and $|f\rangle$.
  - $f(E_f)$, $f(E_i)$: electron occupancy of initial and final states.
  - $|\langle i|p_\pm|f\rangle|^2$: probability of the photon to be absorbed between $\langle i|$ and $|f\rangle$ states for circularly left/right polarized light (non-zero only when electric-dipole selection rules are fulfilled).
  - $\delta(E_f - E_i - \hbar\omega)$ assures energy conservation.

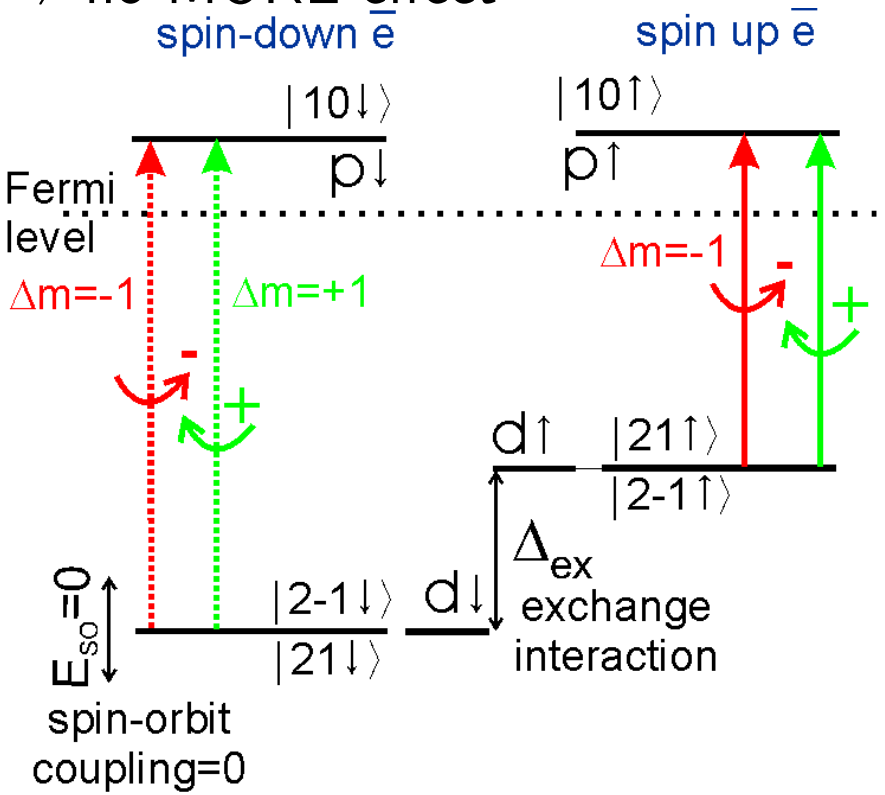
Magneto-optical spectroscopy microscopic picture

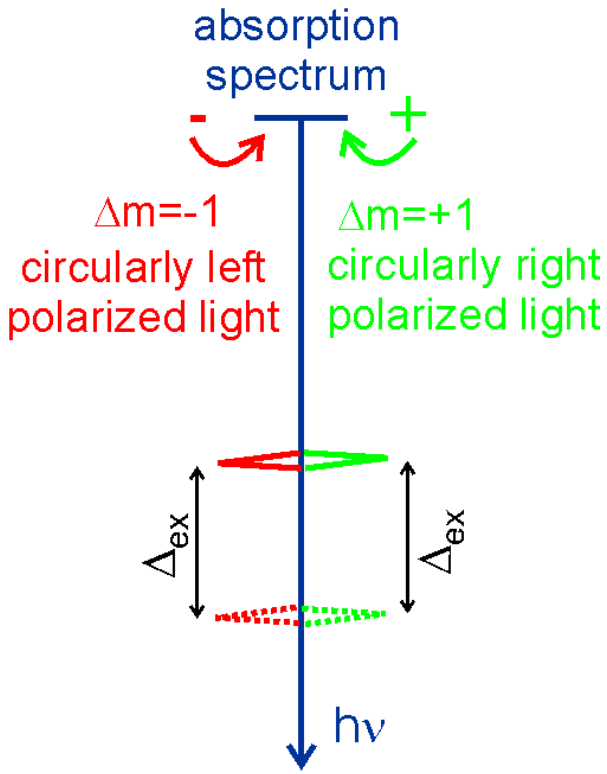


Simplified electronic structure for one point of the k-space.

No spin-orbit coupling assumed:

$\Rightarrow$ no MOKE effect

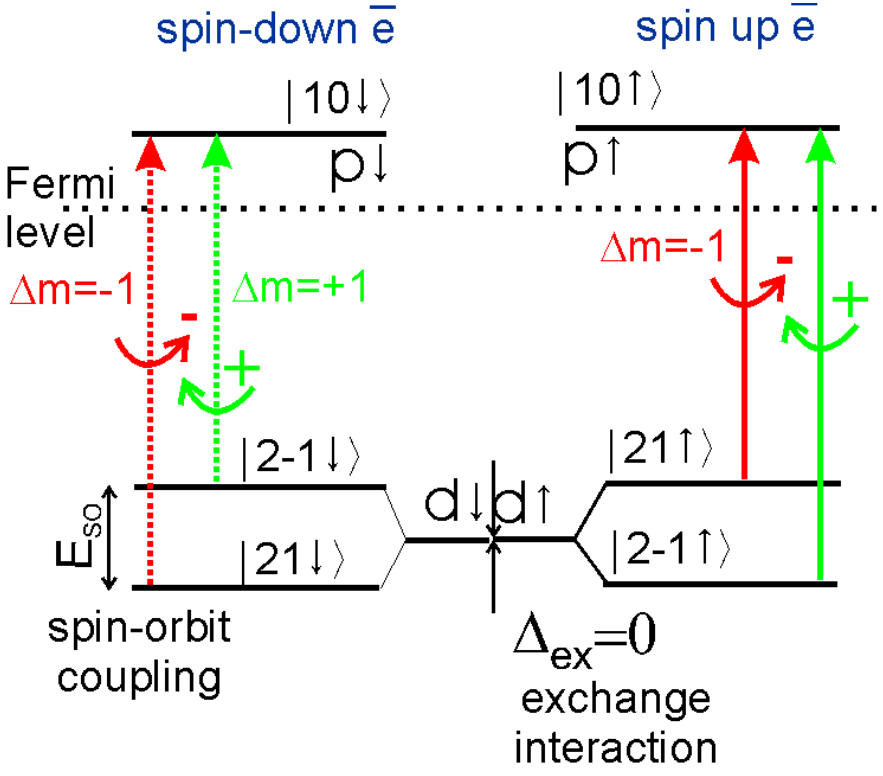


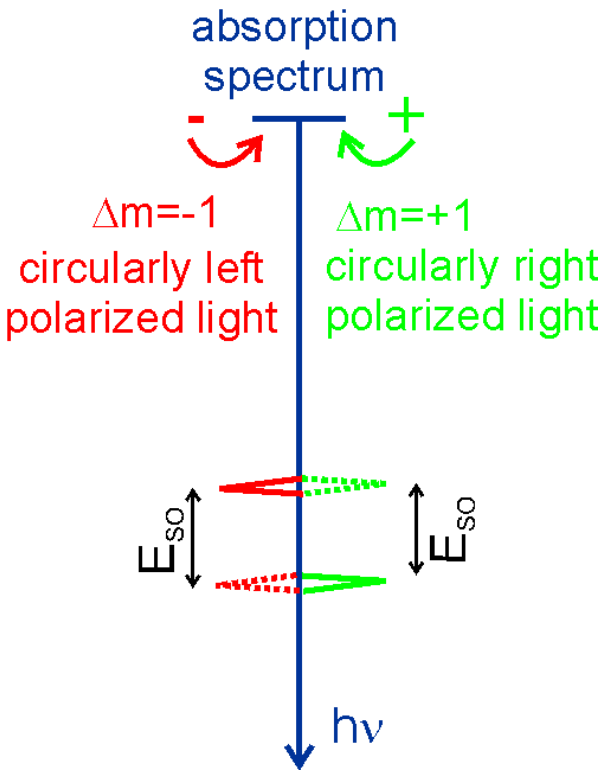


No exchange assumed:

