Light-Matter Interactions

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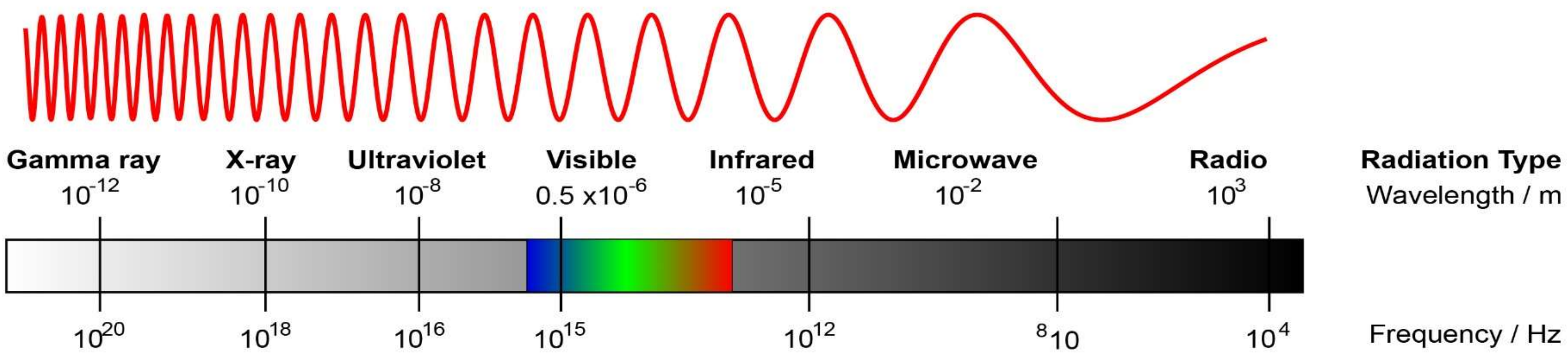
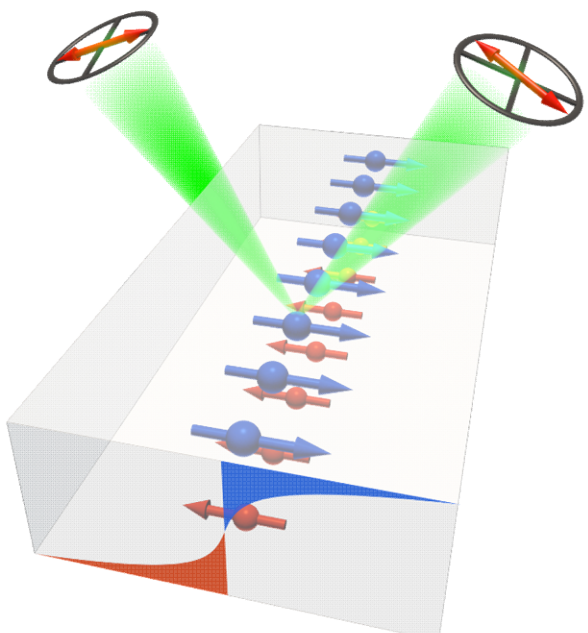
Outline – Lecture I – General Overview

- Introduction – *phenomenology*
- Electronic information vs. structural information
    - Electronic structure picture of materials
- Theory/understanding of light-matter interactions
    - The classical fields' description
    - Quantum theory with classical fields
    - Complete quantum field theory

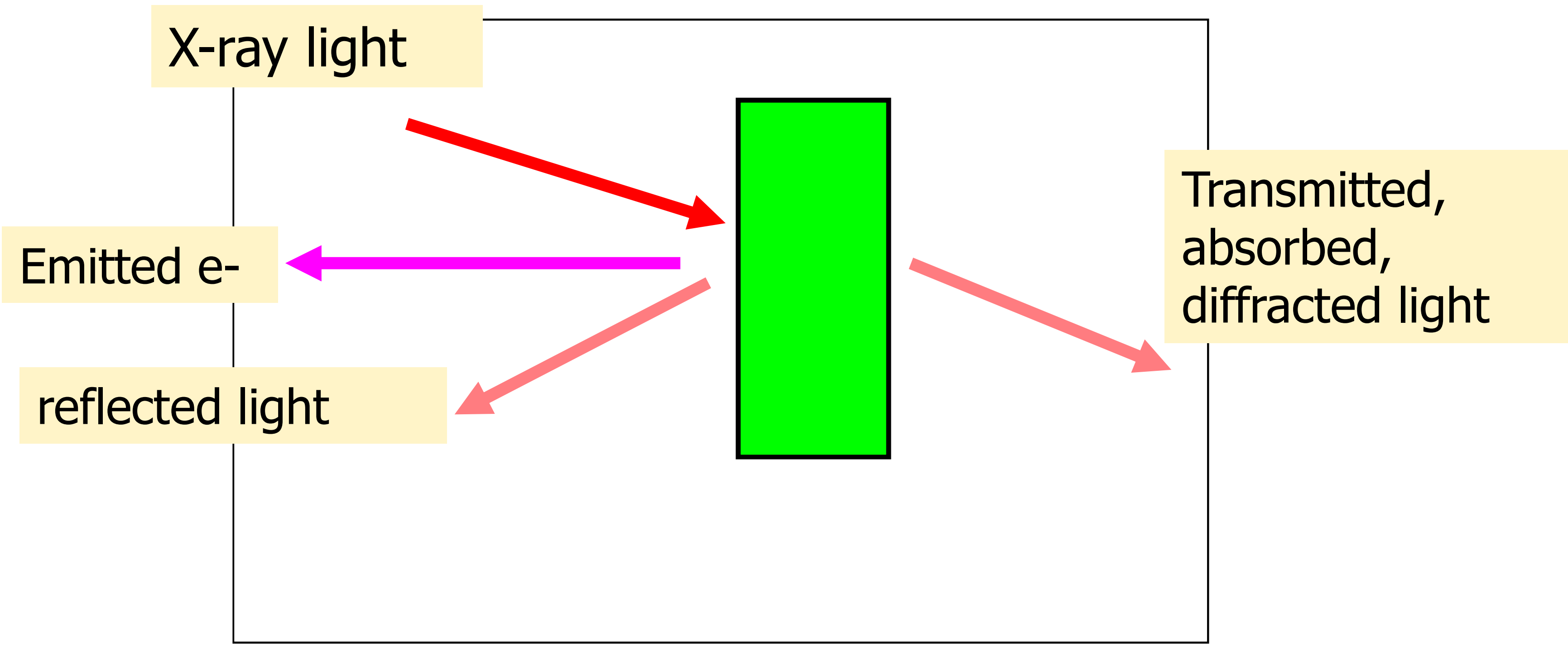


Outline – Lecture II – Light-magnetism Interaction

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



Phenomenology – Types of light-matter interactions



Gain information on two main information areas:
electronic & *magnetic* structure and *structural* information

(with many subdivisions each)

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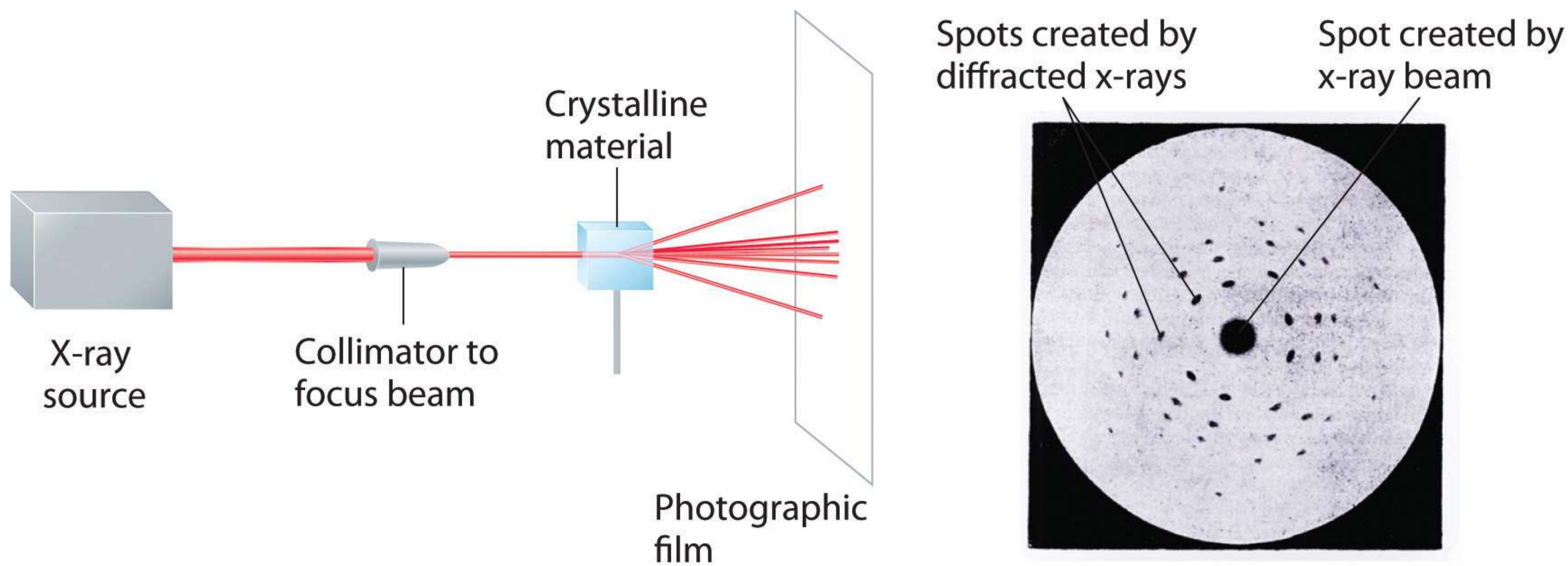
Pure structural information

X-ray diffraction → Positions (crystallography, biomolecules, phonons, etc)

$$I(\boldsymbol{q}) \propto |F(\boldsymbol{q})|^2 = \left|\sum_j f_j \exp(2\pi i(\boldsymbol{q}\cdot\boldsymbol{u}_j))\right|^2$$



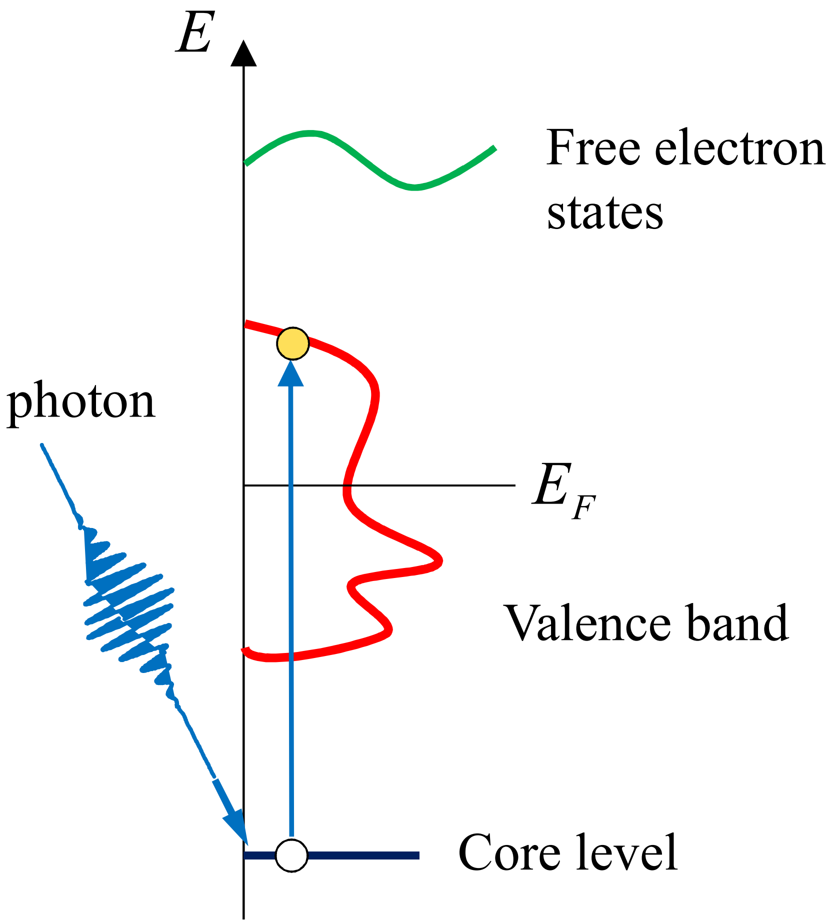
(a) X-ray diffraction

(b) X-ray diffraction pattern captured on photographic film

Often very hard x-rays !

