Synchrotron Radiation for Materials Science Applications

Polarized X-rays from a Bending Magnet and X-ray Magnetic Circular Dichroism (XMCD)

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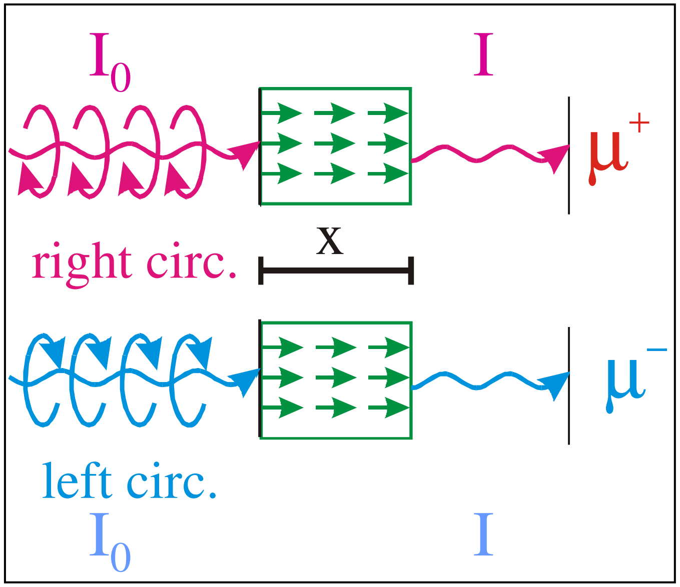
XMCD

X-ray Magnetic Circular Dichroism

Magnetization dependent absorption of circularly polarized light



recall from previous lectures:

$$\frac{I}{I_0} = e^{-\rho\mu x} \qquad (text\, eqn: \quad 1.3a)$$

where

$$\mu = \mu(\hbar\omega, Z, M \cdot \sigma) = \mu(\hbar\omega, Z) + \underline{\mu(M \cdot \sigma)}$$

Another notation: $I = I_0 e^{-\mu_\rho x}$

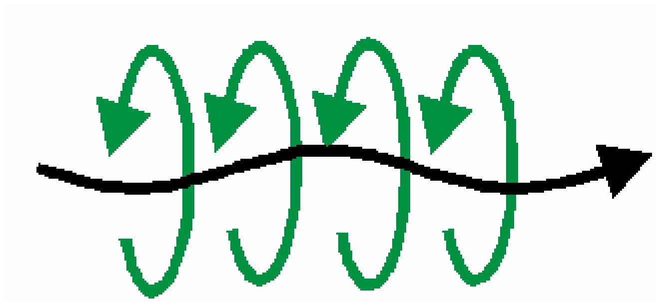
$\mu_\rho = \rho\sigma^{abs}$ *where* σ^{abs} = *X − ray Absorption cross − section*

Circularly Polarized Light

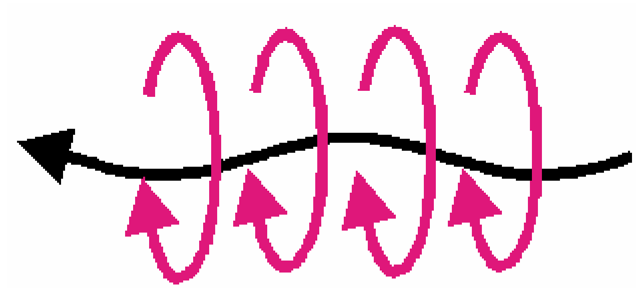
Photon angular momentum: $\sigma = \sigma\hat{k}$