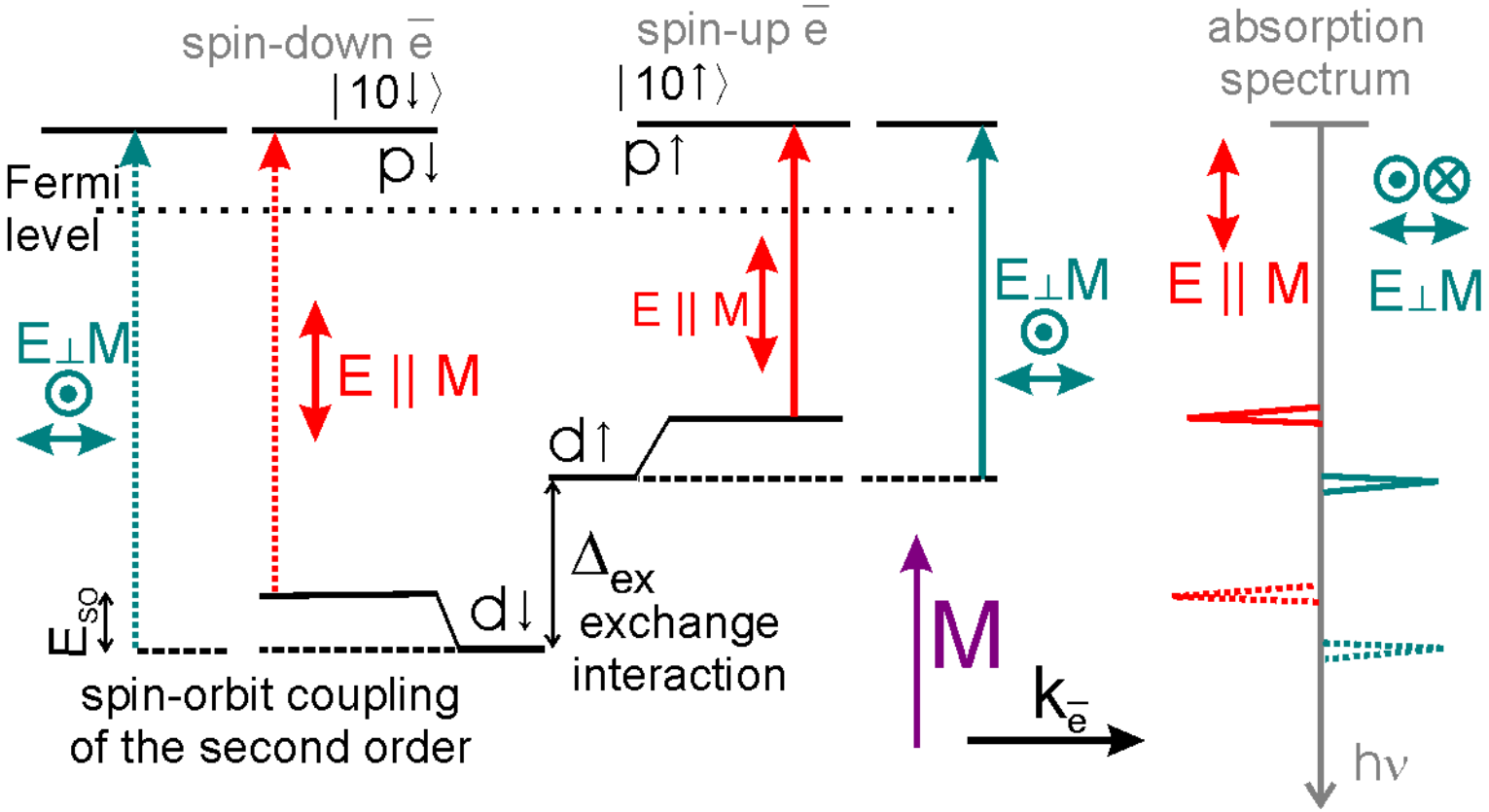
$\Rightarrow$ no MOKE effect

$\Rightarrow$ both SO coupling and exchange must be present to have MOKE.



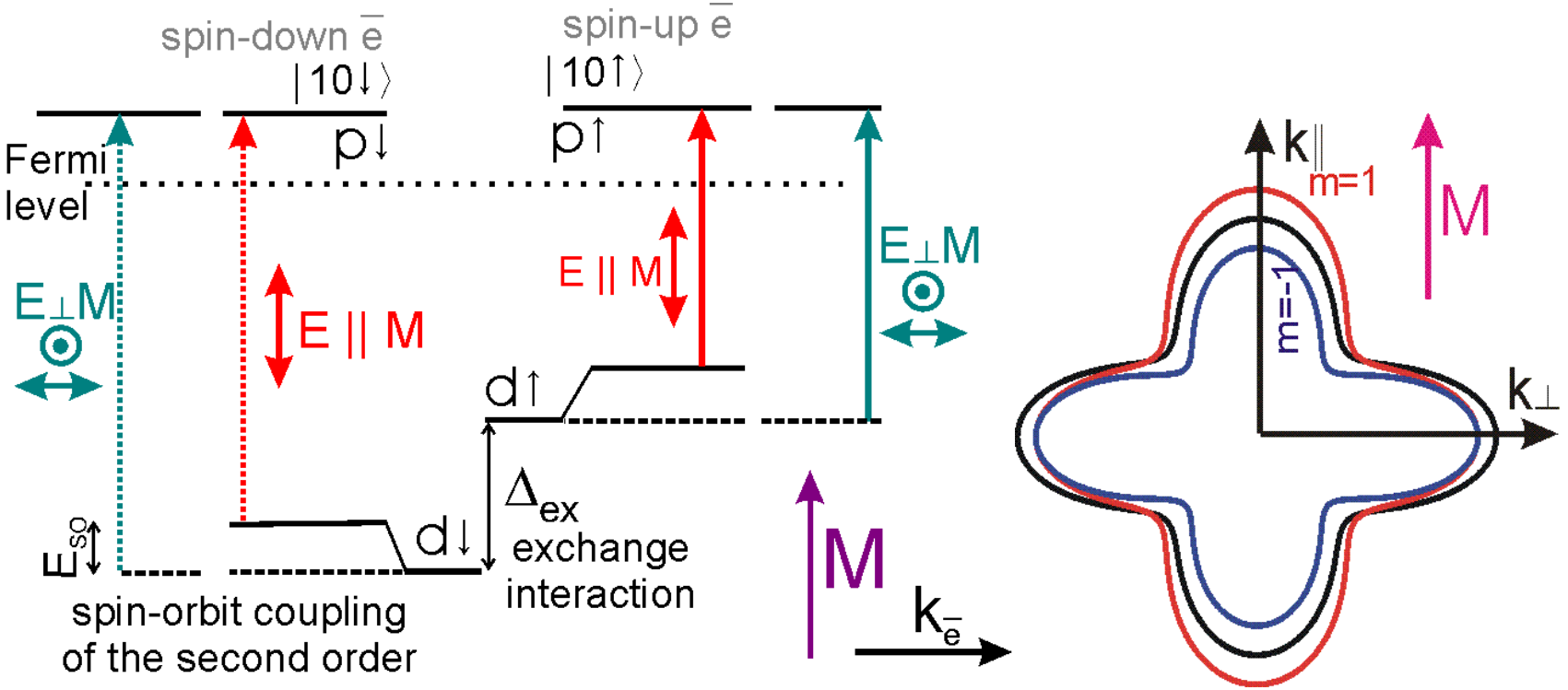


Quadratic Magneto-optical Kerr effect (QMOKE):



QMOKE arises from different absorptions for $\vec{E} \perp \vec{M}$ and $\vec{E} || \vec{M}$.

Quadratic Magneto-optical Kerr effect (QMOKE):



QMOKE arises from different absorptions for $\vec{E} \perp \vec{M}$ and $\vec{E} || \vec{M}$.
$\Rightarrow$ arrises from different electronic structure in $\vec{k}_e \perp \vec{M}$ and $\vec{k}_e || \vec{M}$.

Phenomenological description of MOKE

Inputs are permittivity tensors and layer thicknesses
Phenomenological description based on 4×4 matrix formalism.
(light propagation through layer & continuity of lateral E and H field)
calculated reflectivity matrix
calculated MO Kerr effect

Visible MOKE advantages and disadvantages:

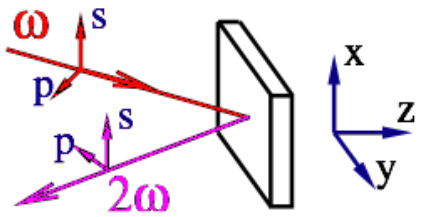
- spatial resolution limited by wavelength limit ($\sim$300nm for visible light) $\rightarrow$ but sub-wavelength resolution demonstrated.
- investigation 'on distance', light can be transported nearby sample by a fibre.
- no need of vacuum or special sample preparation.
- depth resolution about 30nm.
- measurements do not influence sample magnetization.
- high time resolution (down to 20 fs).
- depth selectivity.
- vectorial resolution (possible to determine all magnetization components).
- robust, cheap technique.

BUT:

- spatial resolution limited by wavelength limit.
- easy to overcome Kerr signal by spurious noise (S/N ratio problem).
- not direct information about the electronic structure or magnetic moments etc.

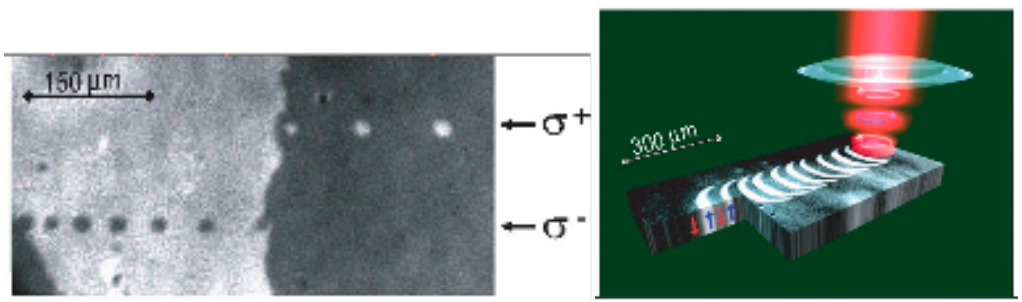
Extensions of MOKE:

- XMCD, XMLD for high photon energy.



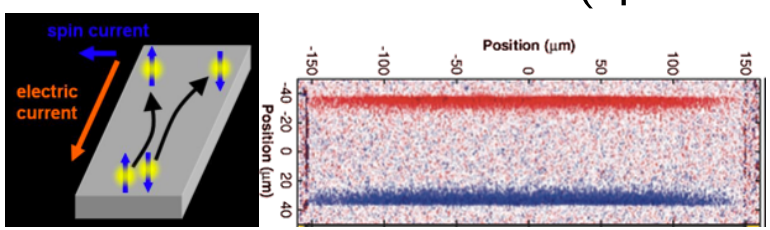
- Non-linear magneto-optics
  $\Rightarrow$ MO second harmonic generation.
- Inverse Faraday effect (ultrafast optical switching).