Electronic structure information

Excitation by photon of one electronic state to another one, provides information on the materials' *electronic structure*

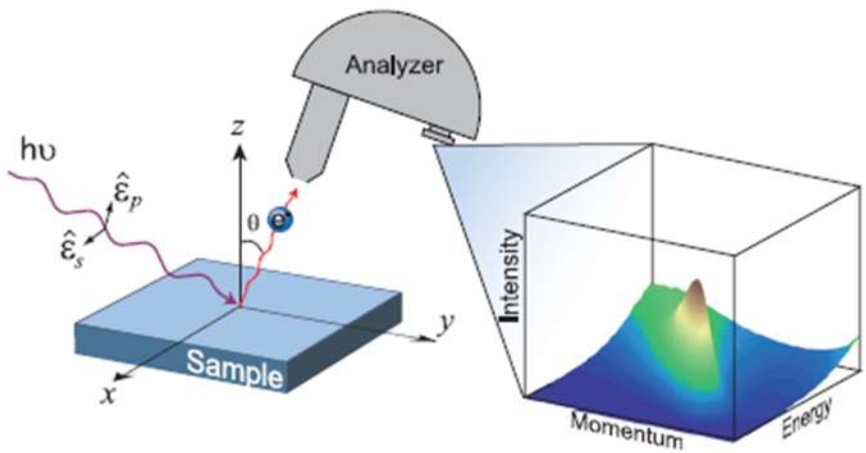


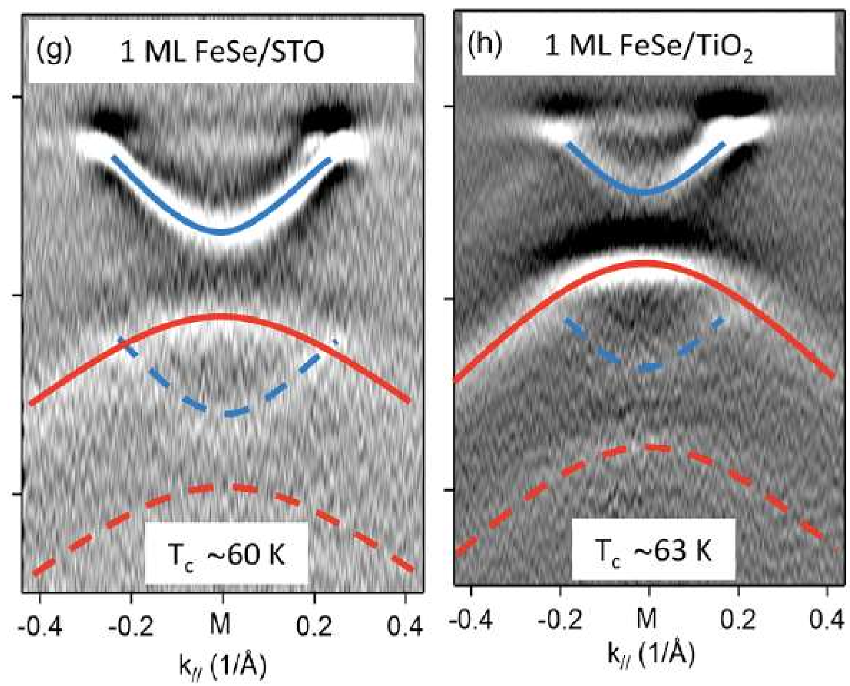
Can measure in
photon-in / photon-out
set-up, *or*
photon-in / electron-out

Information on binding energies, unoccupied states, spin- and orbital properties, electron distributions, quasi-particles etc.

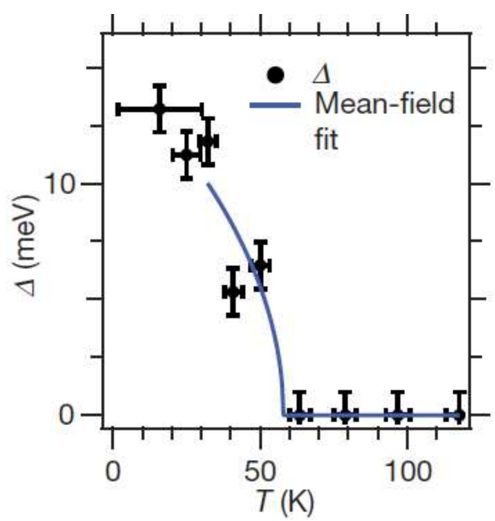
Detailed electronic structure information

High-resolution Angular Resolved Photoemission Spectroscopy (ARPES)





Observation of shadow bands FeSe/STO
Rebec et al, PRL **118**, 067002 (2017)

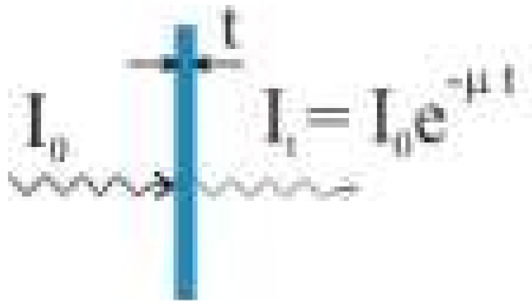


Superconducting gap FeSe/$SrTiO_3$

Lee et al, Nature **515**, 245 (2014)

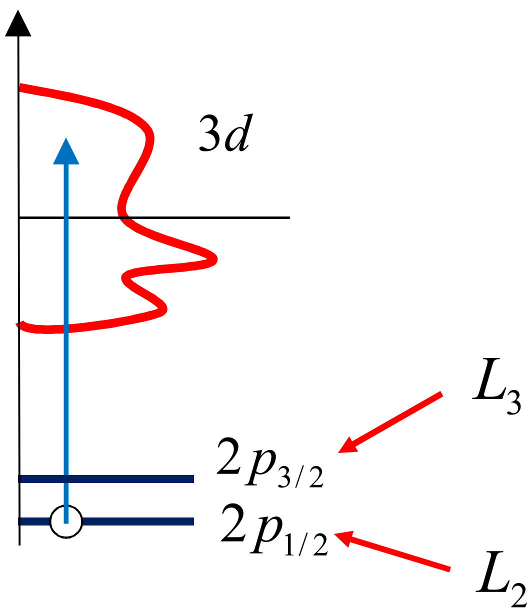
Further example: X-ray absorption

Transmission

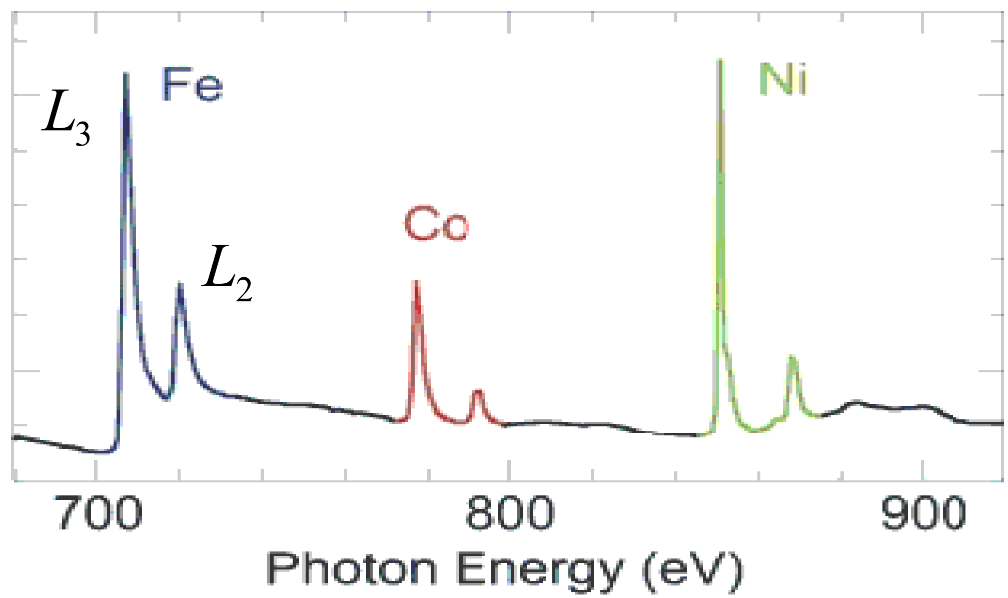


Sample

Beer-Lambert law



Absorption coefficient μ

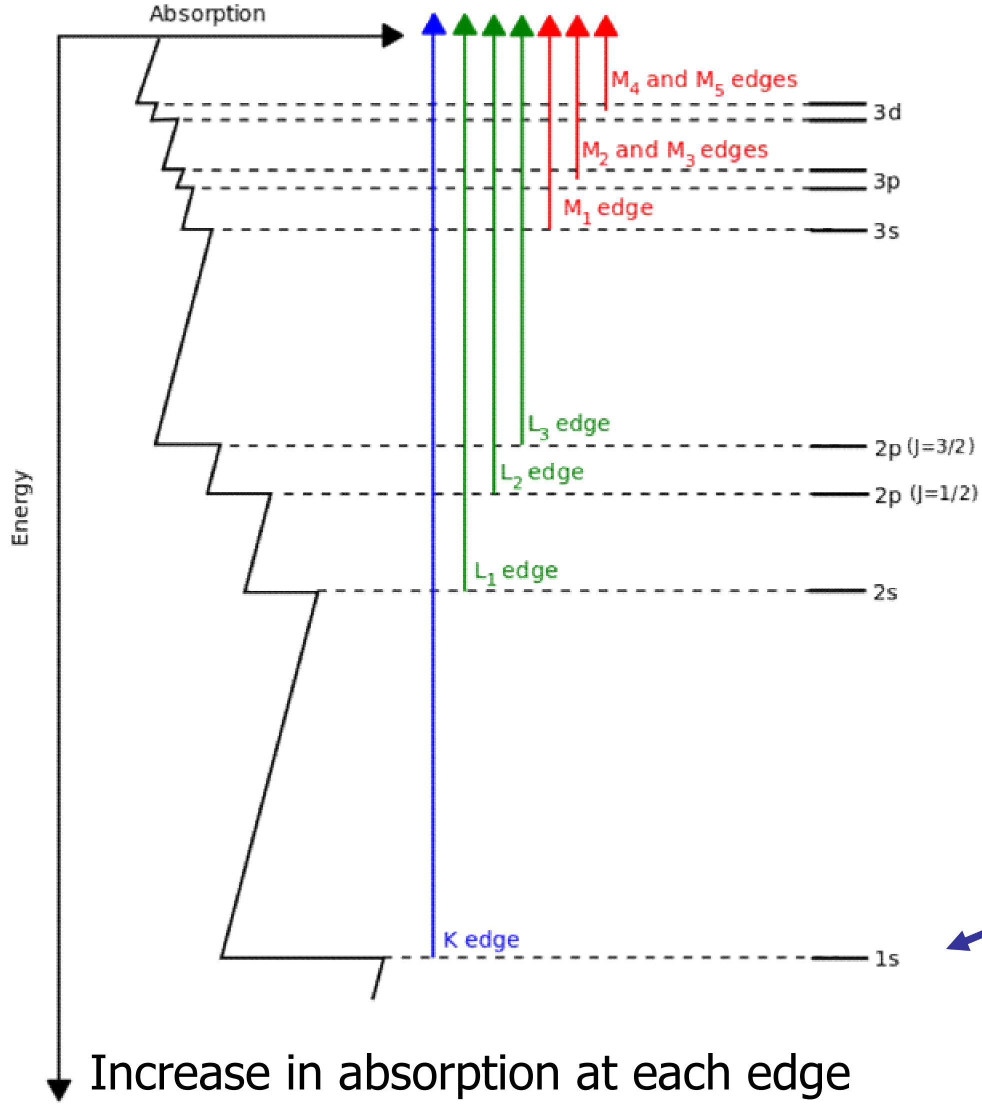


Detailed understanding

Resonant excitation of dipole allowed transitions at edges

$$2p_{1/2};\ 2p_{3/2} \Rightarrow\ 3d$$

Core-level absorption edges



Increase in absorption at each edge

Basic electronic structure:

Positions of the core levels
(here of a 4p element)

Spin-orbit split states ~16 eV
(3d element)