Where σ=photon helicity =±1

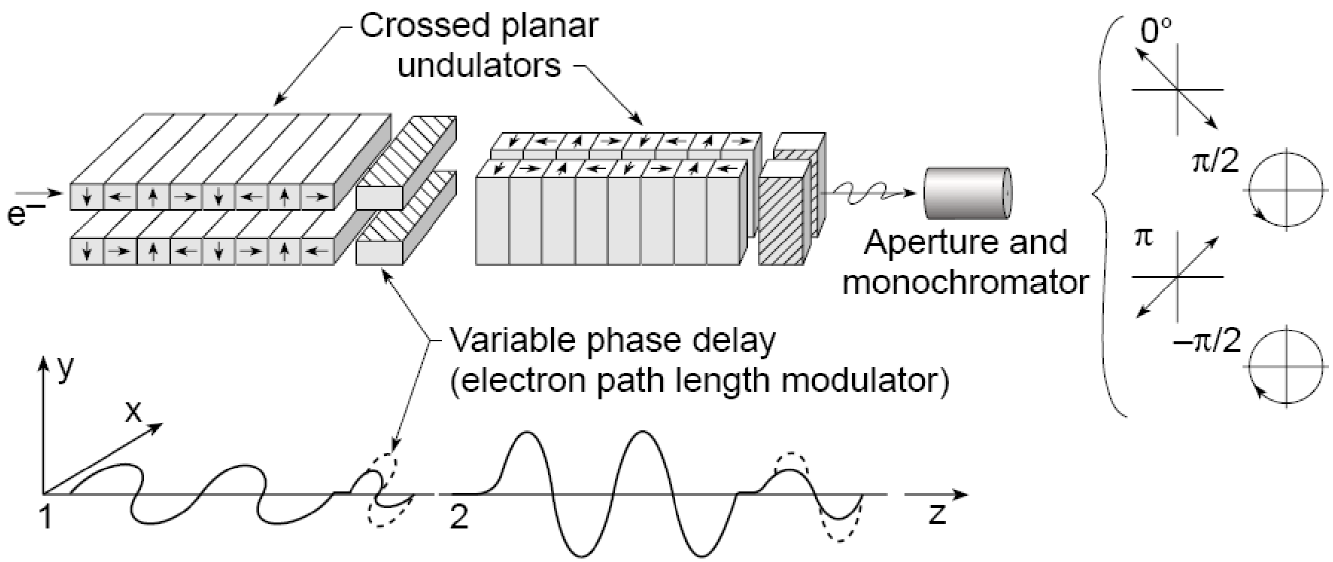
Right circularly polarized
σ=+1



Left circularly polarized
σ=-1

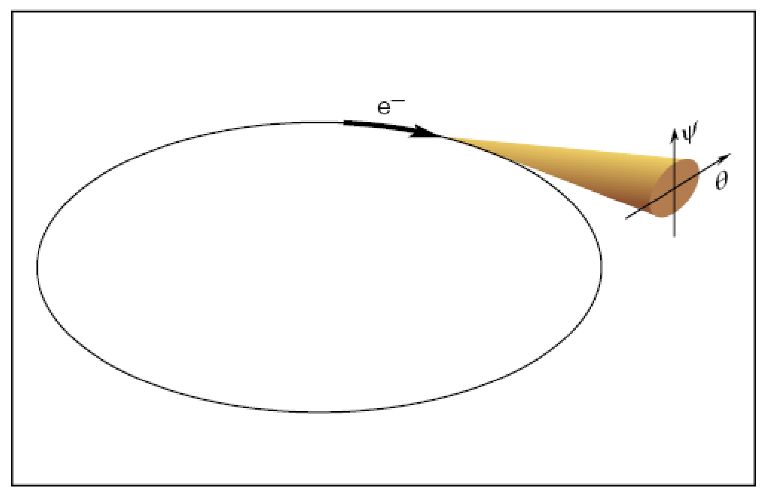


Polarized Radiation at a Synchrotron Source

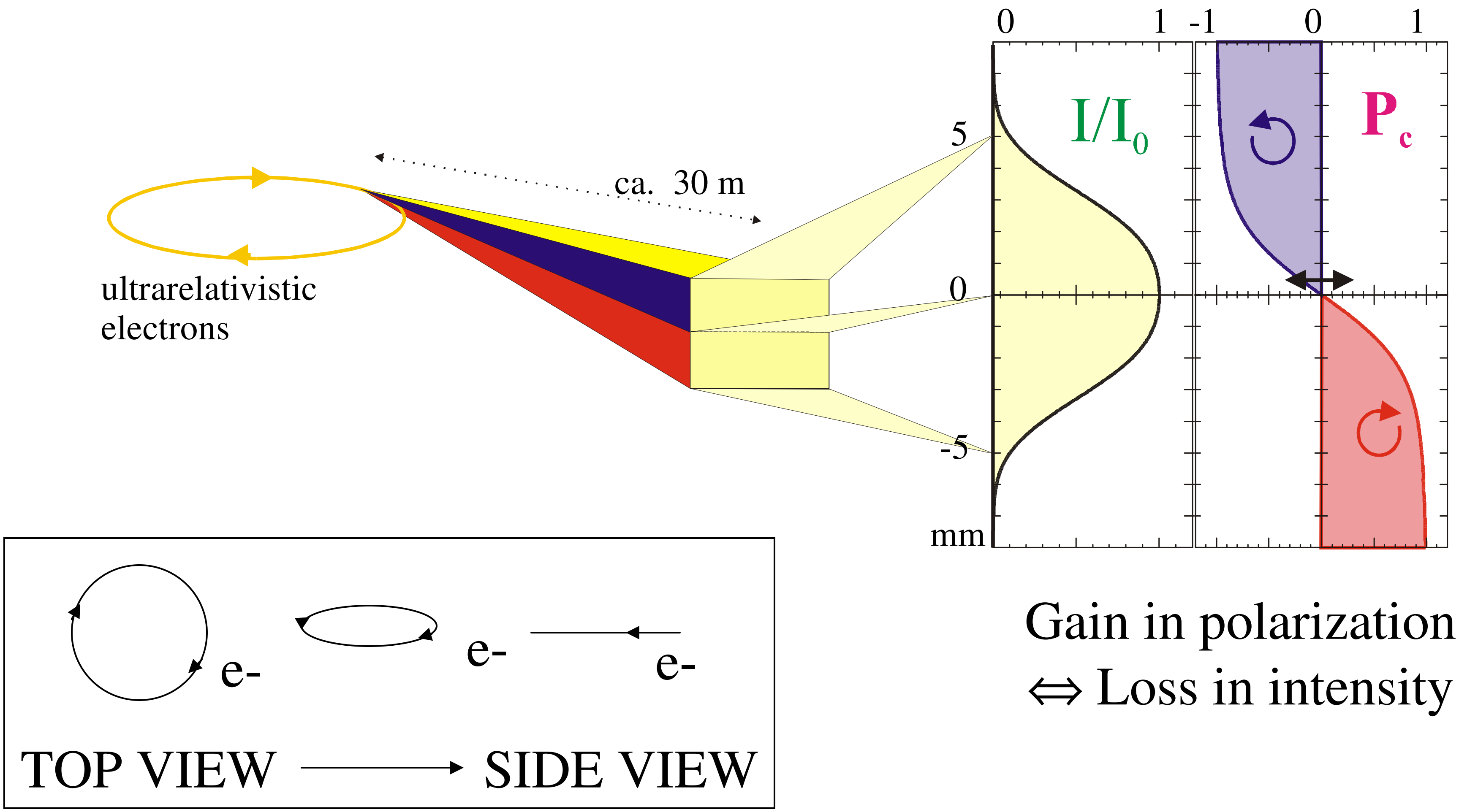


(Courtesy of Kwang-Je Kim)

Polarized light from an EPU or from off-axis bending magnet radiation

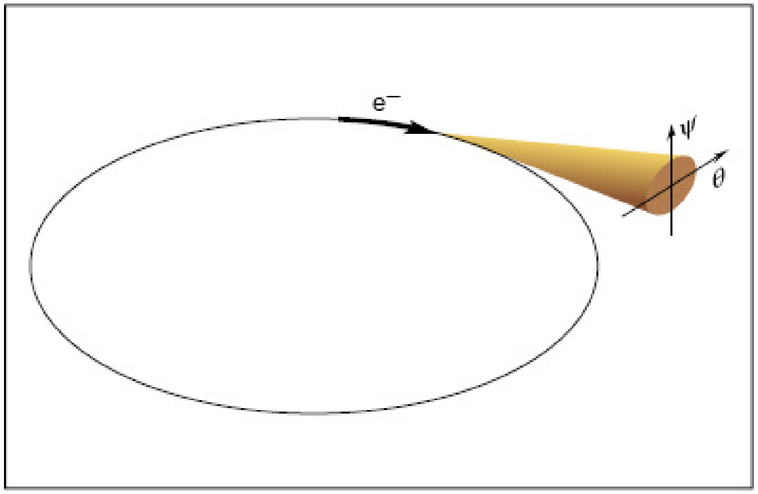


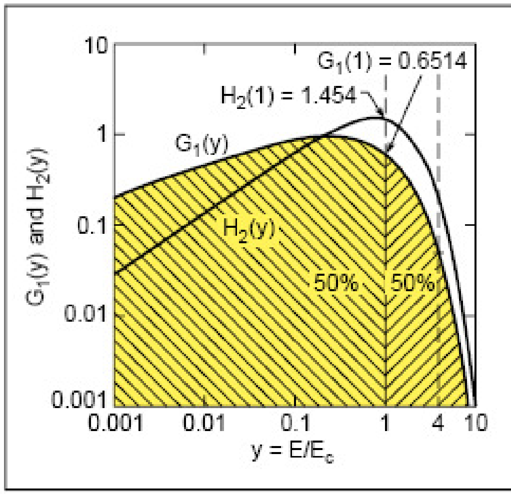
Polarization properties of SR



Gain in polarization
⇔ Loss in intensity

Bending Magnet Radiation On-axis





y	$G_1(y)$	$H_2(y)$
0.0010	2.131×10^{-1}	2.910×10^{-2}
0.0100	4.450×10^{-1}	1.348×10^{-1}
0.1000	8.182×10^{-1}	6.025×10^{-1}
0.3000	9.177×10^{-1}	1.111×10^{0}
0.5000	8.708×10^{-1}	1.356×10^{0}
0.7000	7.879×10^{-1}	1.458×10^{0}