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



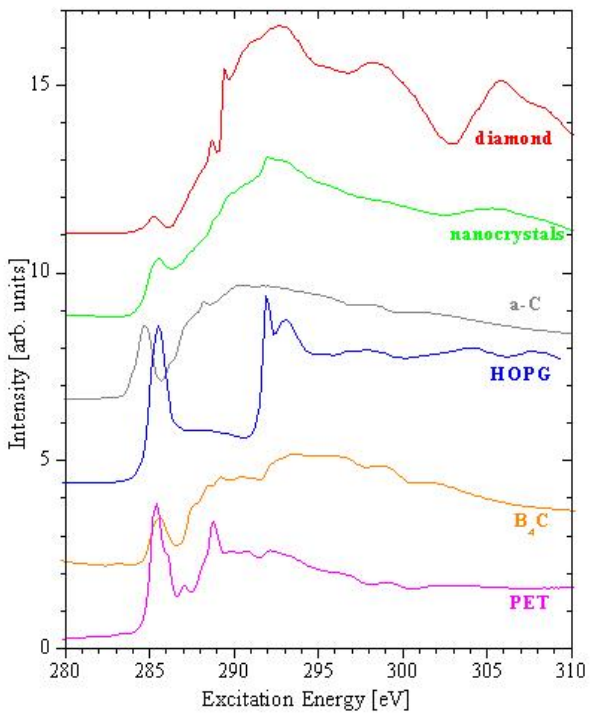
- Observation of spin accumulation in GaAs (spin Hall effect).

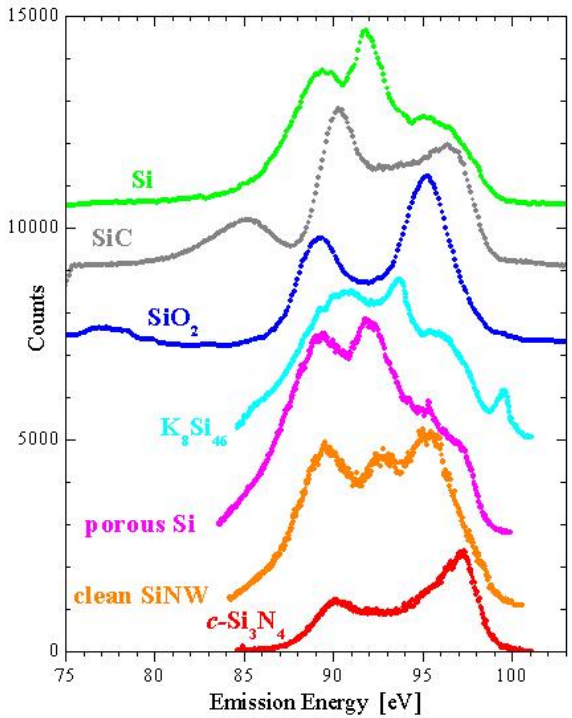
(Kato et al, Science, 2004)



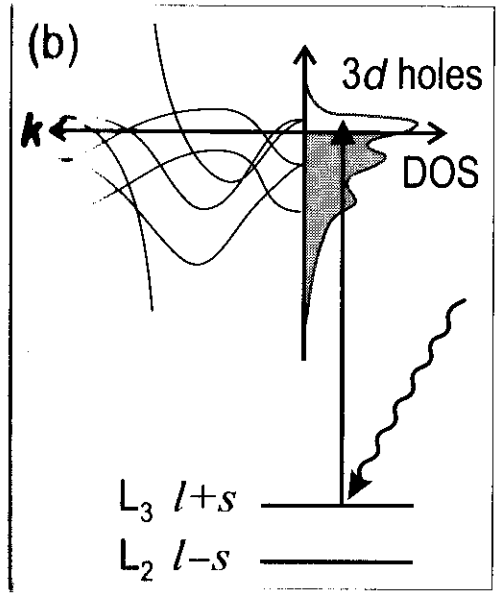
X-ray absorption spectroscopy (XAS):

XAS is extremely sensitive to the chemical state each element, as each element have its own characteristic binding energies. XAS measurements can distinguish the form in which the element crystallizes (for example one can distinguish diamond and graphite, which both entirely consist of C), and can also distinguish between different sites of the same element.

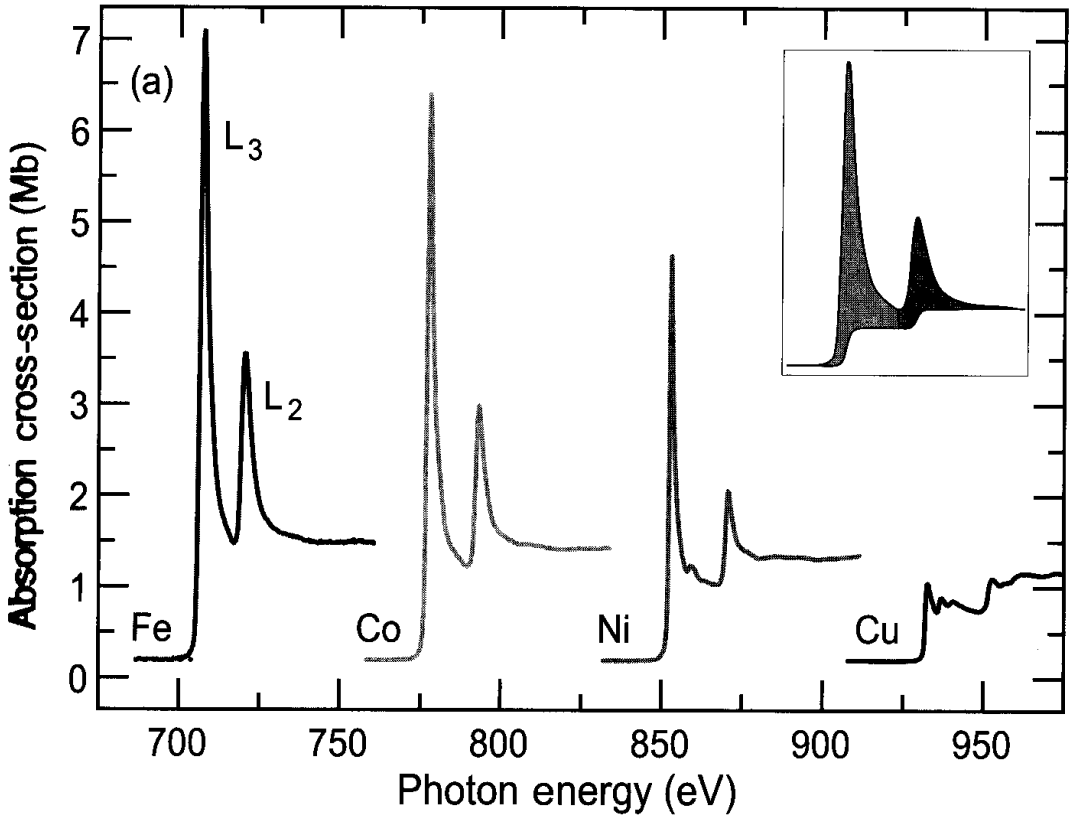




XAS on Fe:



Stöhr, Siegmann, Magnetism: From fundamentals to nanoscale dynamics



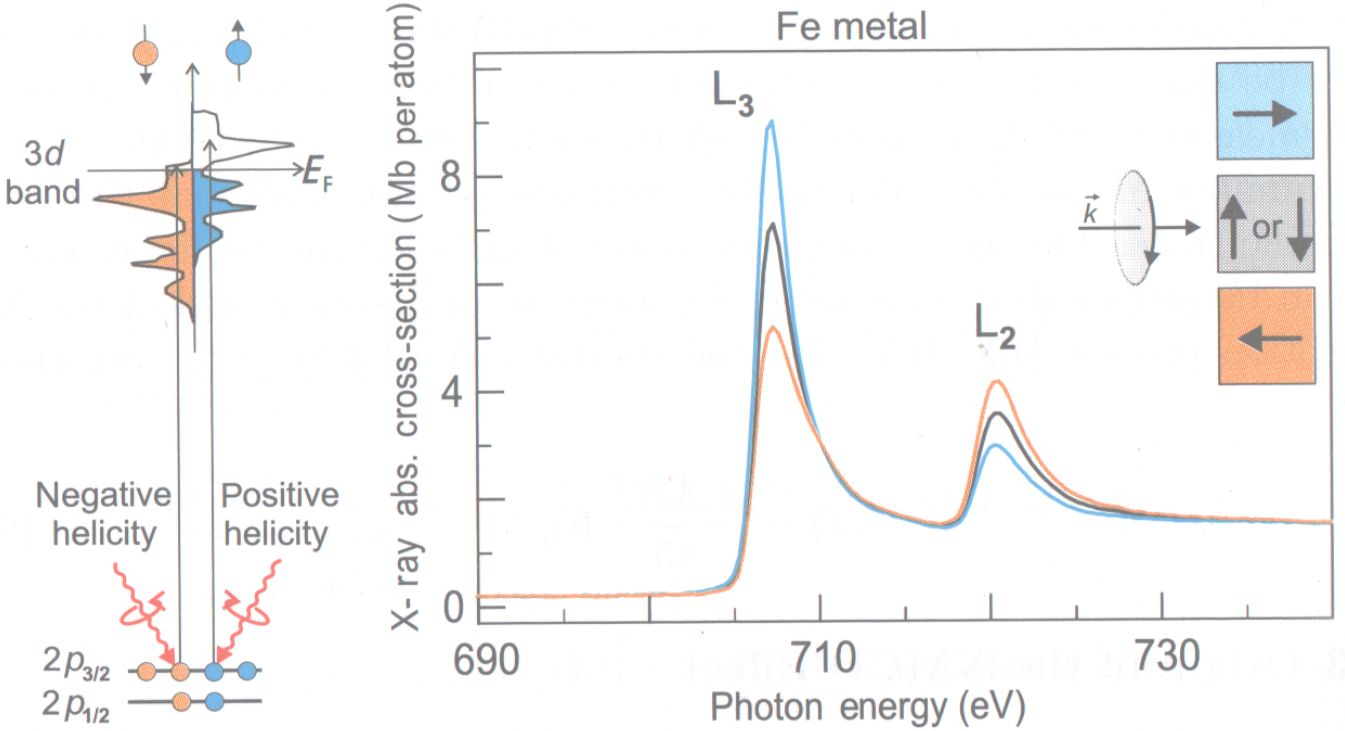
Starting L2, L3 edge (i.e. $2p^{1/2}$, $2p^{3/2}$, respectively):

