Magneto-optical Kerr effect (MOKE)

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Photon-photon spectroscopies (absorption) I:

Type of investigations (polarized light x non-polarized light, reflection x transmission, different properties for different polarizations):

- ellipsometry, reflectometry, transmittivity, magneto-optical Kerr effect spectroscopy, magnetic linear/circular dichroism/birefringence, etc.

Photon-photon spectroscopies (absorption) II:

Different energy ranges probes different part of the electronic structure of the matter.

dc conductivity, THz=far-infrared: energy about $kT \approx 30\,\mathrm{meV}$. Excites vicinity of the Fermi surface (all dc conductivities, VA characteristics,).

extended visible light (mid-infrared – far-UV): $\sim 30\,\mathrm{meV}$ - $100\,\mathrm{eV}$. Excites band structure of the matter. (reflectometry, ellipsometry, etc). Both starting and final states are in the band structure (both unknown).

X-ray: $\sim 120\,\mathrm{eV}$ – $120\,\mathrm{keV}$. Excites deep core levels of the atoms. XAS (X-ray absorption spectroscopy). Starting levels are from core levels and hence they are well known.

Photon-photon spectroscopies (absorption) III:

The underlying physics is based on absorption (the same as emission) of the photons.

1. material's absorption of photons i.e. imaginary part of permittivity $\Im(\varepsilon)$ [determined usually by electric-dipole approximations].
2. real part of permittivity $\Re(\varepsilon)$ by Kramers-Kronig relations
   Then, complex optical properties of matter are known (complex permittivity ε or complex refraction index N or complex conductivity σ, $\varepsilon = N^2 = 1 + i\sigma/\omega$)
3. optical response of the multilayer structure (including interface roughness etc.)

Conductivity (and hence absorption of the photon)

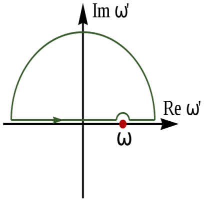
Kubo formula: conductivity determination.

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

where

- $\langle i|$, $|f\rangle$: initial and final states, respectively.
- $p_\pm = p_x \pm ip_y$, $p_x = i\hbar\partial/\partial x$, 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.

Kramers-Kroning relations I



Purely based on mathematical relation between real and imaginary part of 'polite' functions (Cauchy integral).
1) mathematics: Cauchy residual theorem states (for any function without poles in integration area):

$$\oint \frac{\chi(\omega')}{\omega' - \omega}\, d\omega' = 0$$

2) for function $\chi(\omega')/(\omega' - \omega)$, we create pole in point $\omega' = \omega$. Hence Cauchy integral becomes (assuming the integral over 'arc' is zero, i.e. function χ is enough small at infinity):

$$\oint \frac{\chi(\omega')}{\omega' - \omega}\, d\omega' = \mathcal{P}\int_{-\infty}^{\infty} \frac{\chi(\omega')}{\omega' - \omega}\, d\omega' - i\pi\chi(\omega) = 0.$$

I.e. function in point $\chi(\omega)$ equals to the integral over whole ω'!

Kramers-Kroning relations II

Rearranging:

$$\chi(\omega) = \frac{1}{i\pi}\mathcal{P}\int_{-\infty}^{\infty} \frac{\chi(\omega')}{\omega' - \omega}\, d\omega'.$$

3) Thermodynamics shows $\chi(-\omega) = \chi^*(\omega)$ (because time flows only in one direction). Hence, relation between $\Re(\chi)$ and $\Im(\chi)$ is