1.000	6.514×10^{-1}	1.454×10^{0}
3.000	1.286×10^{-1}	5.195×10^{-1}
5.000	2.125×10^{-2}	1.131×10^{-1}
7.000	3.308×10^{-3}	2.107×10^{-2}
10.00	1.922×10^{-4}	1.478×10^{-3}

$$E_c = \hbar\omega_c = \frac{3e\hbar B\gamma^2}{2m} \qquad (5.7a)$$

$$E_c(\text{keV}) = 0.6650 E_e^2(\text{GeV}) B(\text{T}) \qquad (5.7b)$$

$$\gamma = \frac{E_e}{mc^2} = 1957 E_e(\text{GeV}) \qquad (5.5)$$

$$\left.\frac{d^3 F_B}{d\theta\, d\psi\, d\omega/\omega}\right|_{\psi=0} = 1.33 \times 10^{13} E_e^2(\text{GeV}) I(\text{A}) H_2(E/E_c) \frac{\text{photons/s}}{\text{mrad}^2 \cdot (0.1\%\ \text{BW})} \qquad (5.6)$$

$$\left.\frac{d^2 F_B}{d\theta\, d\omega/\omega}\right|_{\psi=0} = 2.46 \times 10^{13} E_e(\text{GeV}) I(\text{A}) G_1(E/E_c) \frac{\text{photons/s}}{\text{mrad} \cdot (0.1\%\text{BW})} \qquad (5.8)$$