$$I_{XAS,\ p\to d} \sim N_h$$

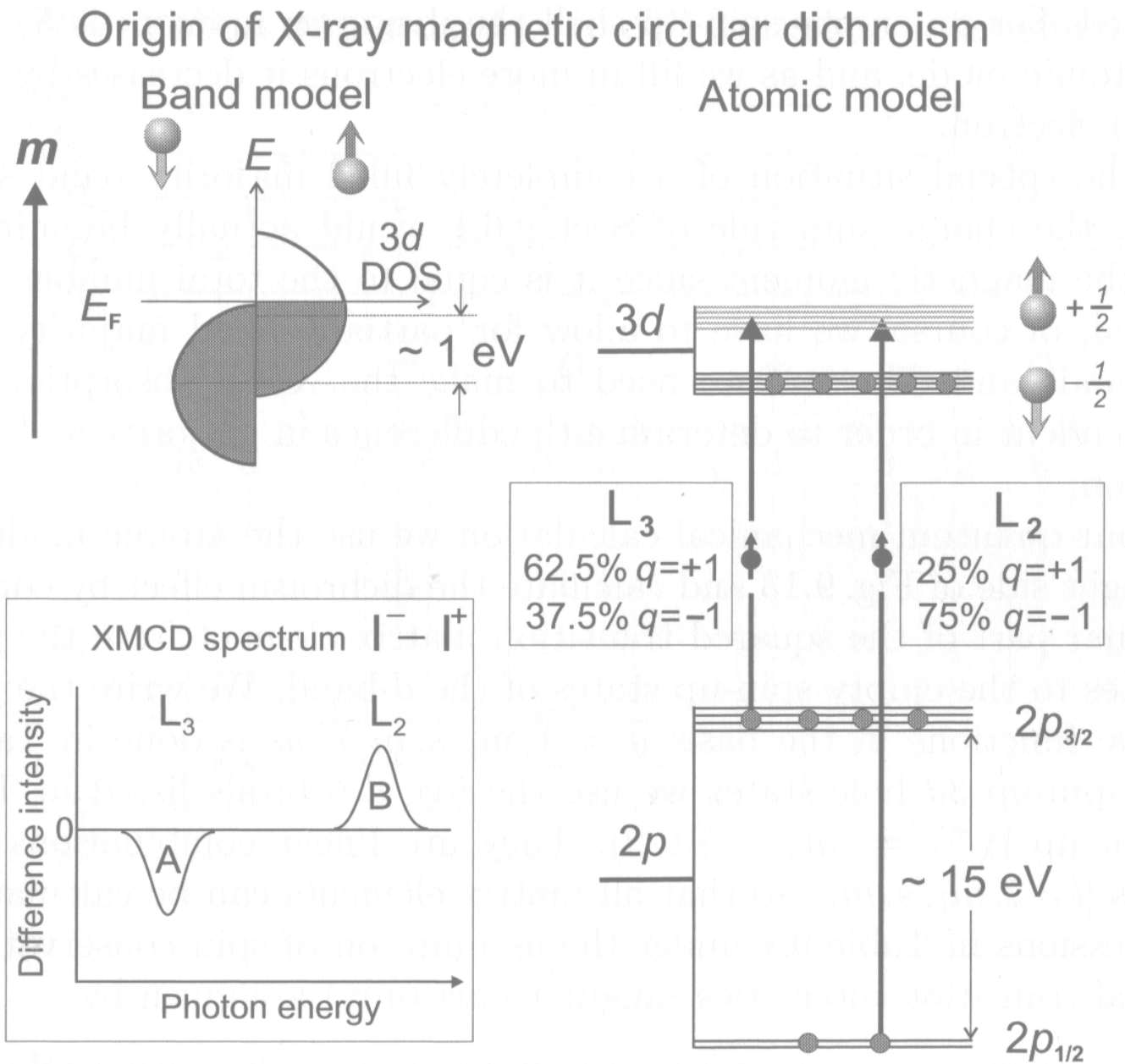
N_h: number of free d-states. $p \to s$ has small contribution.

XMCD: X-ray Magnetic circular dichroism:

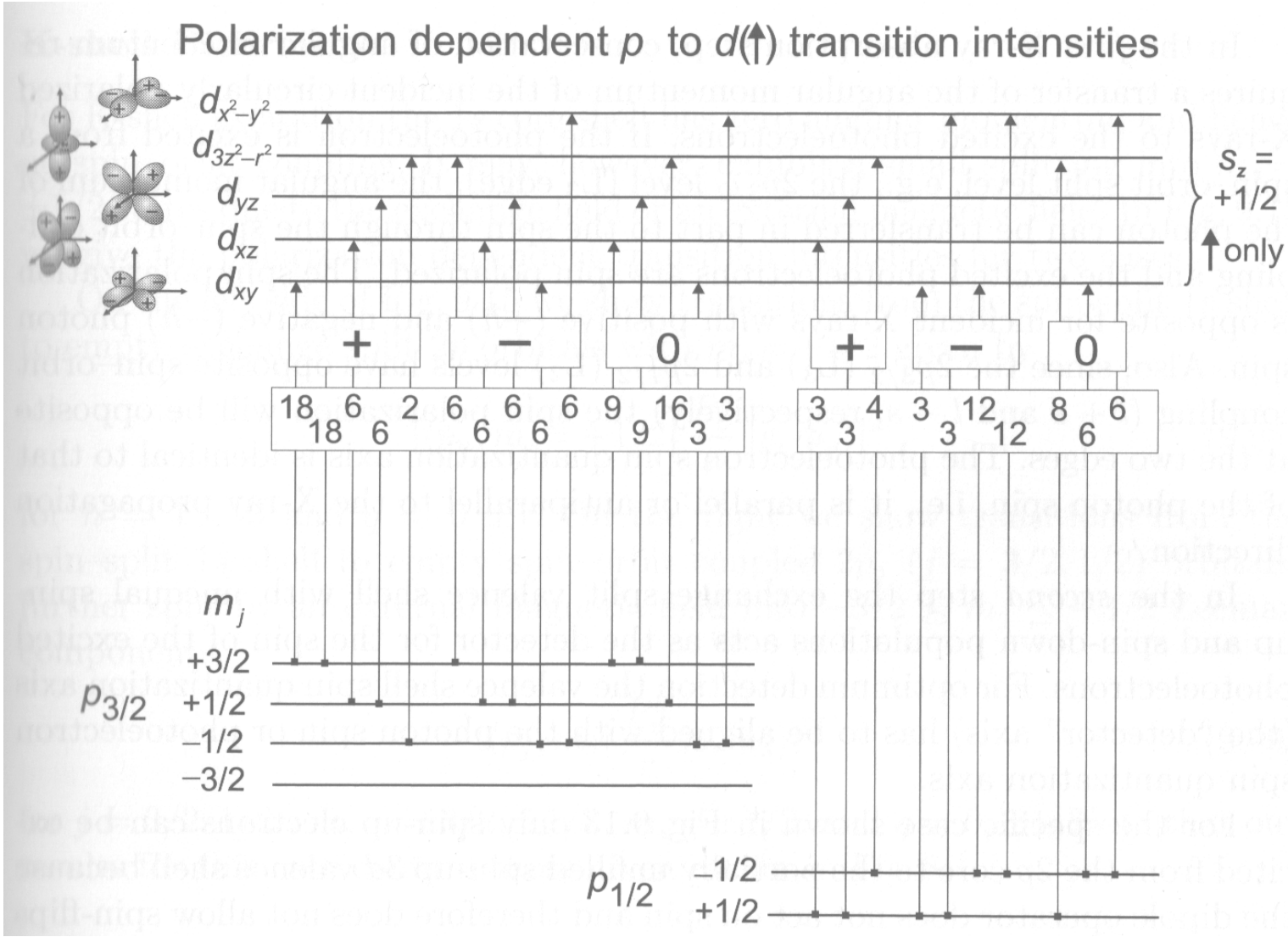
Circular Dichroism: different absorption for circularly left and right light polarization.