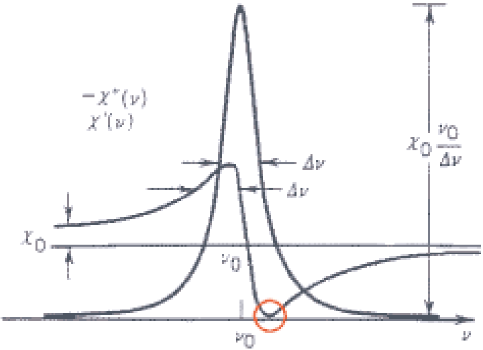
$$\Re(\chi(\omega)) = \frac{2}{\pi}\mathcal{P}\int_{0}^{\infty} \frac{\omega'\Im(\chi(\omega'))}{\omega'^2 - \omega^2}\, d\omega'$$

$$\Im(\chi(\omega)) = -\frac{2\omega}{\pi}\mathcal{P}\int_{0}^{\infty} \frac{\Re(\chi(\omega'))}{\omega'^2 - \omega^2}\, d\omega'$$

Which are famous Kramers-Kronig relations.

Kramers-Kroning relations III

Example: Lorenzian function

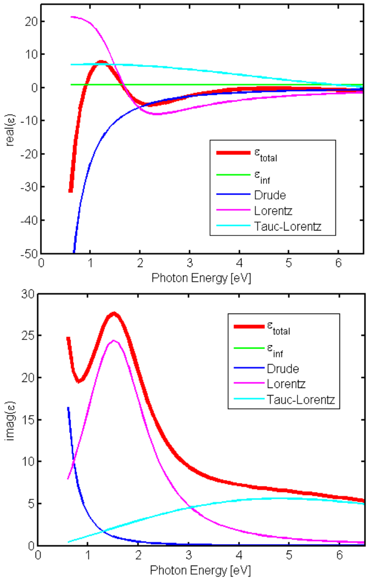


Absorption line (i.e. $\Im(\varepsilon)$) is described by Lorenzian function. Kramers-Kronig then determines its real part.

Description of optical properties of materials I:

Optical permittivity ε: $\varepsilon = \Re(\varepsilon) + i\Im(\varepsilon)$

- $\Im(\varepsilon)$ means light absorption.
- $\Re(\varepsilon)$ and $\Im(\varepsilon)$ are related by Kramers-Kronig (KK) relations.
- in order to assure KK-relations when determining ε, it is advantageous to describe ε by set of optical functions, each one fulfilling KK-relations. For example:
  1. Drude term (free electron contribution).
  2. Lorentz term (resonance line).
  3. Tauc-Lorentz (semiconductor gap).
  4. etc.

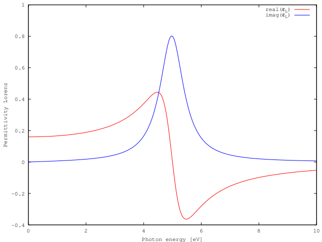


Description of optical properties of materials II:

Lorentz term - description of absorption between two energy levels

$$\varepsilon = \frac{A}{\omega^2 - \omega_0^2 + i\Gamma\omega}$$

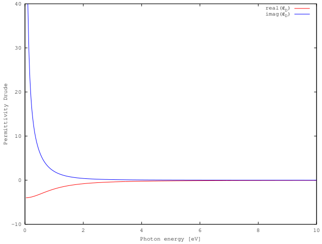
- ω: Photon energy
- ω_0: Energy distance between the lines (i.e. resonance energy)
- A: Amplitude (probability of the absorption)
- Γ: Width of the line (sharpness of the absorption) (i.e. full-width-at-half-maxima: FWHM)



Description of optical properties of materials III:

Drude term - description of absorption due to free electrons (i.e. due to conductivity)

Like Lorenz, for $\omega_0 = 0$



Description of optical properties of materials IV:

Optical permittivity ε: $\varepsilon = \Re(\varepsilon) + i\Im(\varepsilon)$
Another way how to describe spectra:

1. imaginary part $\Im(\varepsilon)$ given as arbitrary spectra for energies E_i, $\Im(\varepsilon_i)$
2. real part $\Re(\varepsilon)$ then calculated from Kramers-Kronig relation

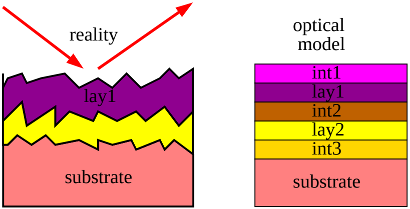
Optics of multilayers I

Total optical response of multilayer described by reflection matrix:

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

Reflection matrix is the only quantity accessible for sample investigations by optical means. Roughness of the interfaces is included by:

1. usually described by effective sub-layers.
2. their optical properties described by e.g. effective-medium-approximation (EMA).