Power per Solid Angle

recall from previous lectures :

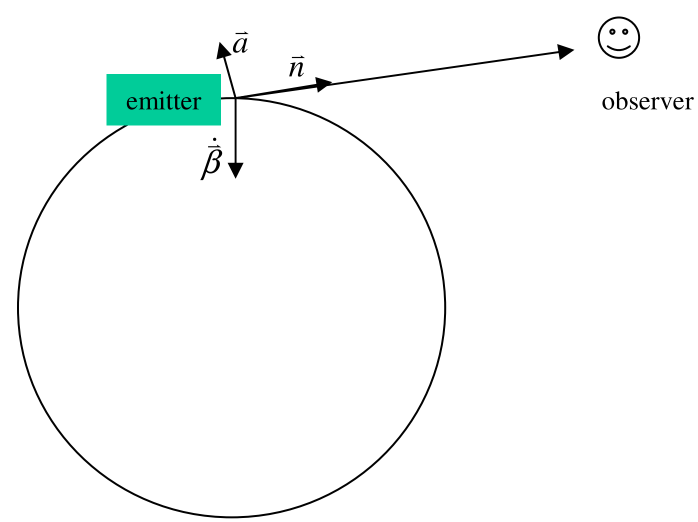
$$\frac{power}{area}: \qquad \vec{S}(\vec{r},t) = \frac{e^2 \left|\vec{a}_T\right|^2}{16\pi^2 \varepsilon_0 c^3 r^2}\vec{k}_0 \qquad (text\ eqn: 2.32)$$

$$and\ \frac{power}{SolidAngle}: \qquad \frac{dP}{d\Omega} = \frac{e^2 \left|\vec{a}\right|^2 \sin^2\Theta}{16\pi^2 \varepsilon_0 c^3} \qquad (text\ eqn: 2.34)$$

$$\Rightarrow \frac{power}{SolidAngle} \propto \left|\vec{a}_T\right|^2$$

$$note: \vec{a}_T = transverse\ acceleration$$

$$\vec{a}_T = -\vec{n}\times(\vec{n}\times\vec{a}) \qquad and \qquad \left|\vec{a}_T\right| = \left|\vec{a}\right|\sin\Theta$$



Flux per Solid Angle

Now, following Kwang–Je Kim:

$$\frac{d^2F_B}{d\theta d\psi} = \alpha \frac{\Delta\omega}{\omega}\frac{I}{e}\left|A(\omega)\right|^2 \qquad \left(\frac{Flux}{SolidAngle}\right)$$

$$A(\omega) = \frac{\omega}{2\pi}\int \vec{a}(t')e^{i\omega t(t')}dt'$$

$$\alpha = 1/137$$

$$I = current \qquad (electron \rightarrow beam\ of\ electrons)$$

$$\frac{\Delta\omega}{\omega} = bandwidth$$

K.-J. Kim,"Characteristics of Synchrotron Radiation,"pp565-632 in *Physics of Particle Accelerators,* AIP Conference Proceedings 184 (American Inst. of Physics, New York 1989)

Horizontal and Vertical Components

We are interested in the polarization of the radiation

$\rightarrow$ break flux up into horizontal and vertical components

$$\begin{pmatrix} \dfrac{d^2 F_{B,h}}{d\theta d\psi} \\ \dfrac{d^2 F_{B,v}}{d\theta d\psi} \end{pmatrix} = \alpha \frac{\Delta\omega}{\omega} \frac{I}{e} \left| \begin{pmatrix} A_h \\ A_v \end{pmatrix} \right|^2$$

now using the modified Bessel functions we can write $A_{h,v}$ as:

$$\begin{pmatrix} A_h \\ A_v \end{pmatrix} = \frac{\sqrt{3}}{2\pi} \gamma \left(\frac{\omega}{\omega_c} \right)^2 (1 + X^2)(-i) \begin{pmatrix} K_{2/3}(\eta) \\ \dfrac{iX}{\sqrt{1+X^2}} K_{1/3}(\eta) \end{pmatrix}$$

$$\omega_c = \frac{3\gamma^2 eB}{2m}$$
$$\eta = \frac{1}{2}\frac{\omega}{\omega_c}(1+X^2)^{3/2}$$
$$X = \gamma\psi$$

(We'll see that this simplifies greatly for $\psi \rightarrow 0$)

K.-J. Kim, "Characteristics of Synchrotron Radiation," pp565-632 in *Physics of Particle Accelerators,* AIP Conference Proceedings 184 (American Inst. of Physics, New York 1989)