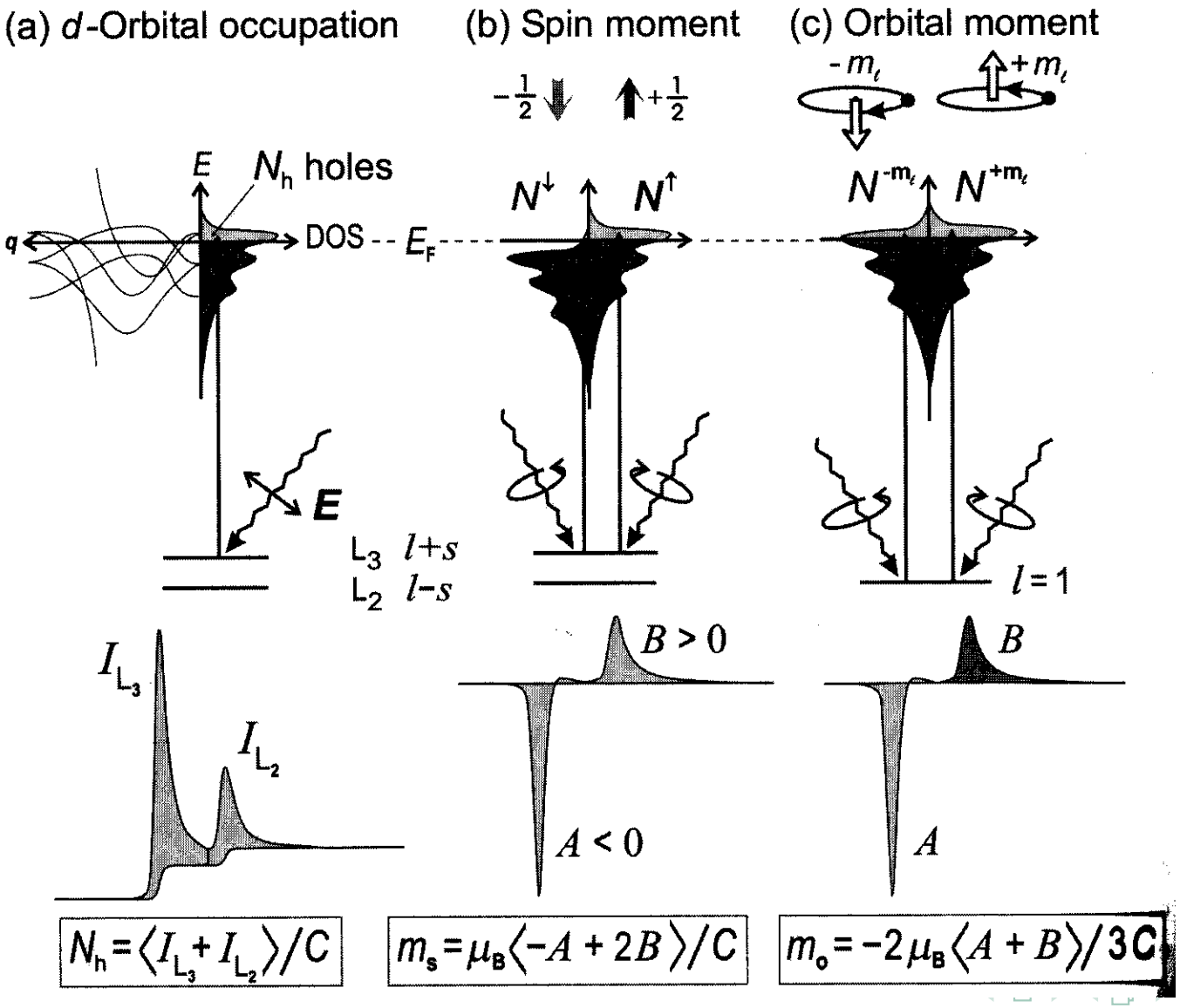
Different absorbed intensity for opposite magnetization orientations.


Origin of X-ray magnetic circular dichroism
Band model
Atomic model
m
E
3d
DOS
E_F
~ 1 eV
3d
+1/2
−1/2
L_3
62.5% q=+1
37.5% q=−1
L_2
25% q=+1
75% q=−1
XMCD spectrum $I^- - I^+$
L_3
L_2
A
B
0
Difference intensity
Photon energy
$2p_{3/2}$
2p
~ 15 eV
$2p_{1/2}$

XMCD: Detailes p $\rightarrow$ d transition



XMCD: sum rules:



Advantages of X-ray spectroscopies:

- element selective.
- quantitative determination of material characterization (e.g. magnetic moment, orbital moment).
- can be both interface or bulk sensitive.
- can provide excellent lateral resolution ($\sim 15\,\mathrm{nm}$).
- can provide excellent time resolution ($\sim 100\,\mathrm{fs}$).

Disadvantages:

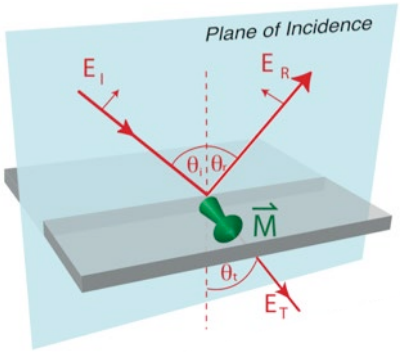
- due to width of the initial (core) line, the energy resolution is limited to $\sim$1 eV.
- synchrotron needed (or high harmonics, see next slides)

DC conductivity:

DC conductivity can be understand as a limit of absorption spectroscopy for $\omega \to 0$.
Due to different history and different available experimental techniques, different names are used in each area:

Transport (dc)	Optics	X-ray
conductivity	absorption	$\sim$ X-ray absorption (XAS)
Hall effect	MOKE effect	XMCD
quadratic-Hall effect	quadratic MOKE (QMOKE)	$\sim$ X-ray linear magnetic dichroism
Anisotropy magneto-resistance (AMR)	Cotton-Mouton, Voigt effect	X-ray linear magnetic dichroism

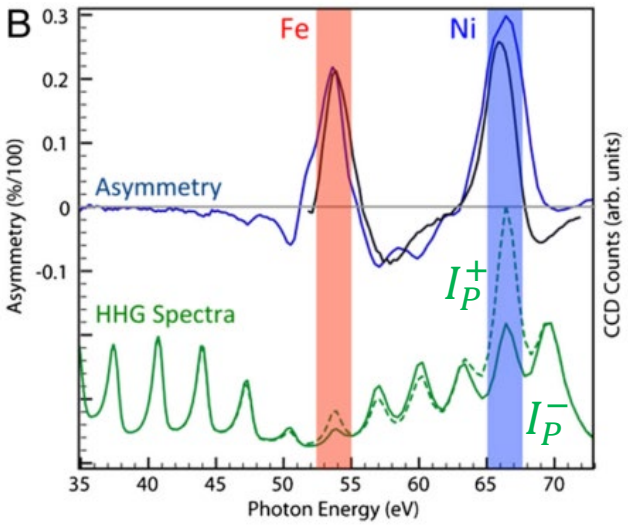
Unique Tabletop Experimental setup: EUV MOKE

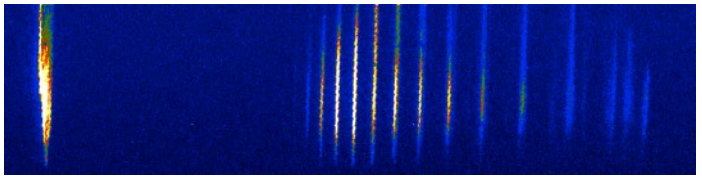


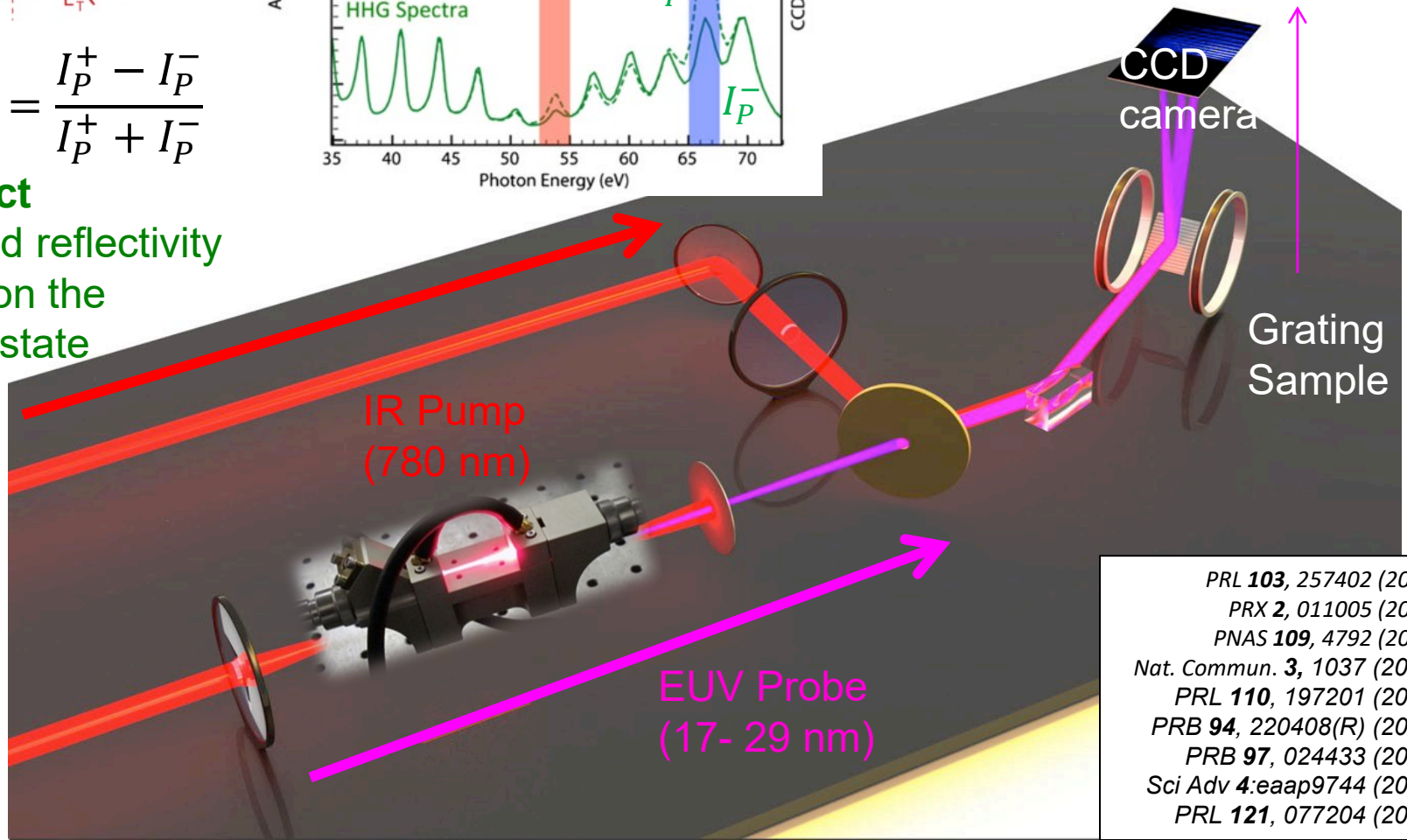
$$\vec{M} \;\alpha\; A = \frac{I_P^+ - I_P^-}{I_P^+ + I_P^-}$$