Optics of multilayers II

Total optical response of multilayer described by reflection matrix:

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

Different quantities can be investigated on reflection:

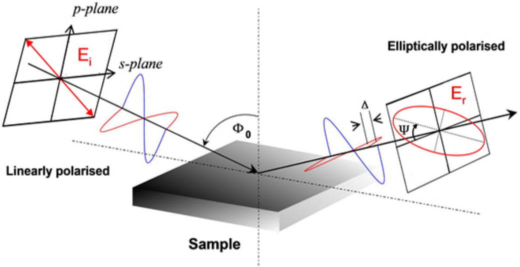
- reflectivity (unpolarized light): $I = 1/2(|r_{ss}|^2 + |r_{pp}|^2)$
- reflectivity of s-polarized light: $I_s = |r_{ss}|^2$
- magneto-optical Kerr s-effect: $\Phi_s = \theta_s + i\epsilon_s = \frac{r_{sp}}{r_{ss}}$
- ellipsometry: $\rho = \tan\Psi \exp(-i\Delta) = \frac{r_{pp}}{r_{ss}}$
- magnetic linear dichroism for s-wave ($\vec{M}$ in-plane): $MLD = |r_{ss}(M \parallel s)|^2 - |r_{ss}(M \parallel p)|^2$

Ellipsometry I

Ellipsometry measures complex ratio of diagonal reflection coefficients:

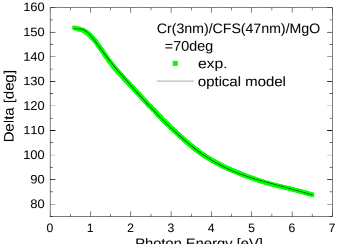
$$\rho = \tan\Psi \exp(-i\Delta) = \frac{r_{pp}}{r_{ss}}$$

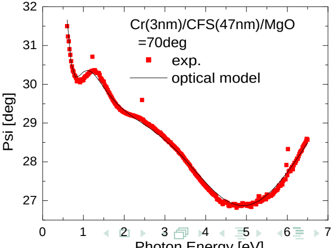
- Ψ, Δ: so-called ellipsometric angles Ψ, Δ
- Ψ expresses ratio of reflected s- and p-waves
- Δ expresses phase difference of reflected s- and p- waves.



Ellipsometry II

- experimental setup provides spectra of Ψ, Δ
- those spectra are fitted into optical model, where various parameters can be free parameters in the fit (but not all at the same fit):
  - optical constants of a given layer (can be further described by a various functions)
  - layer thicknesses
  - interface roughnesses
- Example of fit to spectra of Ψ, Δ





Outline

Magneto-optical Kerr effect:

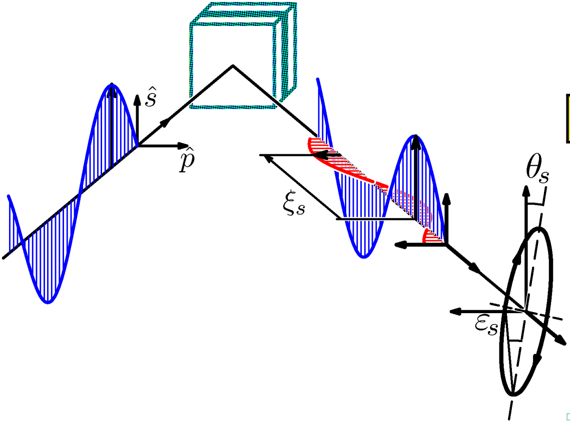
- Change of optical properties (polarization state, reflectivity) by presence/change of magnetization of the sample.