Flux as a function of angle

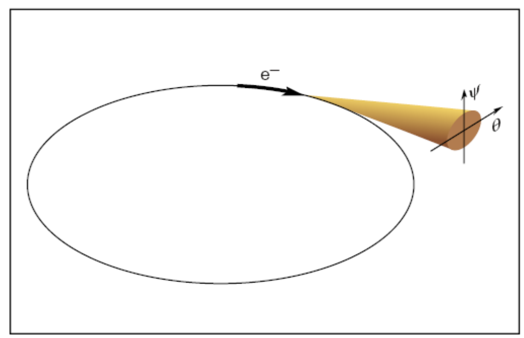
$$\begin{pmatrix} \dfrac{d^2F_{B,h}}{d\theta d\psi} \\ \dfrac{d^2F_{B,v}}{d\theta d\psi} \end{pmatrix} = \frac{3\alpha}{4\pi^2}\gamma^2 \frac{\Delta\omega}{\omega}\frac{I}{e}\left(\frac{\omega}{\omega_c}\right)^2 (1+X^2)^2 \begin{pmatrix} K_{2/3}^2(\eta) \\ \dfrac{X^2}{1+X^2}K_{1/3}^2(\eta) \end{pmatrix}$$

$$\omega_c = \frac{3\gamma^2 eB}{2m}$$
$$\eta = \frac{1}{2}\frac{\omega}{\omega_c}(1+X^2)^{3/2}$$
$$X = \gamma\psi$$

$$Let\ \psi = 0 \Rightarrow \ X = 0, \quad \frac{d^2F_{B,v}}{d\theta d\psi} = 0 \qquad \eta = \frac{1}{2}\left(\frac{\omega}{\omega_c}\right)$$

$$\left.\frac{d^2F_B}{d^2 d\theta d\psi\, d\omega/\omega}\right|_{\psi=0} = 1.33\times 10^{13}\, Ee^2(GeV)I(A)H_2(E/E_c)\frac{photons}{mrad^2\cdot(0.1\%BW)} \qquad (text\ eqn:\ 5.6)$$

$$where \quad H_2(E/E_c) = \left(\frac{\omega}{\omega_c}\right)^2 K_{2/3}^2\left(\frac{1}{2}\frac{\omega}{\omega_c}\right)$$



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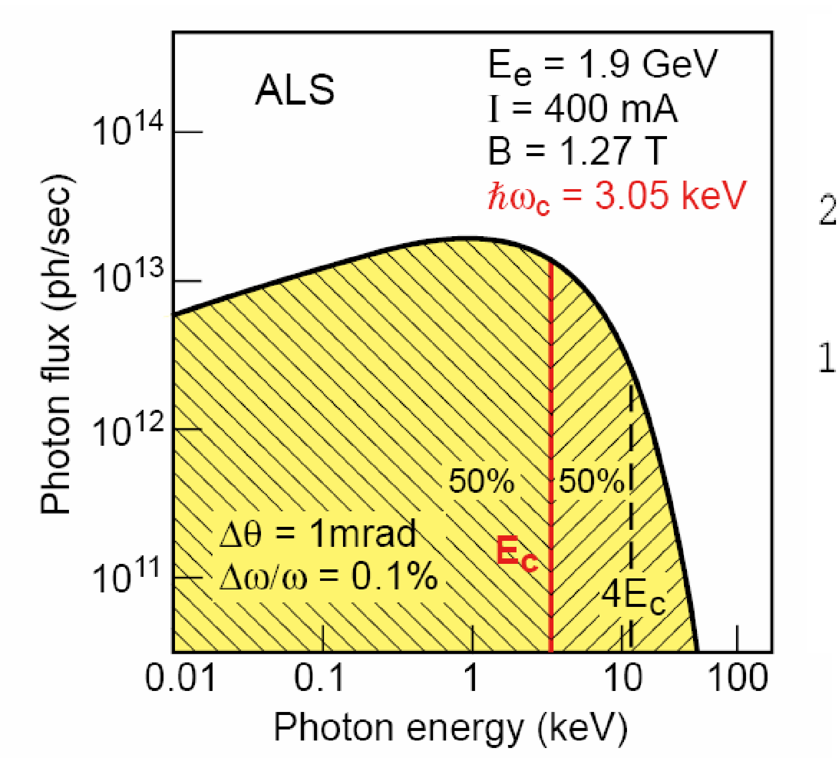
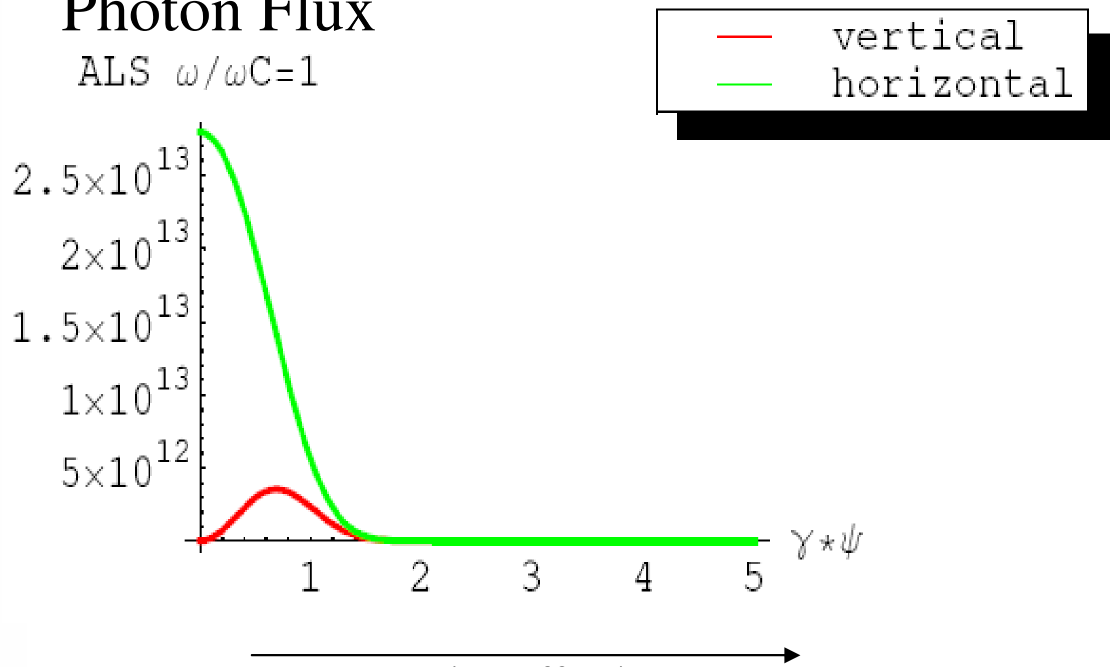
Behavior at Critical Energy