Kerr Effect

p-polarized reflectivity depends on the magnetic state

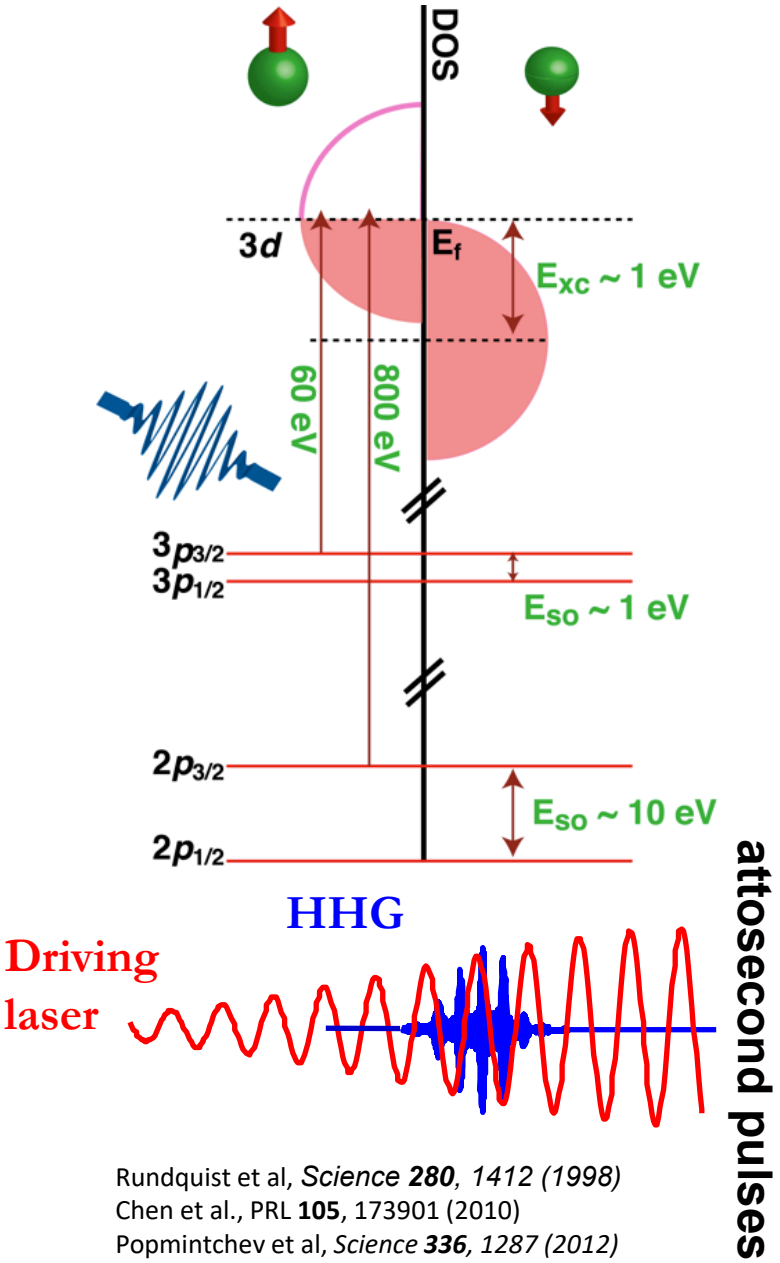




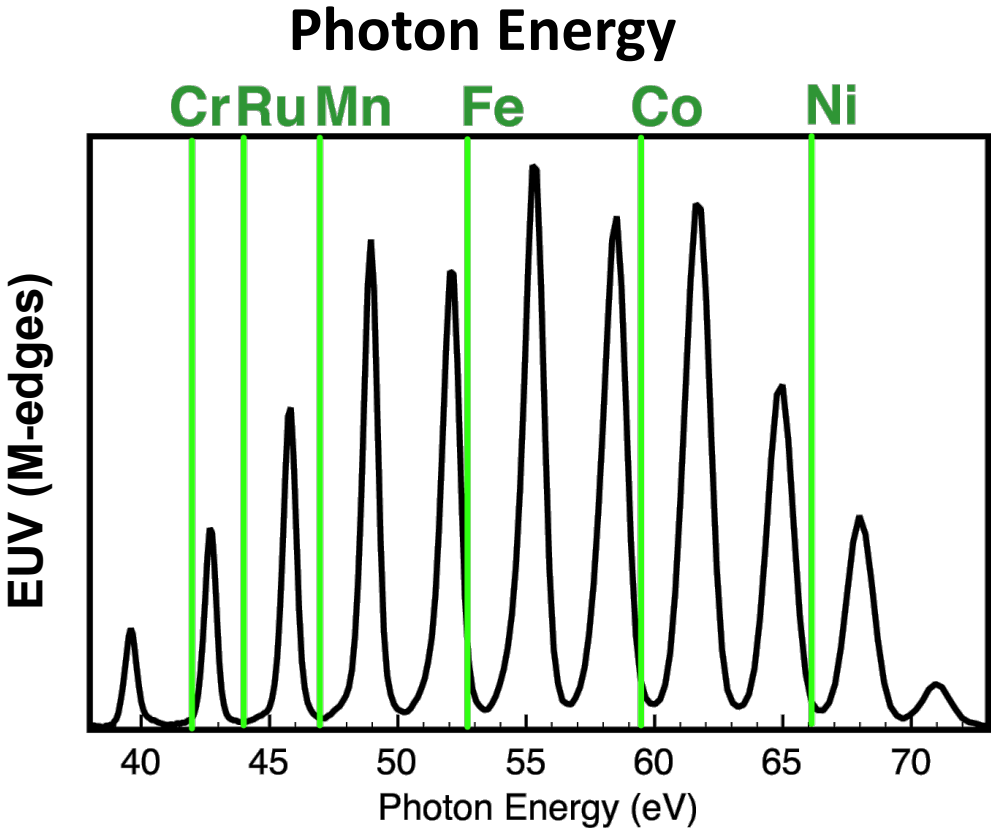


PRL ***103***, 257402 (2009)
PRX ***2***, 011005 (2012)
PNAS ***109***, 4792 (2012)
Nat. Commun. ***3***, 1037 (2012)
PRL ***110***, 197201 (2013)
PRB ***94***, 220408(R) (2016)
PRB ***97***, 024433 (2018)
Sci Adv ***4***:eaap9744 (2018)
PRL ***121***, 077204 (2018)

M-edge Spectroscopy: a unique tool



Rundquist et al, *Science* ***280****, 1412 (1998)*
Chen et al., PRL **105**, 173901 (2010)
Popmintchev et al, *Science* ***336****, 1287 (2012)*



UNIQUE ADVANTAGES

- Element selective
- Multiple elements simultaneously
- <<10fs time resolution, no jitter
- Span entire M edges, L edges

PHYSICAL REVIEW B VOLUME 12, NUMBER 11 1 DECEMBER 1975

Calculation of the M_{23} magneto-optical absorption spectrum of ferromagnetic nickel

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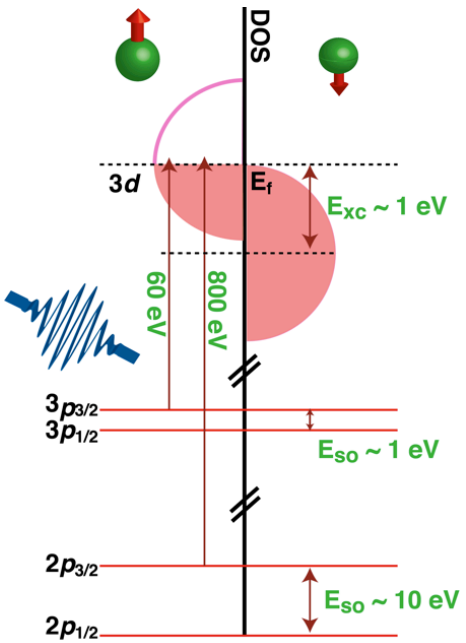
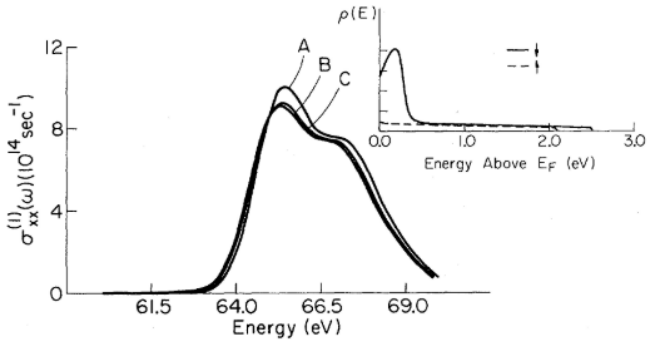
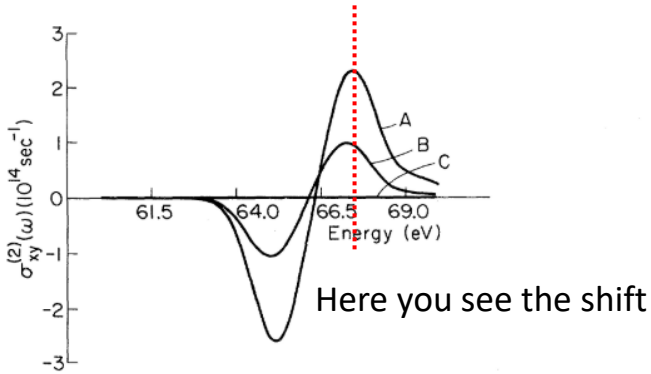
(Received 28 April 1975)

The M_{23} magneto-optical absorption spectrum of ferromagnetic nickel is calculated using an approach similar to the component state-density method that has been successfully used in obtaining valence-band emission and absorption x-ray spectra of metals. The M_{23} magneto-optical effects result predominantly from spin-orbit splitting of the $3p$ core state in conjunction with the final d-state spin polarization. The calculated spectrum exhibits features that are directly related to electronic structure parameters including the $3p$ core spin-orbit splitting, and the unfilled d-band spin polarization. Temperature variations in the magneto-optical structure can be used to determine separately the exchange-splitting variation and spin-wave excitation contributions to the decrease in the magnetization. Experimental verification of these predictions should provide insight into the applicability of the Stoner model to ferromagnetic nickel and may be helpful in resolving some of the apparently conflicting results of other experimental probes of the spin polarization near the Fermi level in nickel.