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:

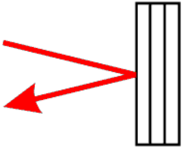
Due to historical reasons, there are different names for MO effects measured in reflection and transmission.

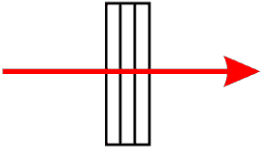
Kerr effect:

- measured in reflection.
- discovered 1876.

Faraday effect:

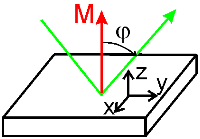
- measured in transmission.
- discovered 1845.

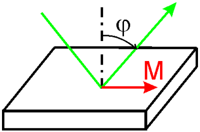


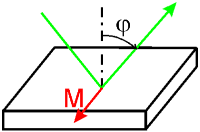


MOKE configurations and permittivity tensor:

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







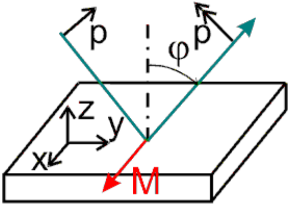
Polarization induced by magnetization: $\Delta \vec{P}_M = \varepsilon_1 (\vec{M} \times \vec{E})$

$$\begin{bmatrix} \varepsilon_0 & -\varepsilon_1 m_z & 0 \\ \varepsilon_1 m_z & \varepsilon_0 & 0 \\ 0 & 0 & \varepsilon_0 \end{bmatrix} \quad \begin{bmatrix} \varepsilon_0 & 0 & \varepsilon_1 m_y \\ 0 & \varepsilon_0 & 0 \\ -\varepsilon_1 m_y & 0 & \varepsilon_0 \end{bmatrix} \quad \begin{bmatrix} \varepsilon_0 & 0 & 0 \\ 0 & \varepsilon_0 & -\varepsilon_1 m_x \\ 0 & \varepsilon_1 m_x & \varepsilon_0 \end{bmatrix}$$

$\Phi_{s/p}(m_z)$ $\quad$ $\Phi_{s/p}(m_y)$ $\quad$ $\Delta r_{pp}(m_x)$

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, on change in polarization of the reflected light appears).



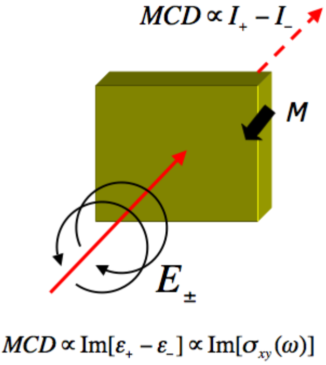
MO effect III: Magnetic dichroism and birefringence:

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

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

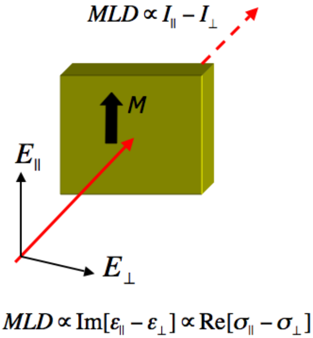
Magnetic circular dichroism (MCD):

- 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.



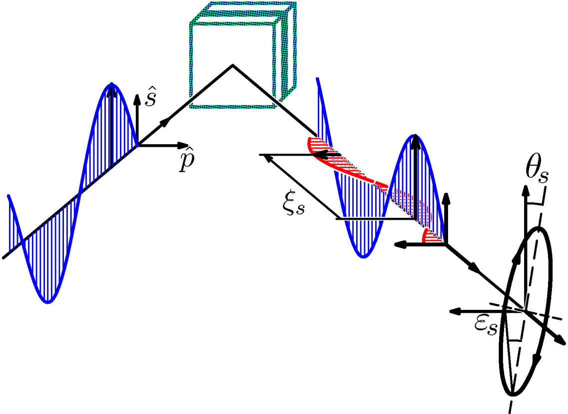
MO effect IV: Voigt effect:

- 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).