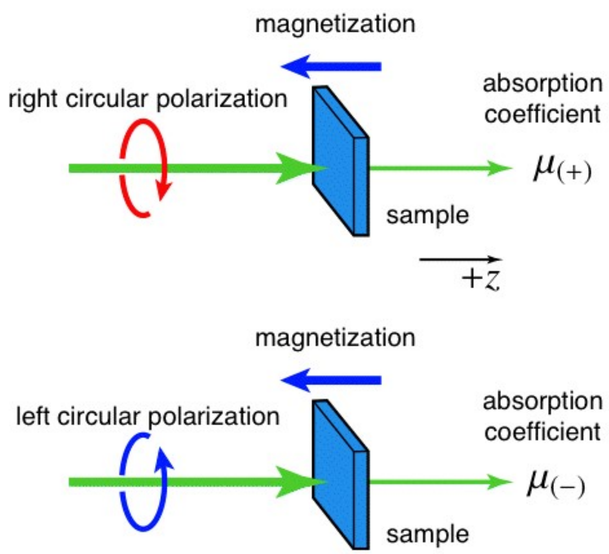
SO splitting of core states:

$$j = \ell \pm s = \ell \pm 1/2 \qquad \ell \geq 1$$

$$j = s = 1/2 \qquad \ell = 0$$

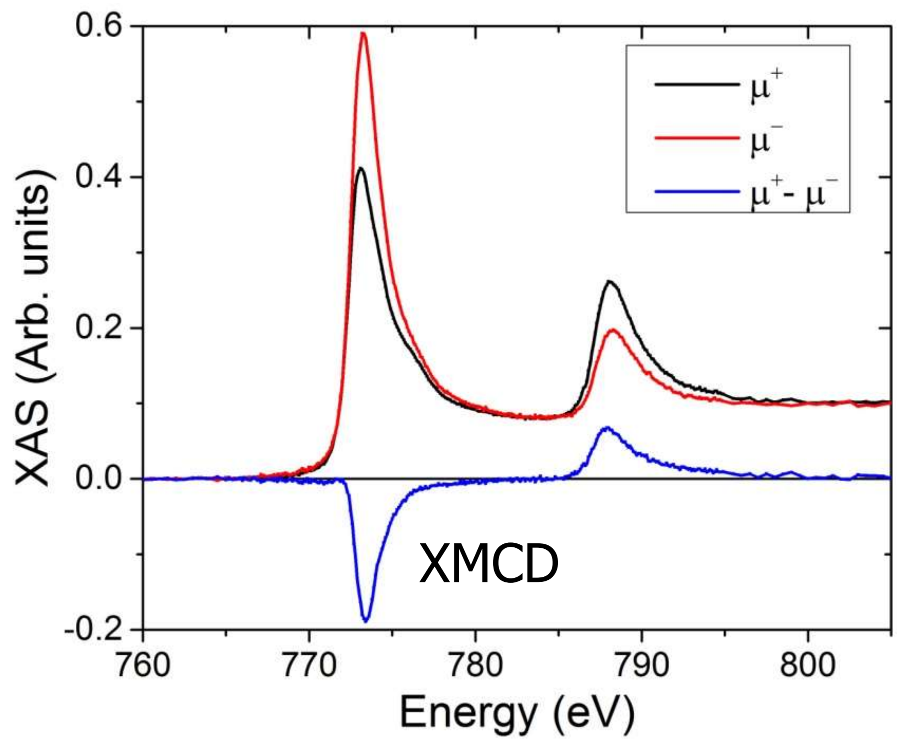
Magnetic information - XAS of ferromagnetic materials

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



X-ray magnetic circular dichroism (XMCD)

$\Delta\mu = \mu_{(+)} - \mu_{(-)}$



Provides a powerful tool to measure element-selectively the atomic magnetic moment

Fundamentals of Light – Matter Theory

Theory/understanding of light-matter interactions – 3 levels

- The classical fields' description
- Quantum theory with classical fields
- Complete quantum field theory

First level:

Maxwell theory and Fresnel theory (classical fields), macroscopic materials' quantities (no quantum physics)

Second level:

Maxwell theory and Fresnel theory (classical fields), materials' quantities given by quantum theory for materials

Third level:

Quantized photon fields, coupled to quantum theory for materials (i.e., 2nd quantization of photon fields)

First level: Maxwell-Fresnel theory

To describe the interaction between matter and the E-M wave field there are several ingredients:

(1) eigenwaves in vacuum & material and (2) the boundary conditions

Both (1) & (2) follow from the Maxwell equations:

$$\begin{aligned}
\nabla \cdot \boldsymbol{D} &= 4\pi\rho\,, \\
\nabla \cdot \boldsymbol{B} &= 0\,, \\
\nabla \times \boldsymbol{H} &= \frac{4\pi}{c}\boldsymbol{j} + \frac{1}{c}\frac{\partial \boldsymbol{D}}{\partial t}\,, \\
\nabla \times \boldsymbol{E} &= -\frac{1}{c}\frac{\partial \boldsymbol{B}}{\partial t}\,,
\end{aligned}$$

(in CKS units!)

Materials equations are also needed:

$$\begin{aligned}
\boldsymbol{D} &= \boldsymbol{\epsilon} \cdot \boldsymbol{E} \\
\boldsymbol{j} &= \boldsymbol{\sigma} \cdot \boldsymbol{E} \\
\boldsymbol{B} &= \boldsymbol{\mu} \cdot \boldsymbol{H}
\end{aligned}$$

D : displacement field
E : electrical field
B : magnetic induction
H : magnetic field

j : current density
ρ : charge density

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Materials relations

Just as important are the *materials relationships* :

$$D = \epsilon \cdot E = E + 4\pi P \,,$$

$$B = \mu \cdot H = H + 4\pi M \,,$$

$$j = \sigma \cdot E$$

With the material specific(!) tensors:

ε : permittivity tensor

μ : permeability tensor

σ : conductivity tensor

And: ***P*** : electrical polarization

M : magnetization

Note: we use here $\varepsilon_0=1$, $\mu_0=1$

Note: materials fields are not uniquely defined.

These equations are valid for *constant* ε, μ, and σ. This is usually not the case!

A closer look at the materials relationships

If we don´t have constant material´s tensors, things become nastier when we consider the full dependence on the space and time coordinates:

$$\boldsymbol{D}(\boldsymbol{r},t) = \int d\boldsymbol{r}' \int dt'\, \boldsymbol{\epsilon}(\boldsymbol{r}-\boldsymbol{r}';t-t') \cdot \boldsymbol{E}(\boldsymbol{r}',t')$$

$$\boldsymbol{E}(\boldsymbol{r},t) = \frac{1}{(2\pi)^4} \int d\boldsymbol{k} \int d\omega\, \boldsymbol{E}(\boldsymbol{k},\omega)\, \mathrm{e}^{i\boldsymbol{k}\cdot\boldsymbol{r}-i\omega t}$$

(homogeneous approximation!)

But, going to reciprocal space makes life easy again !

$$\left\{ \begin{array}{l} \boldsymbol{D}(\boldsymbol{k},\omega) = \boldsymbol{\epsilon}(\boldsymbol{k},\omega) \cdot \boldsymbol{E}(\boldsymbol{k},\omega) \\ \boldsymbol{j}(\boldsymbol{k},\omega) = \boldsymbol{\sigma}(\boldsymbol{k},\omega) \cdot \boldsymbol{E}(\boldsymbol{k},\omega) \end{array} \right.$$

With the material specific(!) tensors:

ε : permittivity tensor

μ : permeability tensor

σ : conductivity tensor

And: $\boldsymbol{D} = \boldsymbol{\epsilon} \cdot \boldsymbol{E} = \boldsymbol{E} + 4\pi \boldsymbol{P}$

$$j_{ind.} = \partial P / \partial t \quad \Rightarrow \quad \boldsymbol{\epsilon}(\omega) = \mathbf{1} + \frac{4\pi i}{\omega} \boldsymbol{\sigma}(\omega)$$

Consequences of Maxwell equations

Solutions of the M.E. for isotropic medium: transverse plane E-M waves:

Re-write the equations (vacuum, $\boldsymbol{\varepsilon} = \varepsilon_0$, $\boldsymbol{\mu} = \mu_0$)

$$\boldsymbol{\nabla} \times (\boldsymbol{\nabla} \times \boldsymbol{E}) = \boldsymbol{\nabla} \times (-\frac{\partial \boldsymbol{B}}{\partial t}) = -\mu_0\varepsilon_0 \frac{\partial^2 \boldsymbol{E}}{\partial t^2}$$

$$\Downarrow$$

$$\boldsymbol{\nabla}(\boldsymbol{\nabla} \cdot \boldsymbol{E}) - \nabla^2 \boldsymbol{E} \Longrightarrow \boxed{\nabla^2 \boldsymbol{E} = \mu_0\varepsilon_0 \frac{\partial^2 \boldsymbol{E}}{\partial t^2}}$$

And:

$$\boldsymbol{\nabla} \times (\boldsymbol{\nabla} \times \boldsymbol{B}) = \mu_0\varepsilon_0 \frac{\partial}{\partial t}(\boldsymbol{\nabla} \times \boldsymbol{E}) = -\mu_0\varepsilon_0 \frac{\partial^2 \boldsymbol{B}}{\partial t^2}$$

$$\Downarrow$$

$$\boldsymbol{\nabla}(\boldsymbol{\nabla} \cdot \boldsymbol{B}) - \nabla^2 \boldsymbol{B} \Longrightarrow \boxed{\nabla^2 \boldsymbol{B} = \mu_0\varepsilon_0 \frac{\partial^2 \boldsymbol{B}}{\partial t^2}}$$

Solutions: E-M wave

$$\begin{cases} \boldsymbol{E}(\boldsymbol{r}, t) = \boldsymbol{E}_0 \; e^{i(\boldsymbol{k}\cdot\boldsymbol{r} - \omega t)} \\ \boldsymbol{B}(\boldsymbol{r}, t) = \boldsymbol{B}_0 \; e^{i(\boldsymbol{k}\cdot\boldsymbol{r} - \omega t)} \end{cases}$$

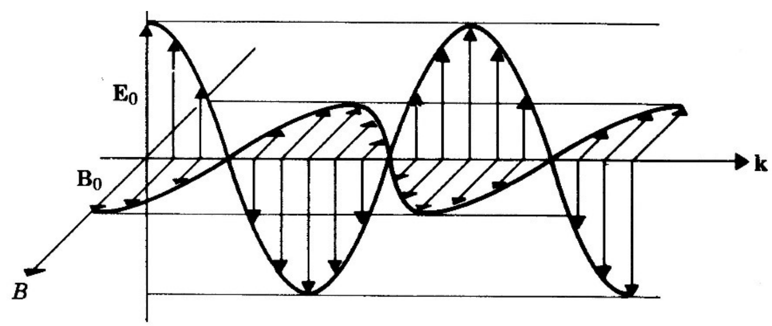
Or: $\boldsymbol{E} \sim \sin(\boldsymbol{k} \cdot \boldsymbol{r} - \omega t)$, $\cos(\boldsymbol{k} \cdot \boldsymbol{r} - \omega t)$

Also:

$$\boldsymbol{\nabla} \cdot \boldsymbol{E} = 0 \Longrightarrow \boldsymbol{k} \cdot \boldsymbol{E} = 0 \Longrightarrow \boldsymbol{k} \perp \boldsymbol{E}$$

$$\boldsymbol{\nabla} \cdot \boldsymbol{B} = 0 \Longrightarrow \boldsymbol{k} \cdot \boldsymbol{B} = 0 \Longrightarrow \boldsymbol{k} \perp \boldsymbol{B}$$

$$\boldsymbol{\nabla} \times \boldsymbol{E} = -\frac{\partial \boldsymbol{B}}{\partial t} \Longrightarrow \boldsymbol{k} \times \boldsymbol{E} = \omega \boldsymbol{B} \Longrightarrow \boldsymbol{E} \perp \boldsymbol{B}$$



Light is a transverse E-M wave

Index of refraction

The plane-wave solution is possible under the condition:

$$(k \cdot k)\vec{E} = \frac{\varepsilon\mu}{c^2}\omega^2\vec{E}, \quad (k \cdot k)\vec{B} = \frac{\varepsilon\mu}{c^2}\omega^2\vec{B} \longrightarrow k^2 = \frac{\varepsilon\mu}{c^2}\omega^2$$

Dispersion relation

Index of refraction: $\vec{n} = \frac{c\vec{k}}{\omega} \Rightarrow n = \sqrt{\varepsilon\mu}\,, \qquad n = c/v$