Here you see the shift

The spin-wave excitation mechanism by itself, if no consequent decrease in exchange splitting occurs, will simply decrease the magnitude of magneto-optical absorption without changing its shape. A decrease in the Stoner exchange splitting will change the shape of the magneto-optical absorption spectrum. Changes in the ordinary optical spectrum are much weaker, and consequently are that much more difficult to detect and interpret.

Time- resolved experiments: we attempted to answer this question: Stoner vs. Heisenberg via reduction of magnetic contrast at the M-edge

PHYSICAL REVIEW B **94**, 220408(R) (2016)

Stoner versus Heisenberg: Ultrafast exchange reduction and magnon generation during laser-induced demagnetization

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PHYSICAL REVIEW B **97**, 024433 (2018)

Editors' Suggestion

Direct measurement of the static and transient magneto-optical permittivity of cobalt across the entire *M*-edge in reflection geometry by use of polarization scanning

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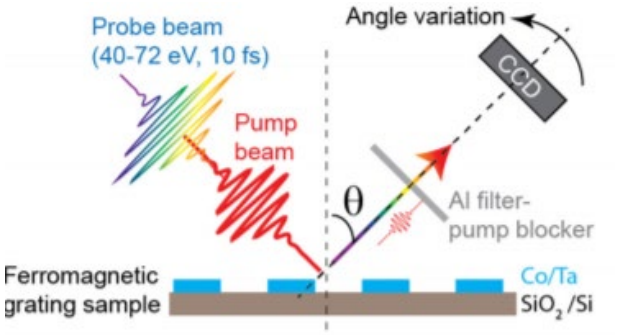
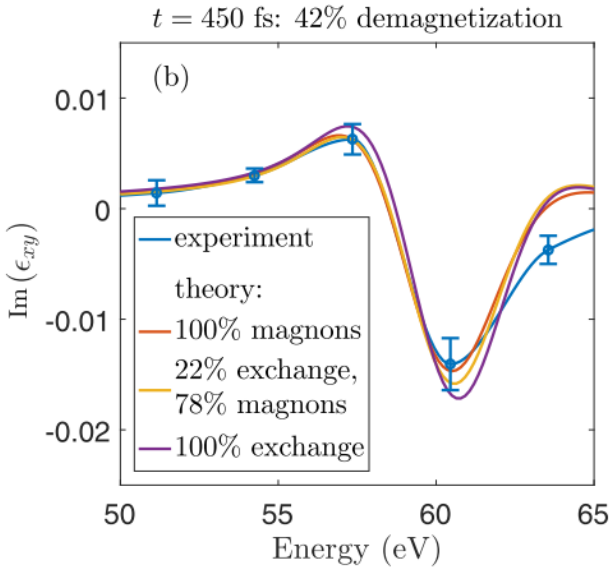
[3]*Quantum Electromagnetics Division, National Institute of Standards and Technology, Boulder, Colorado 80305, USA*
[4]*Department of Physics and Astronomy, Uppsala University, S-75120 Uppsala, Sweden*





Second paper: we showed that you cannot exclude magnons even in the reduction of the moment in Cobalt (which has the strongest Weiss field)

Here we directly compared magneto-optical results to photoemission data

Band structure in Ni

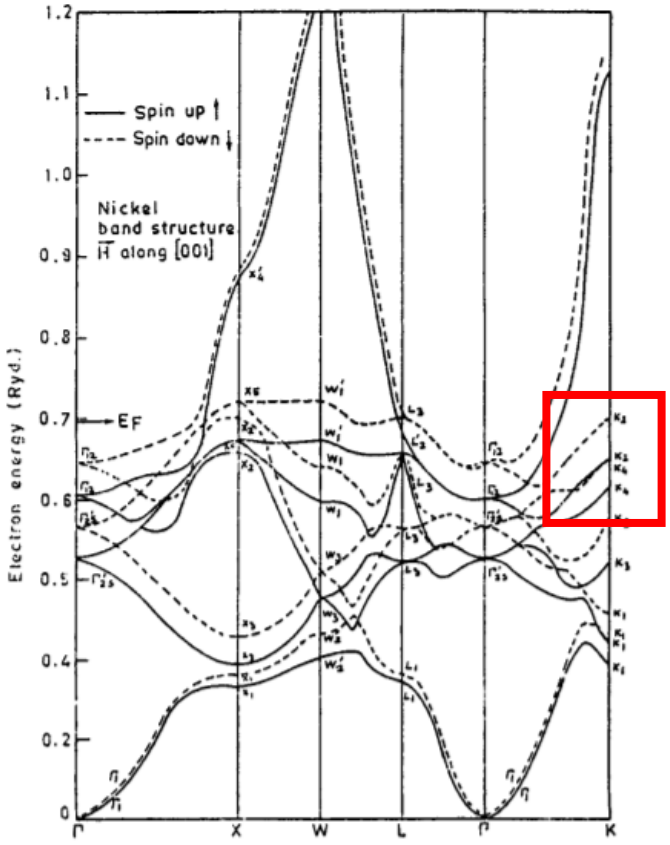
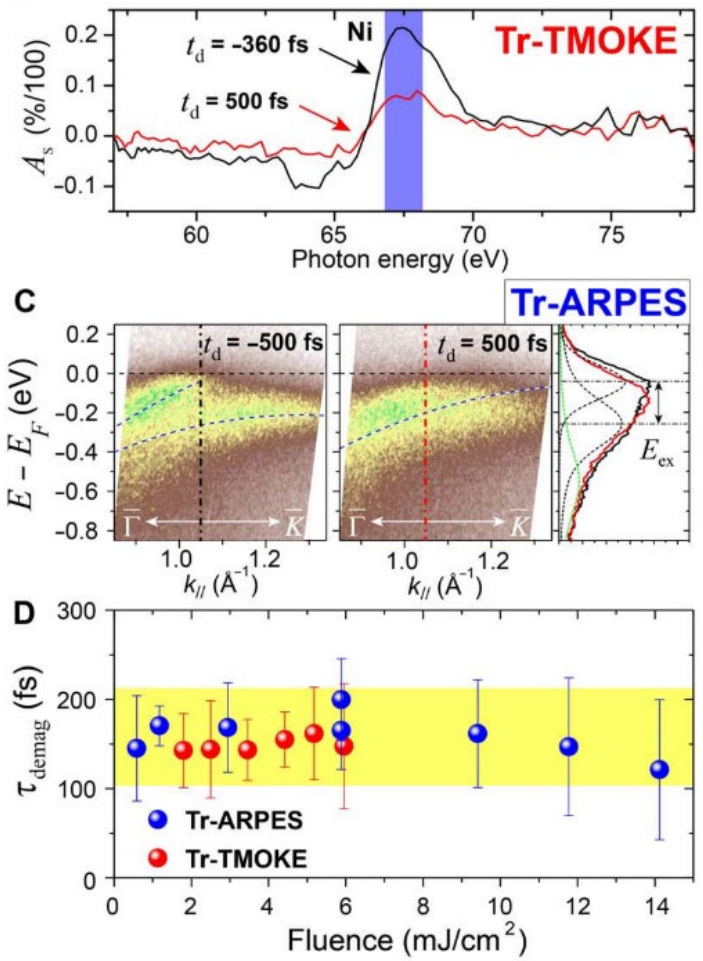


Fig. 1. The band structure of ferromagnetic nickel

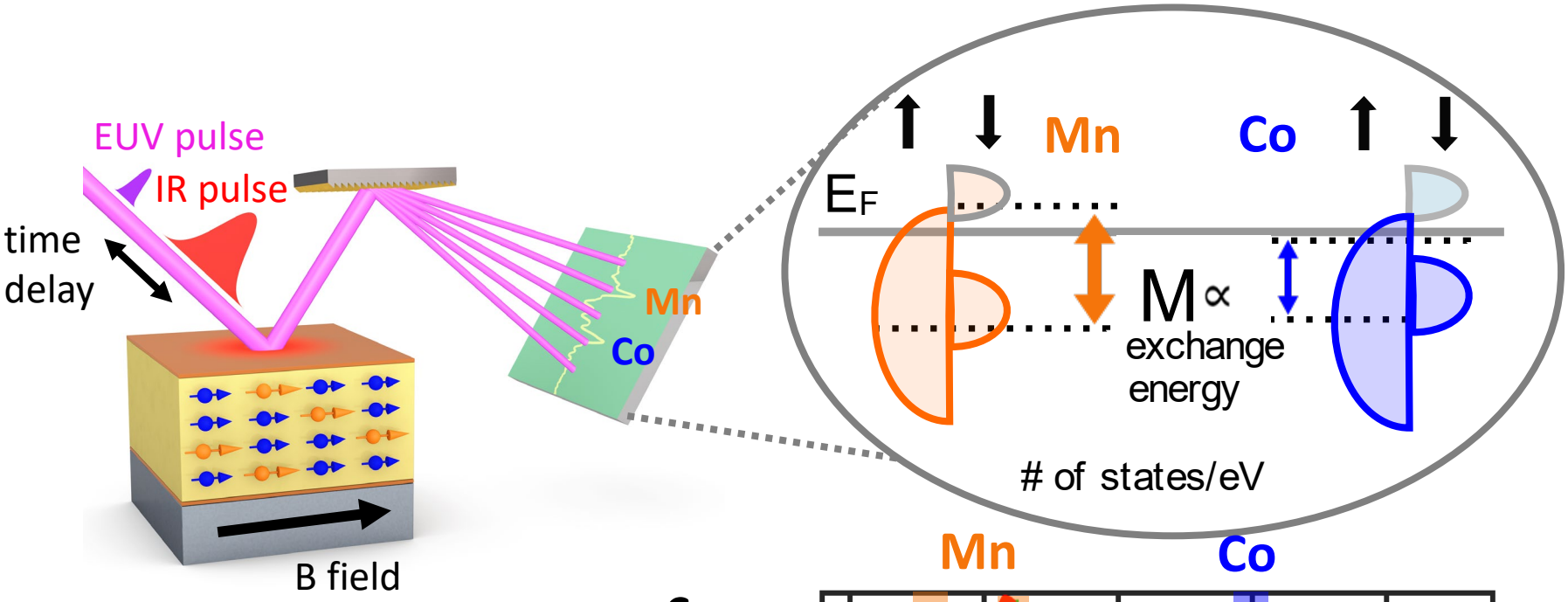
PHYSICS

Critical behavior within 20 fs drives the out-of-equilibrium laser-induced magnetic phase transition in nickel