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 (microscopical):

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]

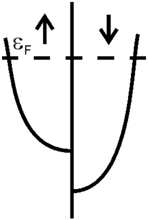
$$R = \begin{bmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{bmatrix}$$

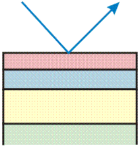
$\Downarrow$

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.]

⇒ Linear MOKE: PMOKE, LMOKE, TMOKE, MCD, MCB, [Hall]

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

⇒ Quadratic MOKE:

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

⇒ 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:

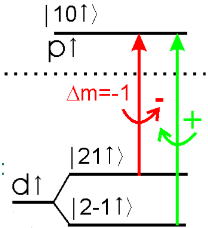
Electric dipole transition is allowed when following conditions are fulfilled:

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.

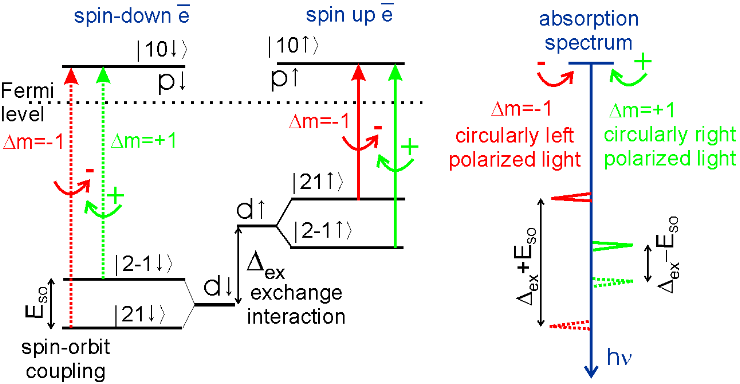
$$\Im[\varepsilon_{xx}] \sim \sum_{i,f}(f(E_i)-f(E_f))\times[|\langle i|p_+|f\rangle|^2+|\langle i|p_-|f\rangle|^2]\times\delta(E_f-E_i-\hbar\omega)$$

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

where

- $\langle i|$, $|f\rangle$: initial and final states, respectively.
- $p_\pm = p_x \pm ip_y$, $p_x = i\hbar\partial/\partial x$, 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.

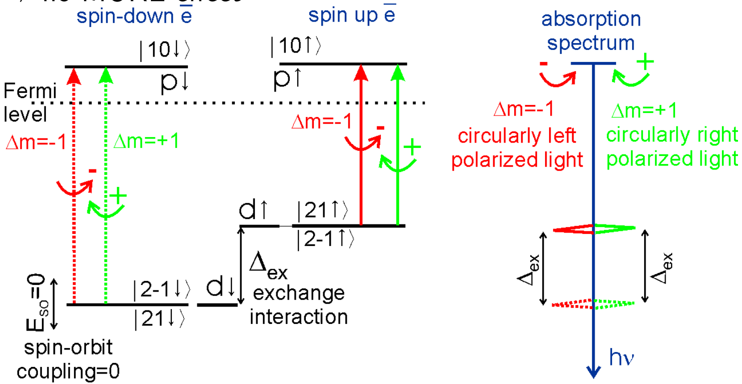
Magneto-optical spectroscopy microscopic picture