<u>Remarks:</u>

1) For materials ε, μ are complex $\longrightarrow$ n is complex & vector

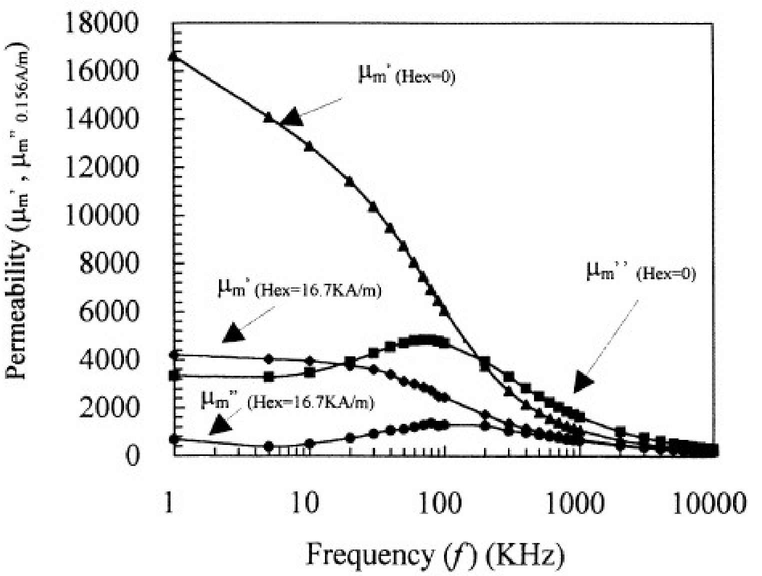
2) The "spins cannot follow the rapid moving H field" $\longrightarrow$ $\mu = 1$

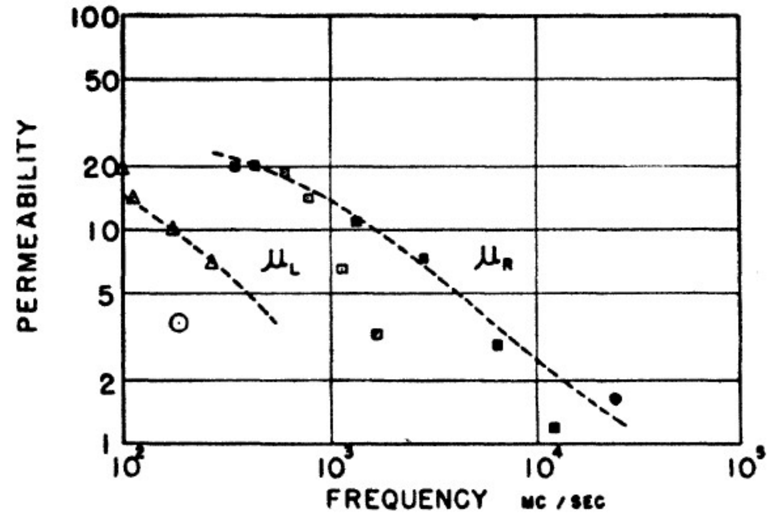
$$\longrightarrow \boxed{n(\omega) = \sqrt{\varepsilon\mu} = \sqrt{\varepsilon(\omega)}}$$

(Dispersion relation)

Nonetheless, all magnetic information is acounted for (see later)

Measured relative permeability $\mu(\omega)$





Ni

Kittel, Phys. Rev. **70**, 281 (1946)

1 eV = 0.25 10^{15} Hz

Arguments

1) no unique separation between *D* and *H* in the Maxwell equations
2) physically: „spins cannot follow the rapidly varying *B* field"

$\mu(\omega)$ => 1 at optical frequencies, $\boldsymbol{B}(\omega) = \boldsymbol{H}(\omega) \quad (k = 2\pi/\lambda \rightarrow 0)$

Energy dispersion of optical constants

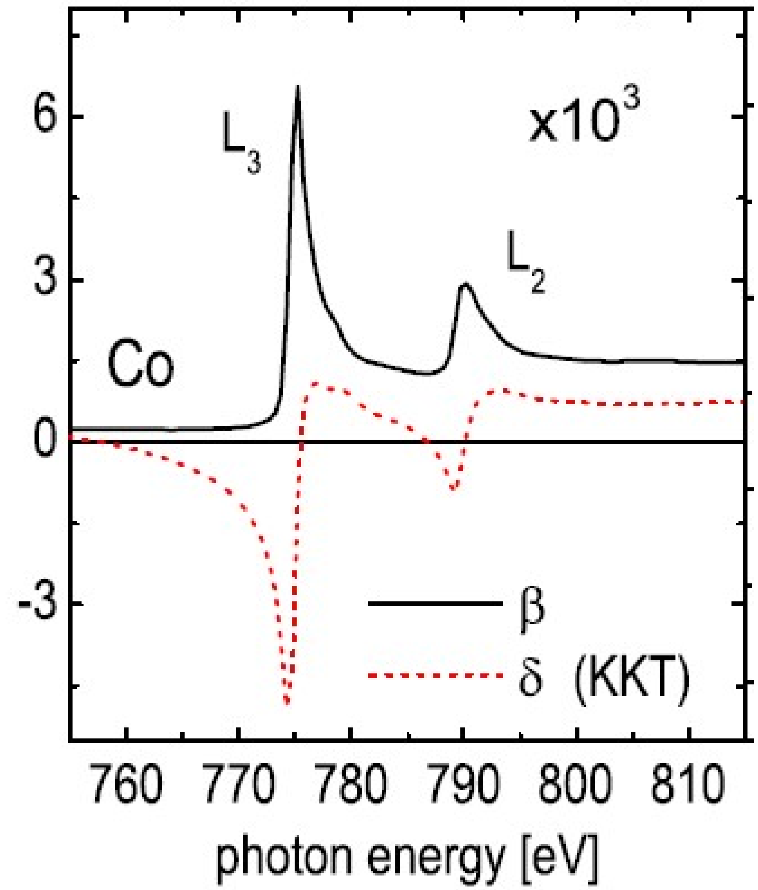
In the x-ray regime, *n* is close to one and complex:

$$n(\omega) = 1 - \delta(\omega) + i\beta(\omega)$$

δ can be positive or negative !

Also, δ and β do depent on the magnetization !

Eventhough δ and β are small they can be measured accurately at modern synchrotrons



Fresnel equation for the material

A combination of the M-E leads to the following wave equation *in the material* :

$$\boxed{-\nabla^2 \boldsymbol{E} + \nabla(\nabla \cdot \boldsymbol{E}) = -\frac{1}{c^2}\frac{\partial^2 \boldsymbol{D}}{\partial t^2}}$$

This is similar to the equation for the isotropic, constant ε case

Substitute:

$$\boldsymbol{E}(\boldsymbol{r}, t) = \boldsymbol{E}_0 \, \mathrm{e}^{i\omega \boldsymbol{n}\cdot\boldsymbol{r}/c - i\omega t}$$

Gives us the Fresnel equation:

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

The solution gives 2 ***n*** in the material and the eigen modes E_0

$$(n:n)_{ij} = n_i n_j$$

Note: we used μ=1

Written in full (SI), it would be:

$$\boxed{\left(n^2 - \frac{\mu\varepsilon}{\mu_0\varepsilon_0} - \boldsymbol{n} : \boldsymbol{n}\right) \boldsymbol{E} = 0}$$

Fresnel equation, continued

The symmetry of ε tensor is an important ingredient for solving the Fresnel equation.

In short, one needs to know about the crystallographic and magnetic symmetry of the material !

Some examples for non-magnetic materials:

Cubic:

(1 quantity)

$$\overleftrightarrow{\varepsilon} = \begin{pmatrix} \varepsilon_{xx} & & \\ & \varepsilon_{xx} & \\ & & \varepsilon_{xx} \end{pmatrix}$$

Monoclinic:

(4 quantities)

$$\overleftrightarrow{\varepsilon} = \begin{pmatrix} \varepsilon_{xx} & \varepsilon_{xy} & \\ \varepsilon_{xy} & \varepsilon_{yy} & \\ & & \varepsilon_{zz} \end{pmatrix}$$

(biaxial)

Tetragonal
hexagonal
trigonal

(2 quantities)

$$\overleftrightarrow{\varepsilon} = \begin{pmatrix} \varepsilon_{xx} & & \\ & \varepsilon_{xx} & \\ & & \varepsilon_{zz} \end{pmatrix}$$

(uniaxial)

Triclinic:

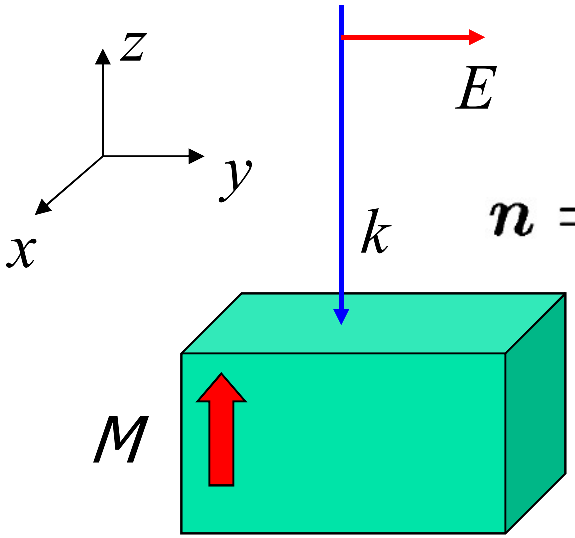
(6 quantities)

$$\overleftrightarrow{\varepsilon} = \begin{pmatrix} \varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} \\ \varepsilon_{xy} & \varepsilon_{yy} & \varepsilon_{yz} \\ \varepsilon_{xz} & \varepsilon_{yz} & \varepsilon_{zz} \end{pmatrix}$$

(biaxial)

Example of Fresnel equation for magnetic medium

Magnetic medium, M||z:



$$\boldsymbol{n} = n\boldsymbol{e}_z$$

Dielectric tensor:

$$\boldsymbol{\epsilon} = \begin{pmatrix} \epsilon_{xx} & \epsilon_{xy} & 0 \\ -\epsilon_{xy} & \epsilon_{xx} & 0 \\ 0 & 0 & \epsilon_{zz} \end{pmatrix}$$

Why? Consequence of magnetism!

Look at σ tensor: $\boldsymbol{\epsilon}(\omega) = \mathbf{1} + \frac{4\pi i}{\omega}\boldsymbol{\sigma}(\omega)$

$\varepsilon_{xy} = \frac{4\pi i}{\omega}\sigma_{xy}$, $\sigma_{xy} \neq 0$ because of the magnetism!

Hall current, σ_{xy}

(SI units: $\epsilon(\omega) = \epsilon_0 \mathbf{I} + \frac{i\sigma(\omega)}{\omega}$)

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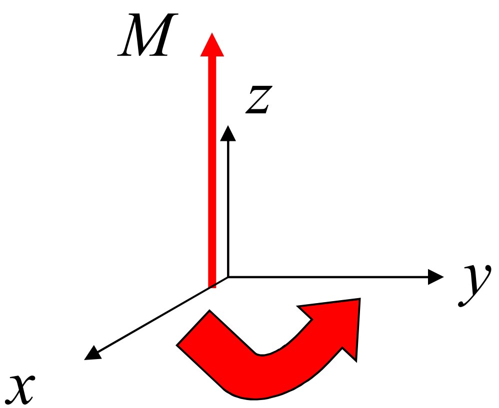
Examples magnetic Fresnel equation, continued



Hall current σ_{xy}

Note: $\sigma_{xy} = -\sigma_{yx}$

Thus: $\varepsilon_{yx} = -\varepsilon_{xy}$

Solve Fresnel equation:

$$\begin{vmatrix} n^2-\varepsilon_{xx} & -\varepsilon_{xy} & \\ +\varepsilon_{xy} & n^2-\varepsilon_{xx} & \\ & & n^2-\varepsilon_{zz}-n^2 \end{vmatrix} = 0$$

$$\left\{ \begin{aligned} & -\varepsilon_{zz}[(n^2-\varepsilon_{xx})^2+\varepsilon_{xy}^2]=0 \\ & \Rightarrow (n^2-\varepsilon_{xx})^2 = -\varepsilon_{xy}^2 \\ & \Rightarrow n^2 = \varepsilon_{xx} \pm i\varepsilon_{xy} \end{aligned} \right.$$

There are two solutions:

$$\boxed{n_{1,2}^2 \equiv n_{\pm}^2 = \varepsilon_{xx} \pm i\varepsilon_{xy}}$$

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Fresnel equation, magnetic case

Eigenmodes:

$$\begin{bmatrix} \pm i\varepsilon_{xy} & -\varepsilon_{xy} & \\ +\varepsilon_{xy} & \pm i\varepsilon_{xy} & \\ & & -\varepsilon_{zz} \end{bmatrix} \bullet \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} = 0 \Rightarrow \begin{bmatrix} \pm i & -1 \\ +1 & \pm i \end{bmatrix} \bullet \begin{pmatrix} E_x \\ E_y \end{pmatrix} = 0 \;\&\; E_z = 0,$$

$$\Rightarrow \begin{pmatrix} E_x \\ E_y \end{pmatrix}_{\pm} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ \pm i \end{pmatrix}$$

(normalized eigenmodes!)