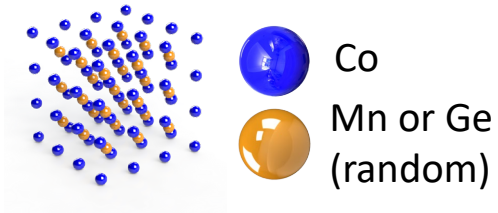
Phoebe Tengdin,[1]* Wenjing You,[1]* Cong Chen,[1] Xun Shi,[1†] Dmitriy Zusin,[1] Yingchao Zhang,[1] Christian Gentry,[1] Adam Blonsky,[1] Mark Keller,[2] Peter M. Oppeneer,[3] Henry C. Kapteyn,[1] Zhensheng Tao,[1†] Margaret M. Murnane[1]



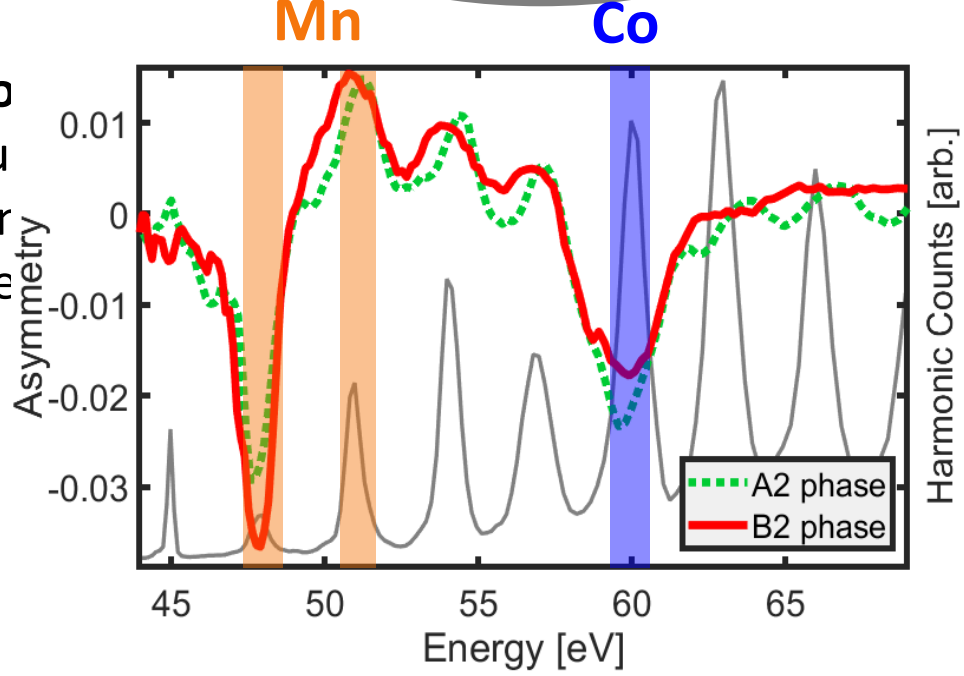
Light induced spin transfer: Half metallic System



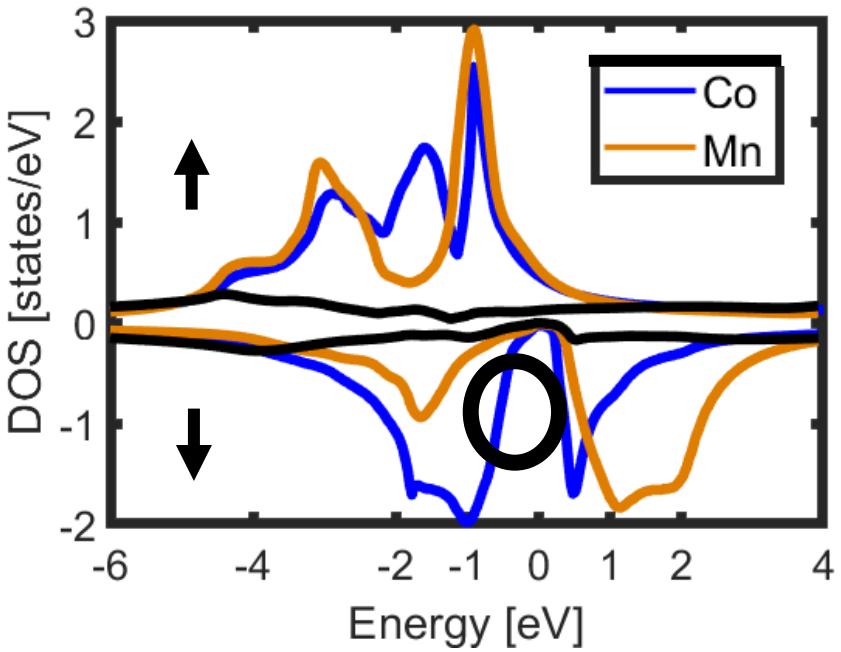
- Half metallic: one spin state condu
- Half-metal gap in Mn is bigger thar
- Expect to see slower dynamics due

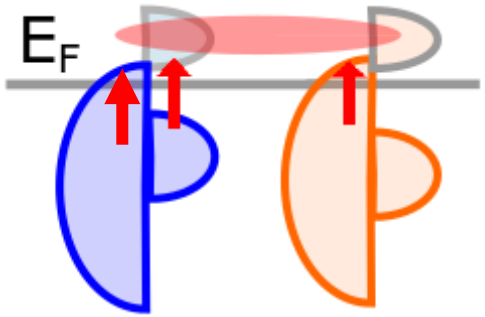


Half-metal Phase



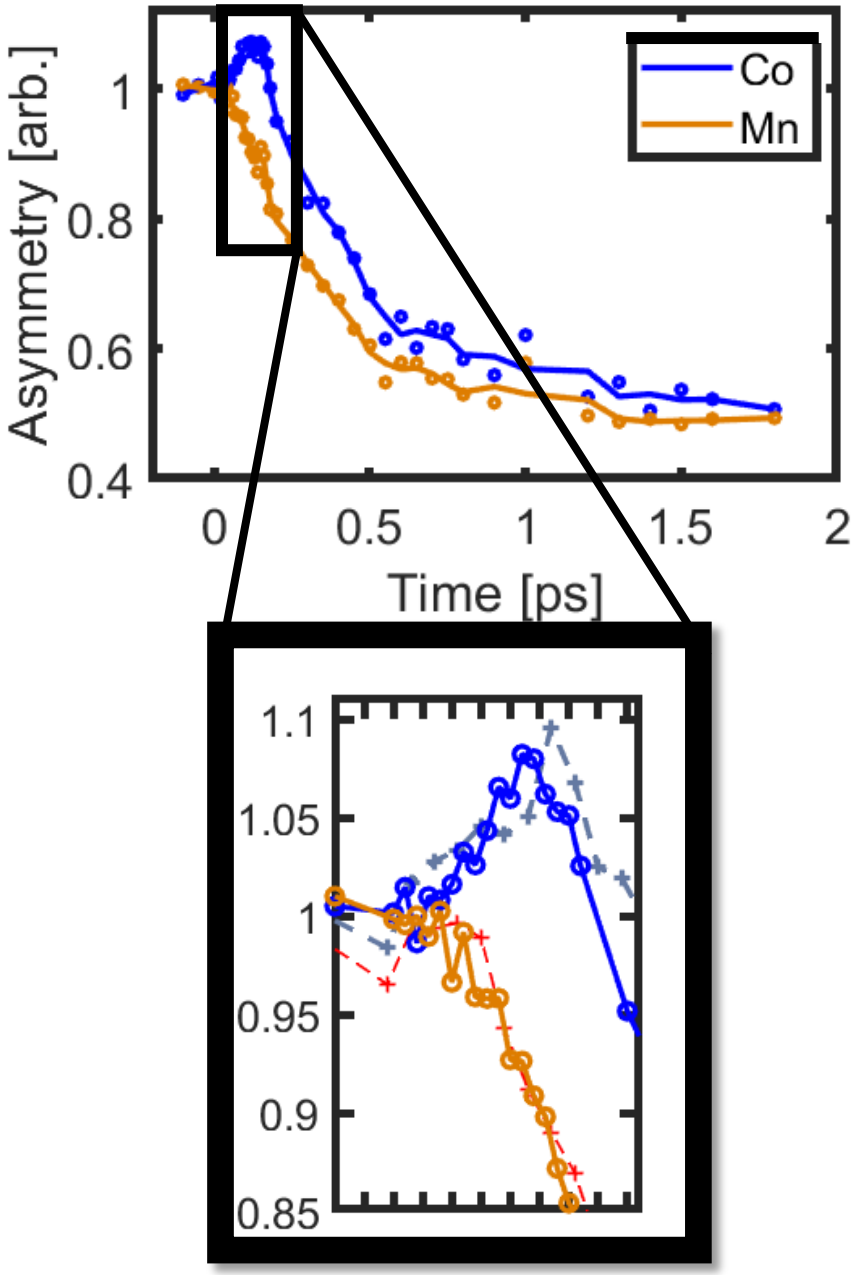
Ultrafast transfer of magnetization between different elements





Co – minority spin excitation

Mn – majority spin excitation



Tengdin, P. et. al., *Sci. Adv.* **6** *:* eaaz1100, (2020)