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

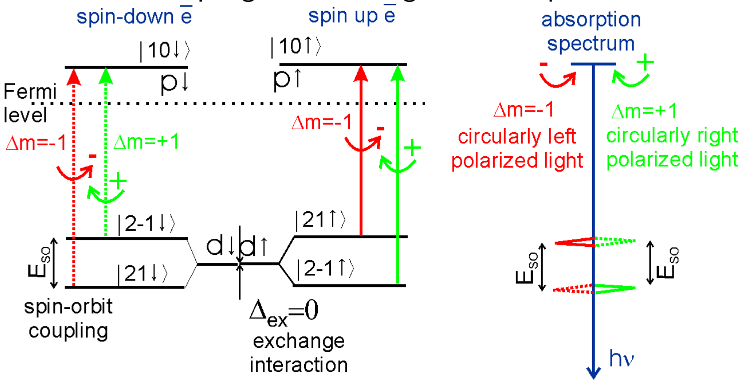
No spin-orbit coupling assumed:

⇒ no MOKE effect

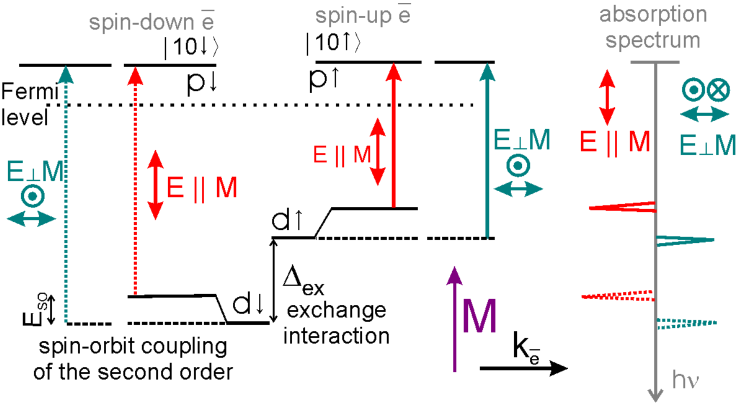


No exchange assumed:

- ⇒ no MOKE effect
- ⇒ both SO coupling and exchange must be present to have MOKE.

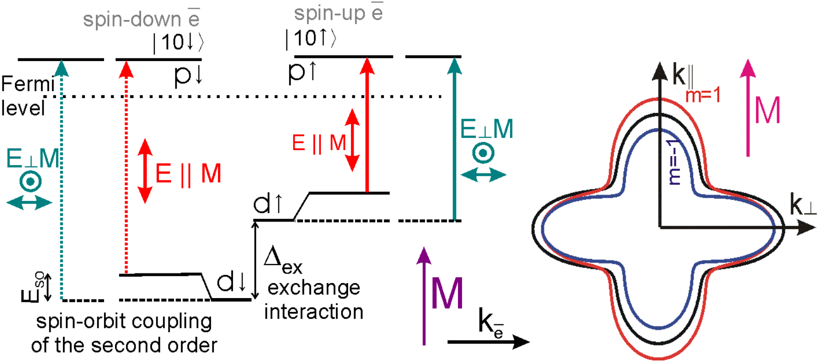


Quadratic Magneto-optical Kerr effect (QMOKE):



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

MOKE advantages and disadvantages:

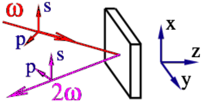
- spatial resolution limited by wavelength limit (∼300nm for visible light) → but sub-wavelength resolution demonstrated.
- investigation 'on distance', light can be transported nearby sample by a fibre.
- no need of vacuum or special sample preparation.
- depth resolution about 30nm.
- measurements 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.

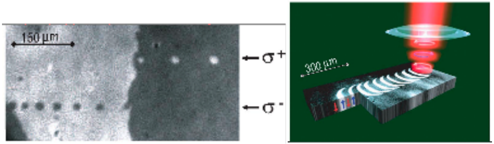
Extensions of MOKE:

- XMCD, XMLD for high photon energy.
- Non-linear magneto-optics
  $\Rightarrow$ MO second harmonic generation.