Solutions are circularly polarized waves (in the material):

$$\vec{E}_{\pm}(r,t) = \frac{1}{\sqrt{2}}(\vec{e}_x \pm i\vec{e}_y)\cdot e^{i\omega/c(n_{\pm}\cdot r) - i\omega t}$$

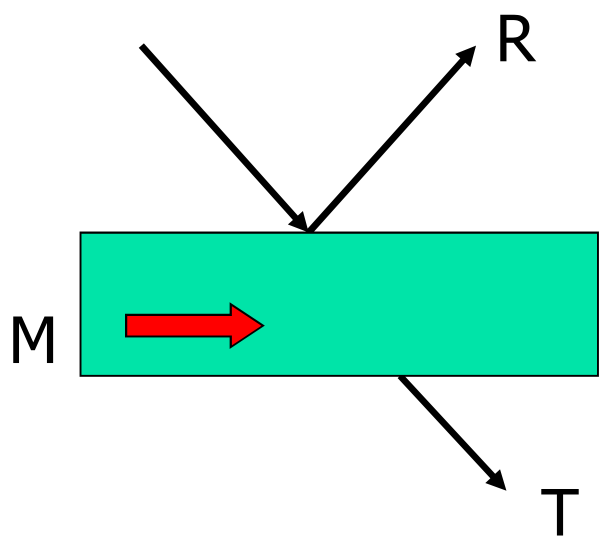
One circularly polarized wave with helicity + corresponds to n_+,
the other one with helicity - to n_-

$\boxed{\text{This situation is called "magnetic circular dichroism", i.e. 2 colors}}$

$$n_{\pm} = \frac{2\pi c}{\omega\lambda_{\pm}}$$

(will apply this to XAS/XMCD in Lecture II)

Materials´ boundary conditions



Experiments always require at least two different media

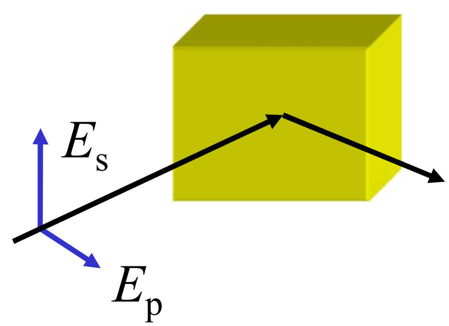
Next to the Fresnel equation (1) we must also know the „matching" conditions (2) at the boundaries !

These will follow (again) from the Maxwell equations

Continuity of temporal and spacial wave parts at interface → 1) Snell´s law
2) reflection/transmission coefficients

Convenient: Jones vector formulation:

2-dim. vector $\vec{E} = \begin{pmatrix} E_s \\ E_p \end{pmatrix}$



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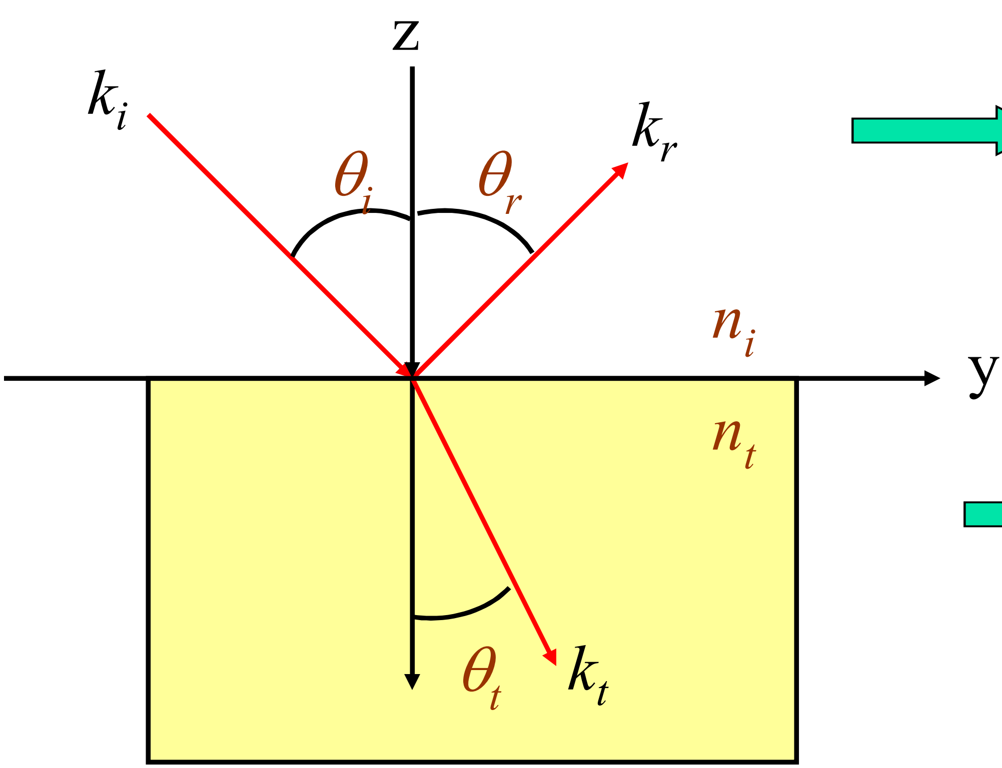
Refresher: Snell´s law

Continuity of temporal and spacial wave parts at interface $z=0$:

$$\Rightarrow \quad e^{ik_i\cdot r-i\omega t}\Big|_{z=0}=e^{ik_r\cdot r-i\omega t}\Big|_{z=0}=e^{ik_t\cdot r-i\omega t}\Big|_{z=0}$$

Use: $\vec{k}=\dfrac{n\omega}{c}\hat{k}$

$$\omega_i=\omega_r=\omega_t=\omega$$



$$\Rightarrow \quad \frac{n_i\omega}{c}(\hat{k}_y^i y)=\frac{n_i\omega}{c}(\hat{k}_y^r y)=\frac{n_t\omega}{c}(\hat{k}_y^t y)$$

$$\hat{k}_y^i=\sin\theta_i\,,\quad \hat{k}_y^r=\sin\theta_r\,,\quad \hat{k}_y^t=\sin\theta_t$$

$$\Rightarrow \quad \begin{cases} \sin\theta_i=\sin\theta_r \\ n_i\sin\theta_i=n_t\sin\theta_t \end{cases}$$

Snell´s law

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Reflection/transmission coefficients, Jones formulation

Definition of reflection matrix:

(similar for transmission)

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

Here r_{sp} means: p-polarized light in, reflected as s-pol. light.
The r_{sp} are magnetic (Fresnel) reflection coefficients

The reflection coefficients follow from the Maxwell equations.

Example: s-polarized light, scalar dielectric constant ε

Calculation gives:

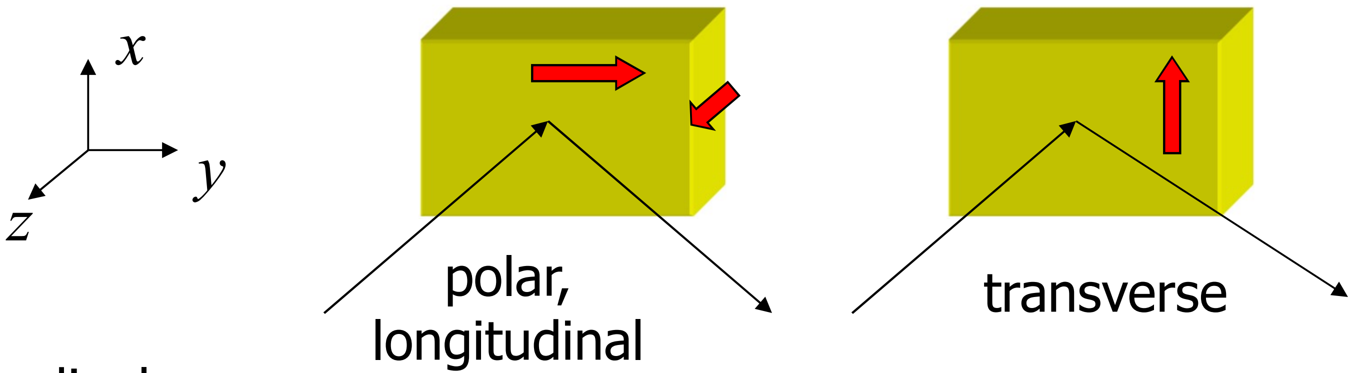
$$\begin{cases} \dfrac{E_s^r}{E_s^i} \equiv r_{ss} = \dfrac{n_i \cos\theta_i - n_t \cos\theta_t}{n_i \cos\theta_i + n_t \cos\theta_t} \\[2ex] \dfrac{E_s^t}{E_s^i} \equiv t_{ss} = \dfrac{2 n_i \cos\theta_i}{n_i \cos\theta_i + n_t \cos\theta_t} \end{cases}$$

Similarly for p-polarized light

Note: $r_{ps} = 0$ here ! (no magnetism!)

Magnetic Fresnel coefficients

The calculation of the Fresnel coefficients in the case of a magnetic material can be teadious!



Polar, longitudinal:

$$\boxed{r_{sp}, r_{ps} \neq 0, \quad \Delta r_{pp} = 0}$$

Transverse:

$$\boxed{r_{sp}, r_{ps} = 0, \quad \Delta r_{pp} \neq 0}$$

$$\overleftrightarrow{r} = \begin{pmatrix} r_{ss} & \boxed{r_{sp}} \\ \boxed{r_{ps}} & r_{pp} + \boxed{\Delta r_{pp}} \end{pmatrix}$$

(more in Lecture II)

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Example: Magnetic reflection coefficients

Result for polar magnetization: $n_{\pm}^2 = \epsilon_{\rm xx} \pm i\epsilon_{\rm xy} \cos\phi_{\rm t}^{\pm}$ (M || z-axis)

$$
\begin{aligned}
r_{ss} &= (n_0 \cos\phi_{\rm i} - \bar{n}\cos\phi_{\rm t}) \,/\, (n_0 \cos\phi_{\rm i} + \bar{n}\cos\phi_{\rm t}) \; , \\
r_{pp} &= (\bar{n}\cos\phi_{\rm i} - n_0 \cos\phi_{\rm t}) \,/\, (\bar{n}\cos\phi_{\rm i} + n_0 \cos\phi_{\rm t}) \; , \\
r_{ps} &= \frac{-i n_0 \,(n_+ - n_-)\cos\phi_{\rm i}}{(\bar{n}\cos\phi_{\rm t} + n_0\cos\phi_{\rm i})(\bar{n}\cos\phi_{\rm i} + n_0\cos\phi_{\rm t})\cos\phi_{\rm t}} \; , \\
r_{sp} &= r_{ps} \; .
\end{aligned}
$$

Result for longitudinal magnetization: Same, but: $r_{sp} \rightarrow -r_{ps}$

$$
\epsilon = \begin{pmatrix} \epsilon_{\rm xx} & 0 & \epsilon_{\rm xz} \\ 0 & \epsilon_{\rm yy} & 0 \\ -\epsilon_{\rm xz} & 0 & \epsilon_{\rm zz} \end{pmatrix} \equiv \begin{pmatrix} \epsilon_1 & 0 & \epsilon_2 \\ 0 & \epsilon_1' & 0 \\ -\epsilon_2 & 0 & \epsilon_1 \end{pmatrix} \Longrightarrow n_{\pm}^2 \approx \epsilon_1 \pm i\epsilon_2 \sin\phi_{\rm t}^{\pm}
$$

(M || y-axis)

See: P.M. Oppeneer, in Handbook of Magnetic Materials, Vol. 13 (2001)

Second level – Quantum theory & classical fields

Spectroscopic quantities can be related to the materials' specific dielectric tensor ε (equivalently, σ)

$$r_{ss} = \frac{n_i \cos\theta_i - n_t \cos\theta_t}{n_i \cos\theta_i + n_t \cos\theta_t} \Longleftrightarrow \left[n^2 \mathbf{1} - \boldsymbol{\epsilon} - \boldsymbol{n} : \boldsymbol{n}\right] \cdot \boldsymbol{E} = 0$$

$$\boldsymbol{\epsilon}(\omega) = \mathbf{1} + \frac{4\pi i}{\omega} \boldsymbol{\sigma}(\omega)$$

Combine quantum theory of the solid with classical (external) E.M. fields as given by Maxwell's equations:

$$\vec{B}(\vec{r},t) = \vec{\nabla} \times \vec{A}(\vec{r},t), \quad \vec{E}(\vec{r},t) = -\partial \vec{A}(\vec{r},t) \big/ \partial t$$

Coulomb gauge

Use electronic structure theory to describe/compute ε (or, σ)

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Electronic structure theory

Complex many-electron problem – many particle Schrödinger equation

$$\hat{H} = \hat{T}_e + \hat{V}_{e-e} + \hat{V}_{e-ion} + \hat{V}_{ion-ion}$$

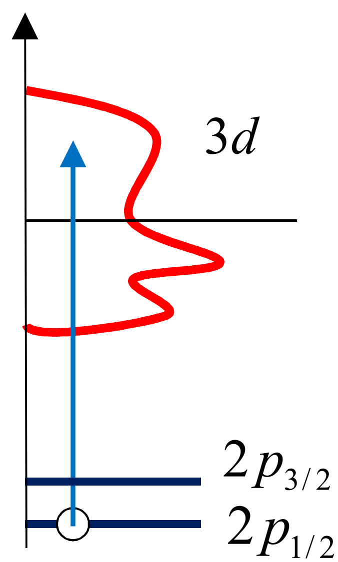
$$\hat{H}\Psi_{\nu_1 \ldots \nu_n}(r_1,\ldots,r_n;R_1,\ldots,R_N) = E_{\nu_1 \ldots \nu_n}\Psi_{\nu_1 \ldots \nu_n}(r_1,\ldots,r_n;R_1,\ldots,R_N)$$

$$\Psi_{\nu_1 \ldots \nu_n}(r_1,\ldots,\ r_n;R_1,\ldots,\ R_N)$$

Many-particle wave-function

Too difficult to solve!