Horizontal (on axis) radiation as a function of energy

Photon Flux



Moving off axis

Behavior at 700eV

L adsorption edges of Fe are ~707eV, ~720eV

ALS:
Ee=1.9GeV, I=400mA, B=1.27T

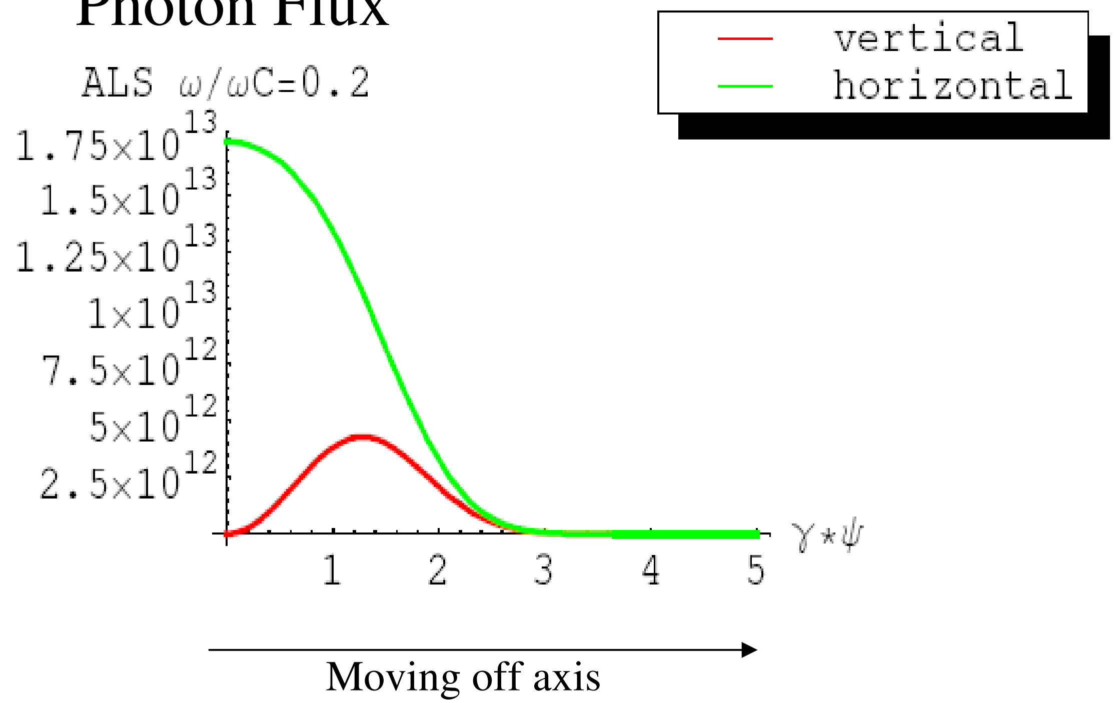
$Ec=0.6650*(Ee)^2[GeV]B[T]$
EcALS=3.05keV