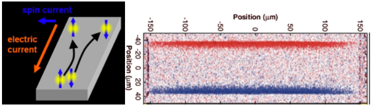
- Inverse Faraday effect (ultrafast optical switching).

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



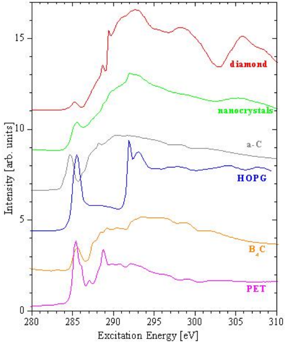
- Observation of spin accumulation in GaAs (spin Hall effect).

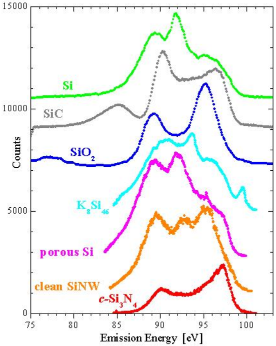
(Kato et al, Science, 2004)



X-ray absorption spectroscopy (XAS):

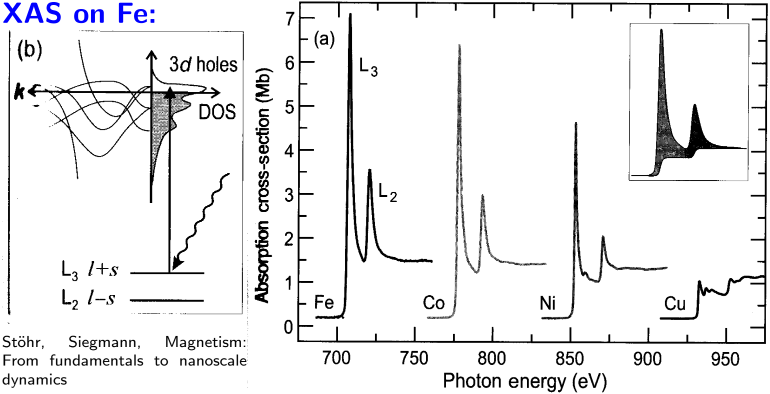
XAS is extremely sensitive to the chemical state each element, as each element have its own characteristic binding energies. XAS measurements can distinguish the form in which the element crystallizes (for example one can distinguish diamond and graphite, which both entirely consist of C), and can also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





http://beamteam.usask.ca/

XAS on Fe:



Stöhr, Siegmann, Magnetism: From fundamentals to nanoscale dynamics

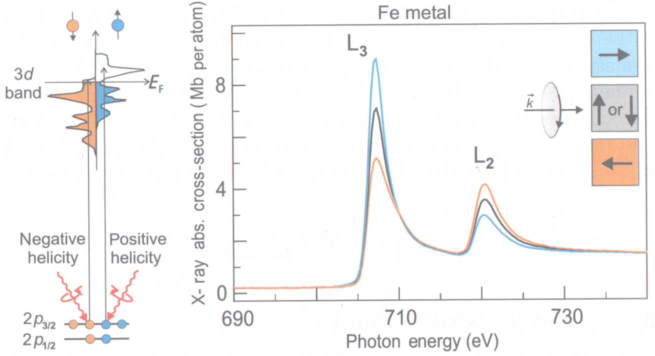
Starting L2, L3 edge (i.e. $2p^{1/2}$, $2p^{3/2}$, respectively):