- Want an effective, non-interacting single electron picture



Ab initio density-functional theory

Complex many-electron problem

exact!

Effective, non-interacting single electron problem

$$\hat{H}\Psi_{\nu_1 \ldots \nu_n}(r_1, \ldots, r_n; R_1, \ldots, R_N) =$$
$$E_{\nu_1 \ldots \nu_n}\Psi_{\nu_1 \ldots \nu_N}(r_1, \ldots, r_n; R_1, \ldots, R_N)$$

$$\hat{H} = \hat{T}_e + \hat{V}_{e-e} + \hat{V}_{e-ion} + \hat{V}_{ion-ion}$$

$$\left[-\frac{\hbar^2}{2m}\nabla^2 + V[n(r)] + V_{xc}[n(r)]\right]\psi_i(r) = E_i\psi_i(r)$$

$$n(r) = \sum_i |\psi_i(r)|^2$$

$\psi_i(r)$ selfconsistent solution

Kohn-Sham single electron equation,
Kohn-Sham density

Density functional theory (DFT):

1) The mapping is exact and provides a unique total energy functional $E[n]$; the exact ground state energy is obtained as its minimum for the ground state density n_G.

2) There is an (not exactly known) exchange-correlation energy $E_{xc}[n]$, which defines the exchange-correlation potential

$$V_{xc}[n(r)] = \delta E_{xc}[n]/\delta n(r)$$

Hohenberg-Kohn, Phys. Rev **136**, B864 (1964)

Kohn-Sham, Phys. Rev. **140**, A1133 (1965)

(See Lecture S. Blügel)

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Single particle, spin-density functional theory

Effective single-particle 2x2 potential (*with spin*):

$$V_{xc}(\vec{r}) = V_0(\vec{r})1 + \vec{B}_{xc}(\vec{r}) \cdot \hat{\vec{\sigma}}, \quad \vec{B}_{xc}(\vec{r}) = -\frac{\delta E_{xc}}{\delta \vec{m}(\vec{r})}$$

Exchange field

Effective Kohn-Sham Hamiltonian:

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

Spin-density (2x2):

$$n(\vec{r}) = \{ n_0(\vec{r})1 + \vec{m}(\vec{r}) \cdot \vec{\sigma} \} / 2$$

$$\begin{cases} n(\vec{r}) = n_\uparrow(\vec{r}) + n_\downarrow(\vec{r}) \\ m(\vec{r}) = \mu_B \{ n_\uparrow(\vec{r}) - n_\downarrow(\vec{r}) \} \end{cases}$$

Not yet fully relativistic; better is Kohn-Sham-Dirac equation to include all relativistic effects.

Combine effective Hamiltonian with classical fields

Classical (external) E.M. fields in Maxwell equations:

$$\boxed{\vec{B}(\vec{r},t) = \vec{\nabla} \times \vec{A}(\vec{r},t), \quad \vec{E}(\vec{r},t) = -\partial \vec{A}(\vec{r},t) \big/ \partial t}$$

Coulomb gauge

With $A(r,t)$ the vector potential

Combine with single-particle electron Hamiltonian:

$$\hat{H}_0 = \left[-\frac{1}{2m} p^2 + V(r) + V_{xc}(r) \right]$$

Use: $\hat{p} \Rightarrow \hat{p} - e\vec{A}$

$$\hat{H} = \left[-\frac{1}{2m} (\hat{p} - e\vec{A})^2 + V(r) + V_{xc}(r) \right] \approx \left[-\frac{1}{2m} \hat{p}^2 + V(r) + V_{xc}(r) \right] + \frac{e}{m} (\hat{p} \cdot \vec{A})$$

Unperturbed H_0

Perturbation H′

P1

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Perturbation treatment of light-matter interaction

The light-matter interaction is given by $\hat{H}' = \frac{e}{m}(\hat{\vec{p}} \cdot \vec{A}) = (\hat{\vec{j}} \cdot \vec{A})$

This can be rewritten as $\hat{H}' = -e\ (\hat{\vec{r}} \cdot \vec{E})$

[Using that $A = (B \times r)/2$ and $(A.r) = 0$]

For linear optics & magneto-optics:
Compute effect of perturbation to first order in *E*

$$\boldsymbol{j}(\boldsymbol{k},\omega) = \boldsymbol{\sigma}(\boldsymbol{k},\omega) \cdot \boldsymbol{E}(\boldsymbol{k},\omega)$$

Linear-order response function

Use perturbation theory or linear-response theory

The described effects will be linear in perturbing field (*E* or *A*).
More work is needed to include non-linear optical effects!

Slide 34

P1 Peter, 6/2/2018

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Result of linear-response theory

Dielectric tensor/ Conductivity tensor

$$\boxed{\boldsymbol{\epsilon}(\omega) = \mathbf{1} + \frac{4\pi i}{\omega}\boldsymbol{\sigma}(\omega)}$$

Conductivity is the response function to the *E*-field:

$$j_\alpha(t) = \int_{-\infty}^{t} dt' \sigma_{\alpha\beta}(t-t') E_\beta(t')$$

$$\boldsymbol{E}(t) = \boldsymbol{E}\,\mathrm{e}^{-i\omega t}$$

Gives:

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

Fermi function

And:

$$\Pi^\alpha_{nn'} \equiv \langle n|p_\alpha|n'\rangle$$

$$\hbar\omega_{n'n} \equiv \epsilon_{n'} - \epsilon_n$$

Single particle eigenstates & eigenenergies

For a derivation, see the Appendix!

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Electronic structure picture

Expression sums contributions from all optical transitions, with 1-photon in, and 1-photon out

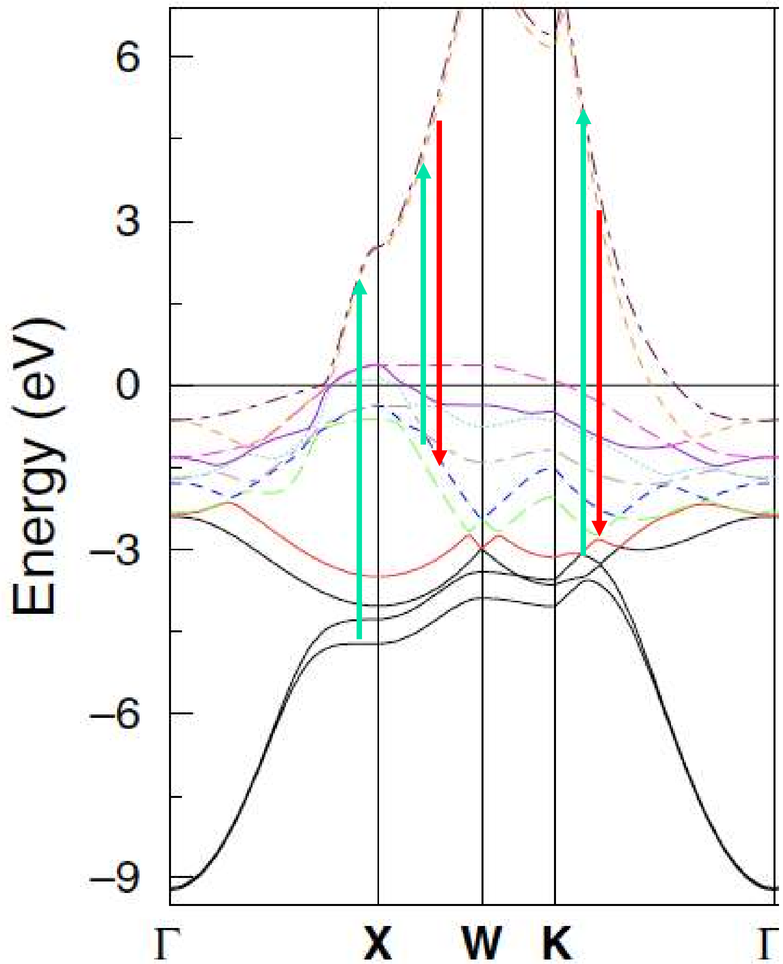
$$k = \frac{2\pi}{\lambda} \Rightarrow k \quad small$$

Dipole transitions:

$$\Delta l = \pm 1, \quad \Delta m = \pm 1, 0$$

Due to matrix elements

Sum all optical transitions



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Expressions for dielectric tensor

Use relation between tensors:

$$\boldsymbol{\epsilon}(\omega) = \mathbf{1} + \frac{4\pi i}{\omega}\boldsymbol{\sigma}(\omega)$$

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

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

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

Thus, we have an electronic structure expression for ε, from which we can in principle compute all spectra!

1) Examples of compute x-ray magnetic spectra come in Lecture II.

2) These equations are equivalent to those of the Fermi´s golden rule.

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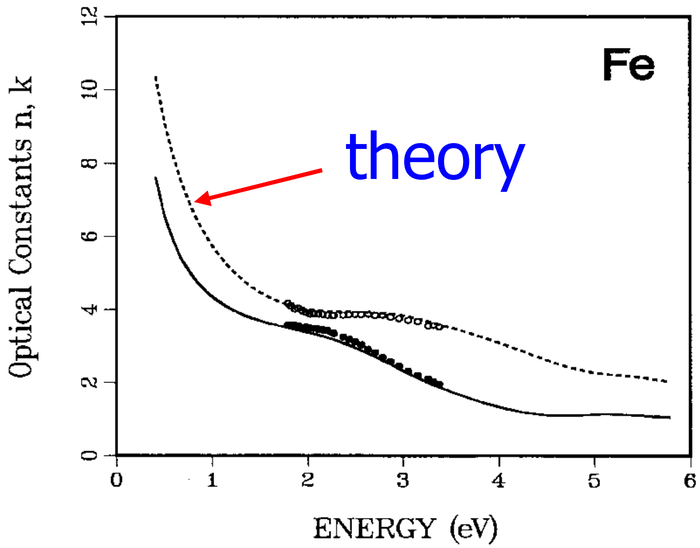
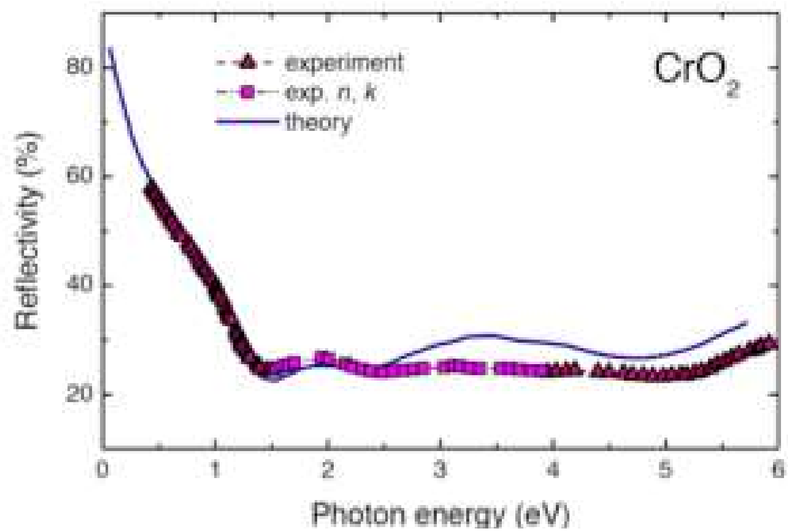
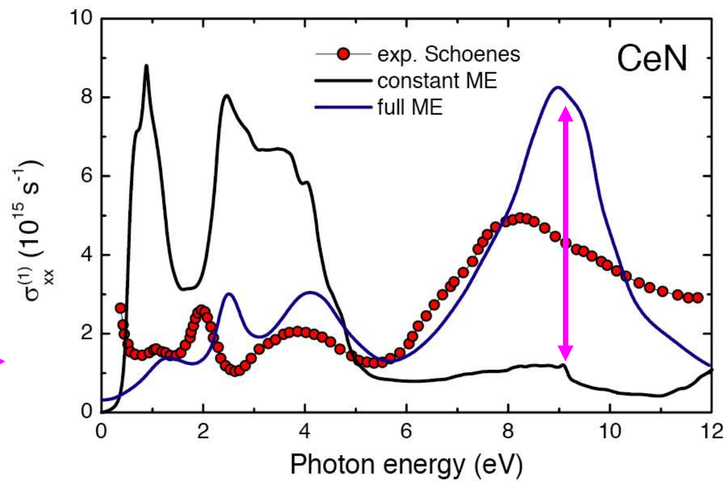
Some examples ...