700eV/3.05keV =0.2

γ=1957Ee[GeV]
γALS=3718.3

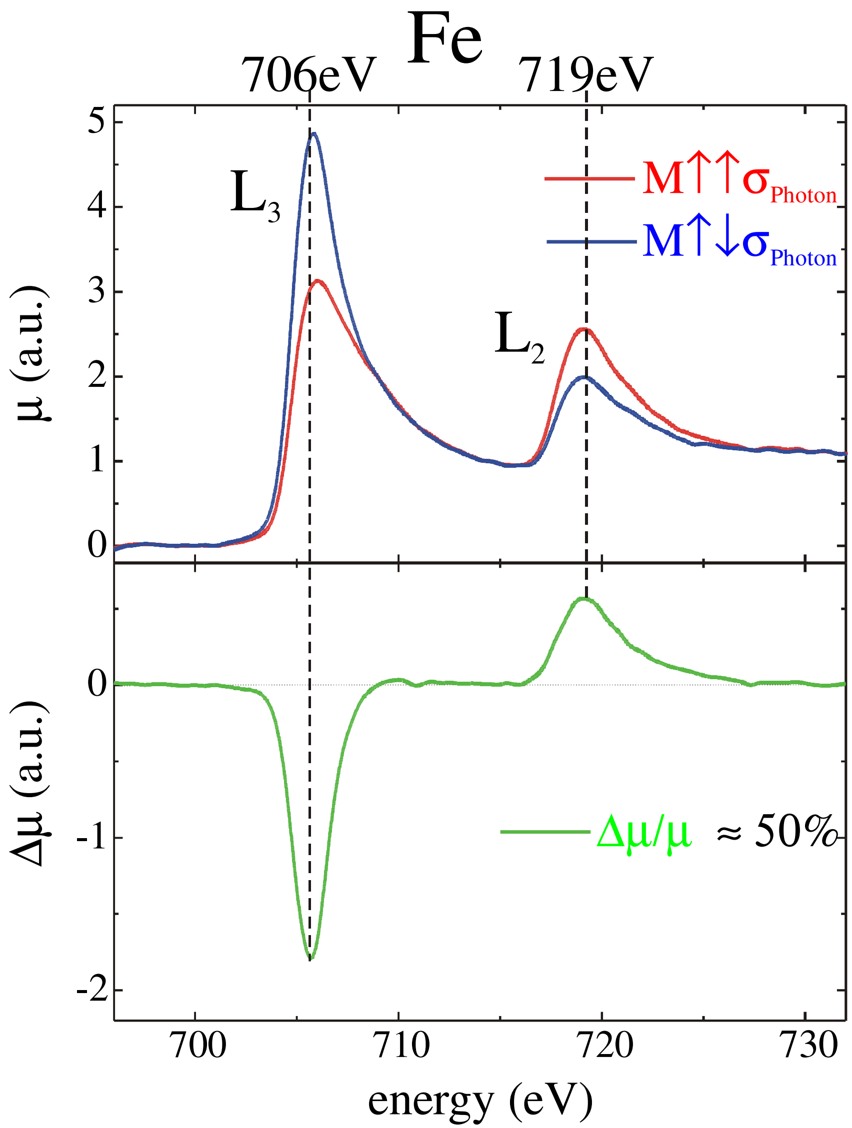
$\gamma*\psi=1 \rightarrow \psi \sim 2.7*10^{-4}$ radians

Photon Flux



Bending magnet radiation is naturally polarized at small angles off-axis

X-ray Magnetic Circular Dichroism (XMCD)