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

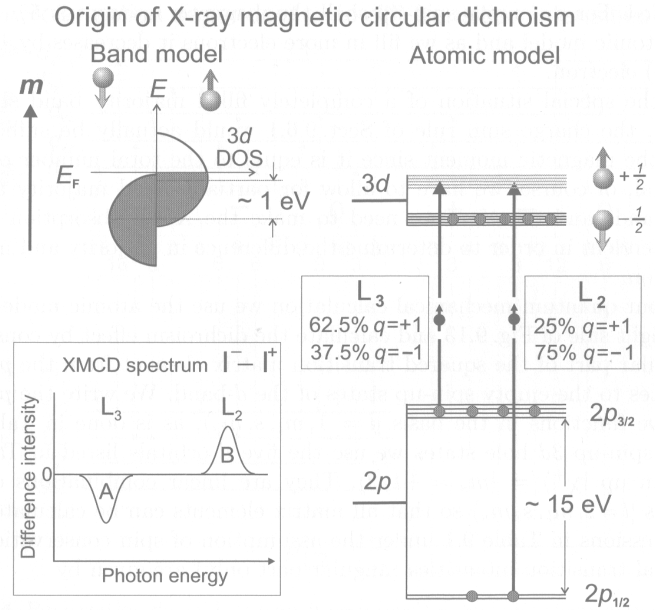
N_h: number of free d-states. $p \rightarrow s$ has small contribution.

XMCD: X-ray Magnetic circular dichroism:

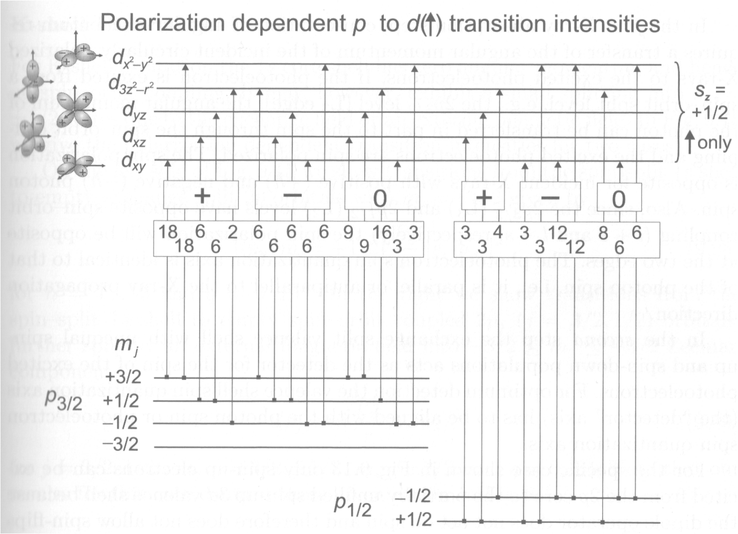
Circular Dichroism: different absorption for circularly left and right light polarization.



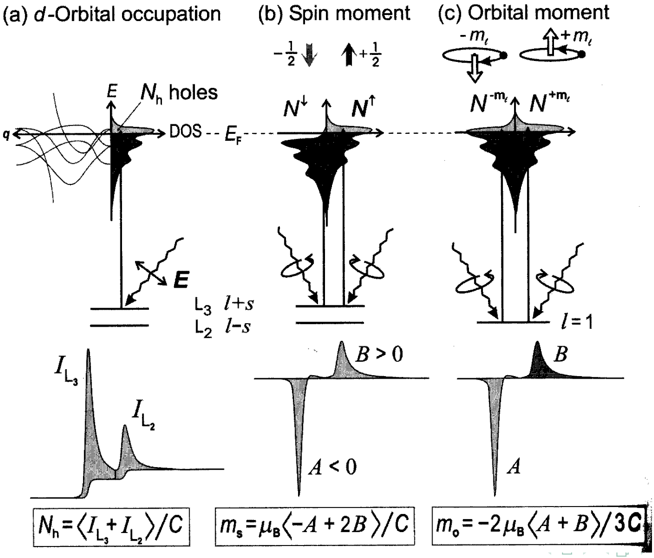
Different absorbed intensity for opposite magnetization orientations.


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⁺
Difference intensity
L3
L2
0
A
B
Photon energy
2p
2p3/2
~ 15 eV
2p1/2

XMCD: Detailes p → 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 nm).
- can provide excellent time resolution ($\sim$ 100 fs).

Disadvantages:

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

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