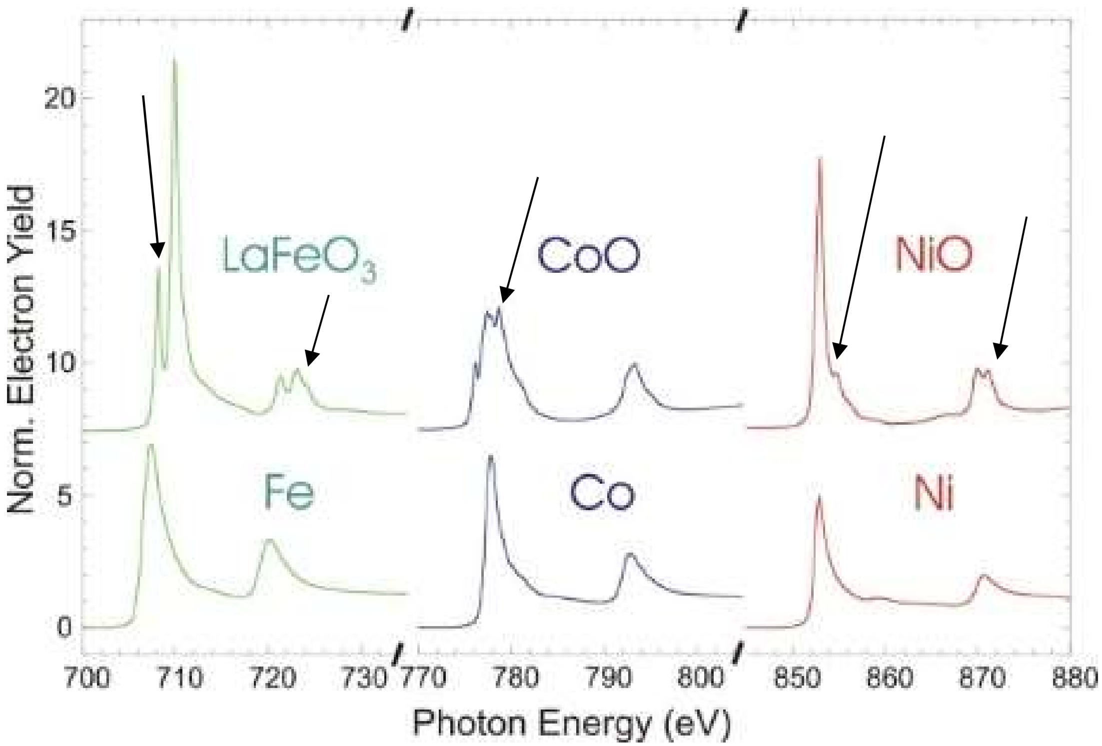


The comparison *ab initio* theory – experiment is often very good!

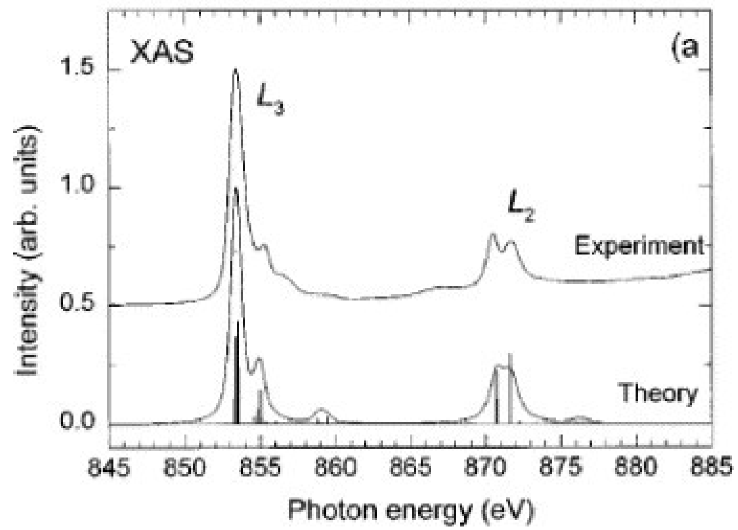
Importance of precise transition matrix elements



Limitations of the single-particle approach



NiO



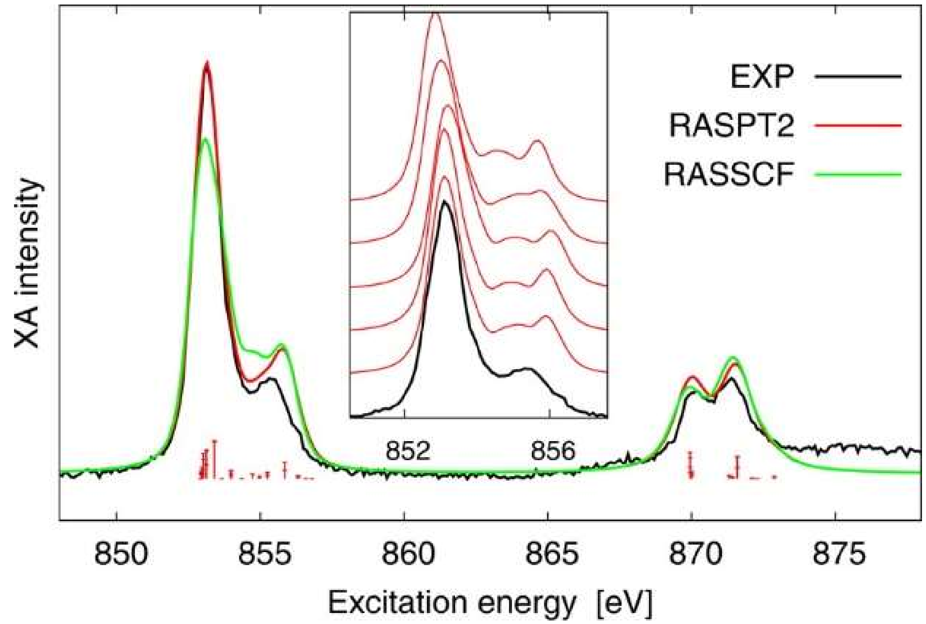
Single-ion calculations $3d^8$

Multiplet structures due atomic multi-electron configurations not included in 1-particle model

De Groot, Coord. Chem. Rev. **249**, 31 (2005)

To go beyond these limitations

Atomic multiplets with multiconfigurational SCF approach
(”active space”)



Josefsson et al, JPCL **3**, 3565 (2012)

Other approaches beyond effective single-e theory

1) Dyson equation:

electron state energy, wave function

$$\left[\frac{\mathbf{p}^2}{2m} + V_{\text{ext}} + V_H\right]\psi_{n\mathbf{k}}(\mathbf{r}) + \int \mathrm{d}^3\mathbf{r}'\, \underline{\Sigma(\mathbf{r},\mathbf{r}';E_{n\mathbf{k}})}\, \psi_{n\mathbf{k}}(\mathbf{r}') = E_{n\mathbf{k}}\ \psi_{n\mathbf{k}}(\mathbf{r})$$

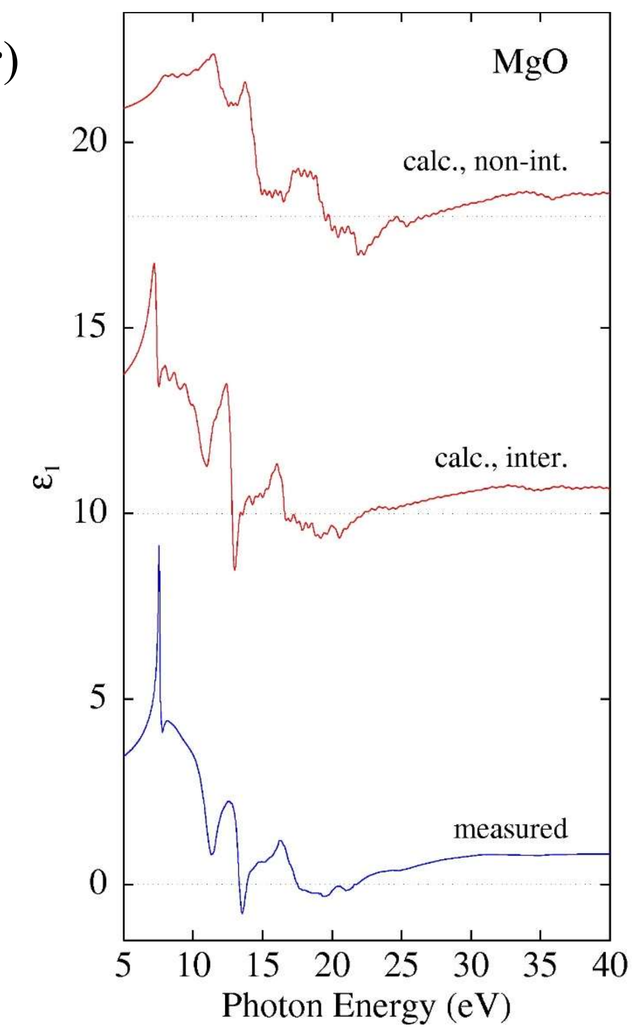
GW self-energy (accounts for many-body electron-electron interaction effects)

2) With explicit core-hole interaction:

core-hole effect

$$\left[\frac{p^2}{2m} + V_{\mathrm{N}}(\mathbf{r}) + V_{\mathrm{H}}(\mathbf{r}) + \underline{V_c(\mathbf{r})}\right] G(\mathbf{r},\mathbf{r}';E) +$$
$$\int \mathrm{d}^3\mathbf{r}''\, \Sigma(\mathbf{r},\mathbf{r}'';E)\, G(\mathbf{r}'',\mathbf{r}';E) = E\ G(\mathbf{r},\mathbf{r}';E)$$

electron-core hole Bethe-Salpeter equation



Improvement especially for non-metallic materials

(E. Shirley, J.J. Rehr)

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3rd level – quantized photon field

The 3rd level is a next step, where the photon is a *quantized field*

$$\hat{H} = K_e + V_{e-e} + V_{e-ion} + V_{ion-ion} + \sum \hbar\omega\, \hat{a}^+_{k\lambda}\hat{a}_{k\lambda} - \int d\vec{r}\left[\vec{j}(\vec{r},t)\cdot\vec{A}^{tot}(\vec{r},t)\right]$$

in 2nd quantization.

The vector potential is not just the external one, but is renormalized due to the electron response (feedback effect on the fields or "photon dressing")

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \nabla^2\right)\vec{A}(\vec{r},t) = \frac{4\pi}{c}\vec{J}(\vec{r},t)$$

Gives set of coupled Maxwell-Kohn-Sham equations that need to be solved selfconsistently!

$$\vec{A}(\vec{r},t) \propto \int d\vec{k} \sum_\lambda \frac{1}{\sqrt{2\omega_k}}\left[\hat{a}_k \varepsilon_\lambda e^{ik\cdot r - i\omega t} + \hat{a}^+_k \varepsilon^*_\lambda e^{-ik\cdot r + i\omega t}\right]$$

Example: small molecule in an optical cavity
(Fick et al, ACS Photon. **5**, 992 (2018)

Summarizing light-matter interaction

Most basic principles of (macroscopic) light-matter interaction are given by the Maxwell-Fresnel theory

Combining classical Maxwell fields with *ab initio* quantum theory (effective single particle theory) gives often quite *accurate* valence band and X-ray optical spectra of many materials

(*Ab initio DFT* approache gives reasonable description of electronic structure properties for relatively low computational costs)

Current frontlines:

1) Beyond DFT single-particle theory to include many-particle interactions in the excited state
2) Quantized photon fields coupled selfconsistently to DFT Kohn-Sham equations

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Appendix I: Linear response theory

Here we briefly go through some steps of the Kubo theory derivation:

Density matrix definition: $\rho(t) = \mathrm{e}^{-\beta H}/Z$ $\quad Z = \mathrm{Tr}\{\exp(-\beta H)\}$

(partition function)

Expectation value of operator ***O***: $\langle \mathcal{O}(t) \rangle = \mathrm{Tr}\{\rho(t)\mathcal{O}\}$

(Schrödinger picture)

For the time-dependence of any expectation value of operator ***O***:

$$\frac{d\rho(t)}{dt} = \frac{1}{i\hbar}[H(t), \rho(t)]$$

Linear-approximation in H_1:

$$\rho^{\mathrm{i}}(t) \approx \rho_0 + \frac{1}{i\hbar} \int_{-\infty}^{t} dt' \left[H_1^{\mathrm{i}}(t'), \rho_0\right]$$

$$A^i(t) = e^{iH_0 t/\hbar} A_s e^{-iH_0 t/\hbar}, \quad |\psi(t)\rangle^i = e^{iH_0 t/\hbar} |\psi(t)\rangle_s$$

(interaction picture)

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Linear response theory, continued

For any operator $\boldsymbol{O}$ we get the time-dependence induced through the perturbing Hamiltonian:

$$\langle \mathcal{O}(t) \rangle \approx \langle \mathcal{O} \rangle_0 + \frac{1}{i\hbar} \int_{-\infty}^{t} dt' \langle [\mathcal{O}^{\mathrm{i}}(t), H_1^{\mathrm{i}}(t')] \rangle_0$$

If you want details, see the appendix!

(Ensamble average with respect to the unperturbed states, interaction picture)

This is already linear-response theory:

$$\langle \mathcal{O}(t) \rangle \approx \int_{-\infty}^{t} dt' \underbrace{R(t-t')}_{\text{„response function"}} F(t')$$

With: $H_1(t) = BF(t)$

Note: $\tau = t\text{-}t'$, response is always causal

$$R(\tau) = \frac{1}{i\hbar}\theta(\tau)\langle [\mathcal{O}^{\mathrm{i}}(\tau), B^{\mathrm{i}}(0)] \rangle_0$$

Conductivity response to EM field

Conductivity is the response function to the E-field:

$$j_\alpha(t) = \int_{-\infty}^{t} dt' \sigma_{\alpha\beta}(t-t') E_\beta(t')$$

$$\boldsymbol{E}(t) = \boldsymbol{E}\,\mathrm{e}^{-i\omega t}$$

The perturbing hamiltonian can be written as:

$$H_1 = e \sum_i \boldsymbol{r}_i \cdot \boldsymbol{E}(t) \equiv B\,\mathrm{e}^{-i\omega^+ t}$$

$$\omega^+ \equiv \omega + i\delta$$

$$\vec{J} = -e \sum_i \dot{\vec{r}}_i$$

Thus, we have to work out: $$J_\alpha(t) = \frac{1}{i\hbar} \int_{-\infty}^{t} dt' \left\langle \left[J_\alpha^{\mathrm{i}}(t), B^{\mathrm{i}}(t') \right] \right\rangle_0 \mathrm{e}^{-i\omega^+ t'}$$

PI for total current J:

$$J_\alpha(t) = \frac{1}{i\hbar} \left\langle \left[J_\alpha^{\mathrm{i}}(t), B^{\mathrm{i}}(t) \right] \right\rangle_0 \frac{\mathrm{e}^{-i\omega^+ t}}{-i\omega^+} - \frac{1}{i\hbar} \int_{-\infty}^{t} dt' \left\langle \left[J_\alpha^{\mathrm{i}}(t), \dot{B}^{\mathrm{i}}(t') \right] \right\rangle_0 \frac{\mathrm{e}^{-i\omega^+ t'}}{-i\omega^+}$$

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Conductivity response

With: $\dot{B}^{\mathrm{i}}(t) = e\sum_i \dot{\boldsymbol{r}}_i^{\mathrm{i}} \cdot \boldsymbol{E} = -\boldsymbol{J}^{\mathrm{i}}(t) \cdot \boldsymbol{E}$

And: $\left\langle \left[J_\alpha^{\mathrm{i}}(t), B^{\mathrm{i}}(t) \right] \right\rangle_0 = -e^2 \sum_{ij} \left\langle \left[\dot{r}_{i,\alpha}(0), r_{j,\beta}(0) \right] \right\rangle_0 E_\beta = \frac{ie^2\hbar}{m} N \delta_{\alpha\beta} E_\beta$

$$\Longrightarrow \quad J_\alpha(t) = \frac{ie^2 N \delta_{\alpha\beta}}{m\omega^+} E_\beta \, \mathrm{e}^{-i\omega^+ t} - \frac{1}{i\hbar} \int_{-\infty}^{t} dt' \left\langle \left[J_\alpha(t), J_\beta(t') \right] \right\rangle_0 \frac{E_\beta \, \mathrm{e}^{-i\omega^+ t'}}{i\omega^+}$$

Comparing with the equation for σ gives:

$$\sigma_{\alpha\beta}(t-t') = \frac{ie^2 N \delta_{\alpha\beta}}{V m \omega^+} \delta(t-t') + \frac{1}{V\hbar\omega^+} \left\langle \left[J_\alpha(t-t'), J_\beta(0) \right] \right\rangle_0$$

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Conductivity response, continued

Fourier transform:

$$\sigma_{\alpha\beta}(\omega) = \frac{ie^2 N\delta_{\alpha\beta}}{Vm\omega^+} + \frac{1}{V\hbar\omega^+}\int_{-\infty}^{\infty} d\tau\,\theta(\tau)\big\langle\big[J_\alpha(\tau), J_\beta(0)\big]\big\rangle_0\, \mathrm{e}^{\,i\omega^+\tau}$$

$$\boxed{\sigma_{\alpha\beta}(\omega) = \frac{ie^2 N\delta_{\alpha\beta}}{Vm\omega^+} + \frac{i}{V\omega^+}\sum_{\kappa\kappa'}\frac{1}{Z}\left[\mathrm{e}^{-E_\kappa/kT} - \mathrm{e}^{-E_{\kappa'}/kT}\right]\frac{\langle\kappa|J_\alpha|\kappa'\rangle\langle\kappa'|J_\beta|\kappa\rangle}{\hbar\omega^+ - (E_{\kappa'} - E_\kappa)}}$$

With: $\vec{J} = -e\sum_i \vec{v}_i = -\frac{e}{m}\sum_i \vec{p}_i = -\frac{e}{m}\sum_i (-i\hbar)\vec{\nabla}_i$

Single particle formulation

Rewrite for single particle states:

$$\sigma_{\alpha\beta}(\omega) = \frac{ie^2 N\delta_{\alpha\beta}}{Vm\omega^+} + \frac{ie^2}{m^2 V\hbar\omega^+}\sum_{nn'}\frac{f(\epsilon_n)-f(\epsilon_{n'})}{\omega^+ - \omega_{n'n}}\,\Pi^\alpha_{nn'}\Pi^\beta_{n'n}$$

With: $\Pi^\alpha_{nn'} \equiv \langle n|p_\alpha|n'\rangle$ $\quad \hbar\omega_{n'n} \equiv \epsilon_{n'} - \epsilon_n$

Can be written as:

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

Fermi function

Use:

$$\begin{aligned} N\delta_{\alpha\beta} &= \sum_n f(\epsilon_n)\,\delta_{\alpha\beta} = \sum_n f(\epsilon_n)\,\langle n|[r_\alpha, p_\beta]|n\rangle\frac{1}{i\hbar} \\ &= \sum_{nn'}\frac{f(\epsilon_n)-f(\epsilon_{n'})}{i\hbar}\,r^\alpha_{nn'}\Pi^\beta_{n'n} \\ &= -\frac{1}{m\hbar}\sum_{nn'}\frac{f(\epsilon_n)-f(\epsilon_{n'})}{\omega_{nn'}}\,\Pi^\alpha_{nn'}\Pi^\beta_{n'n}\,. \end{aligned}$$

Appendix II: Transition matrix elements

The matrix elements have special properties, called selection rules

Rewrite: $\boldsymbol{\Pi} = \frac{im}{\hbar}[H, \boldsymbol{r}] \Longrightarrow \boldsymbol{\Pi}_{nn'} = \frac{im}{\hbar}(\epsilon_n - \epsilon_{n'}) \langle \psi_n | \boldsymbol{r} | \psi_{n'} \rangle$

and consider an atomic basis: $\psi_{n\vec{k}}(\vec{r}) \sim \sum_{lm} C_{lm}^{n}(\vec{k}) f_{lm}^{n}(r) Y_{lm}(\hat{r})$

This leads to: $\langle \psi_n | \vec{r} | \psi_{n'} \rangle \rightarrow \int_S d\Omega Y_{lm}^{*}(\hat{r}) \underbrace{\hat{r} Y_{l'm'}(\hat{r})}_{Y_{l'\pm1, m'\pm1,0}}$

$$Y_{l'\pm1, m'\pm1,0} \rightarrow \quad l = l'\pm1, \quad m = m'\pm1,0$$

$\Longrightarrow$ Dipolar transitions have: $\Delta l = \pm1, \quad \Delta m = \pm1,0$

Example:

2p states -> 3d states,
4s states (L-edge)

Selection rules on m :

$$\left\{ \begin{array}{l} \varepsilon_+ = \varepsilon_{xx} + i\varepsilon_{xy} \Rightarrow \Delta m = +1 \\ \varepsilon_- = \varepsilon_{xx} - i\varepsilon_{xy} \Rightarrow \Delta m = -1 \\ \varepsilon_0 = \varepsilon_{\|} \Rightarrow \Delta m = 0 \end{array} \right.$$

Appendix III: Fermi's golden rule

$$\mu(\omega) \propto \sum_j \left| \left\langle \psi_j \left| \vec{\varepsilon} \cdot \vec{r} \right| \psi_i \right\rangle \right|^2 \delta(E_f - E_i - \hbar\omega)$$

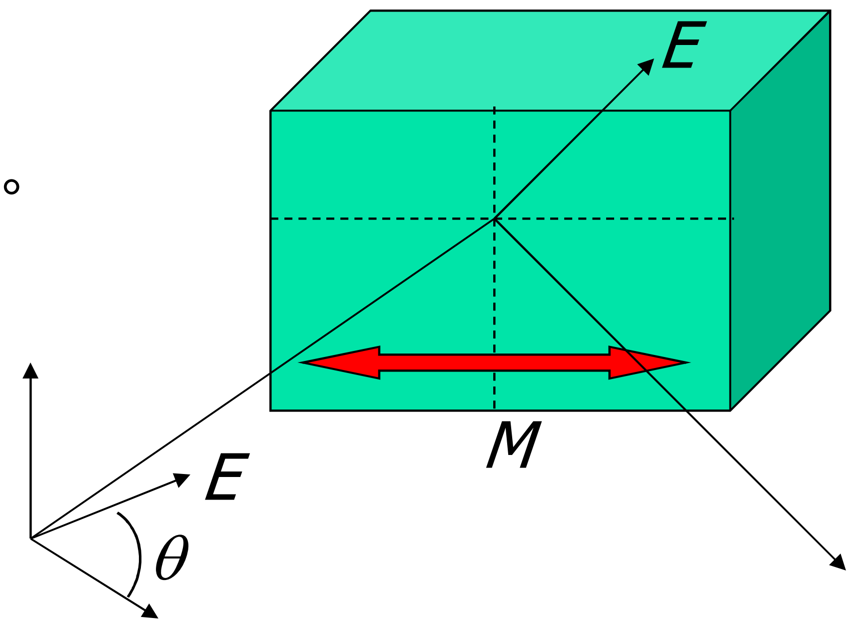
From Fermi's golden rule $W_{i \to f} = \dfrac{2\pi}{\hbar} \left| M \right|^2 \delta(E_f - E_i - \hbar\omega)$

with $M = \left\langle \psi_f \left| H' \right| \psi_i \right\rangle = \int \psi_f^*(\vec{r}) H'(\vec{r}) \psi_i(\vec{r}) d\vec{r}$

and $H' = e(\vec{\varepsilon} \cdot \vec{r}) e^{i\vec{k} \cdot \vec{r}}$ (perturbation due to radiation field)

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:

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