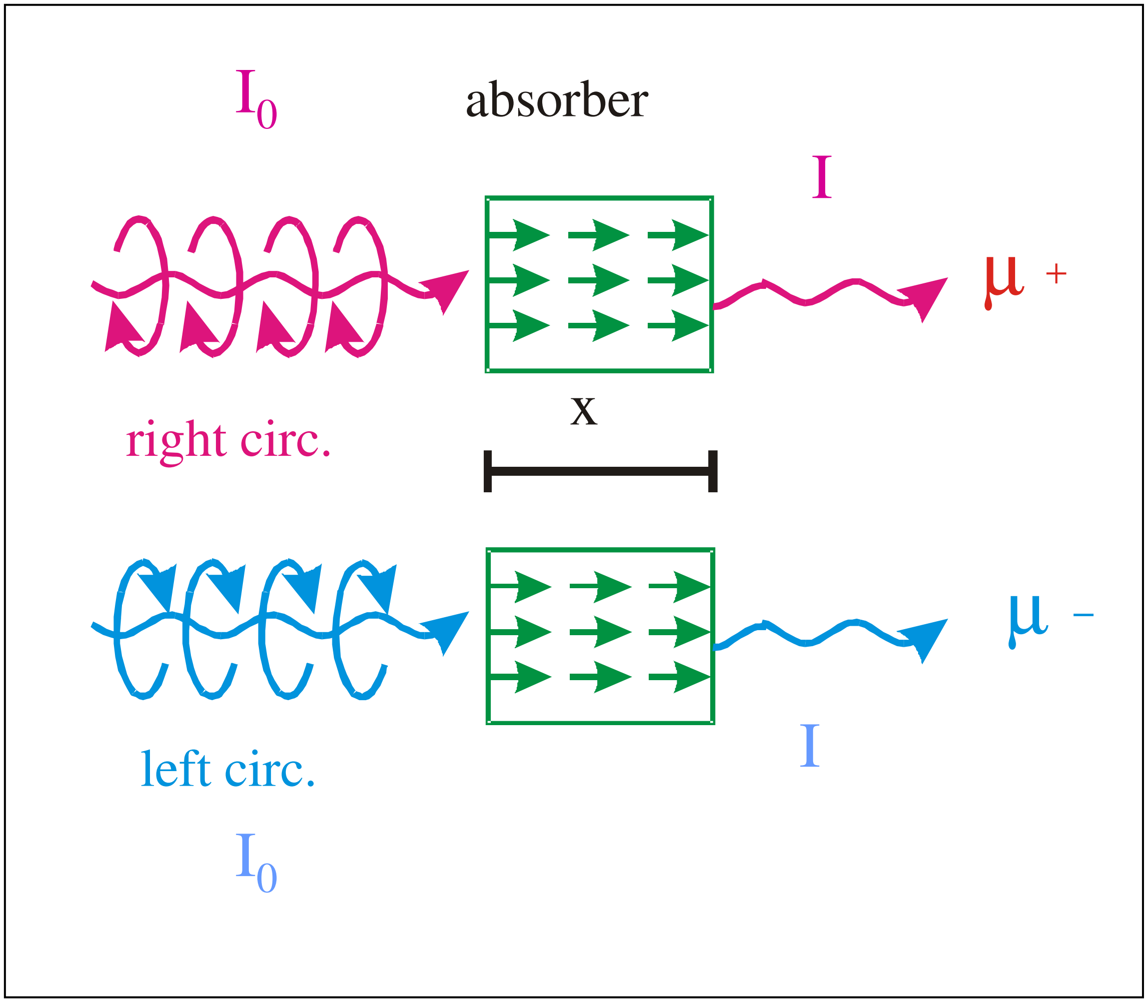


A typical XMCD result @ Fe $L_{3,2}$ absorption edges

- element specific
- huge magnetic contrast
- $\underline{M}\cdot\underline{\sigma}_{Photon}$
- Quantitative probe of spin and orbital moments

M=magnetization
= magnetic moment per volume

XMCD Measurement

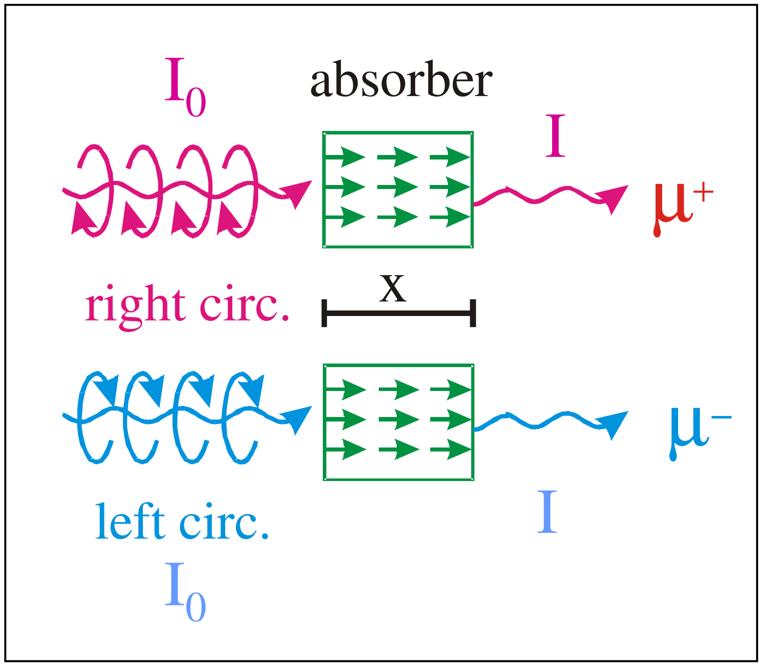


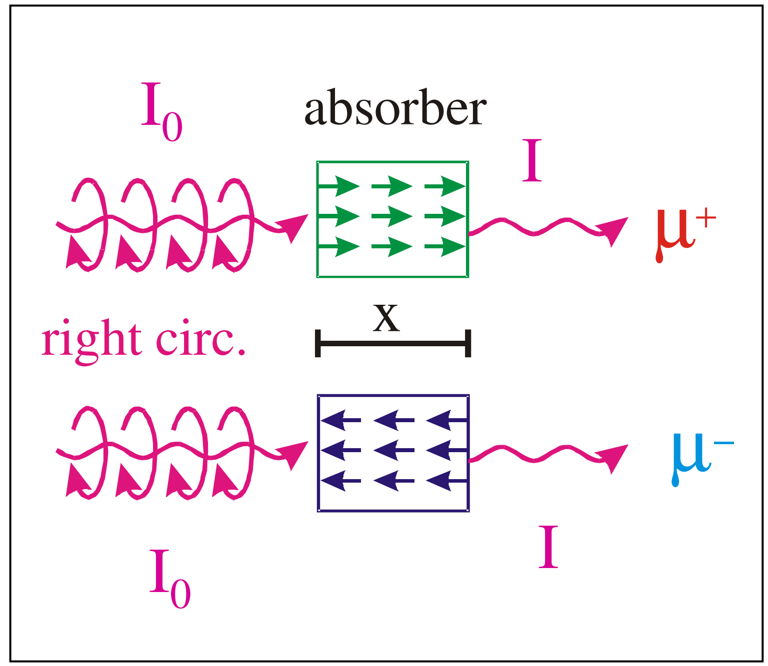
transmission mode

$$\left(\frac{I}{I_0}\right)^{\pm} = e^{-\rho\mu^{\pm}x}$$

dichroic signal $\mu^+ - \mu^- = \Delta\mu$

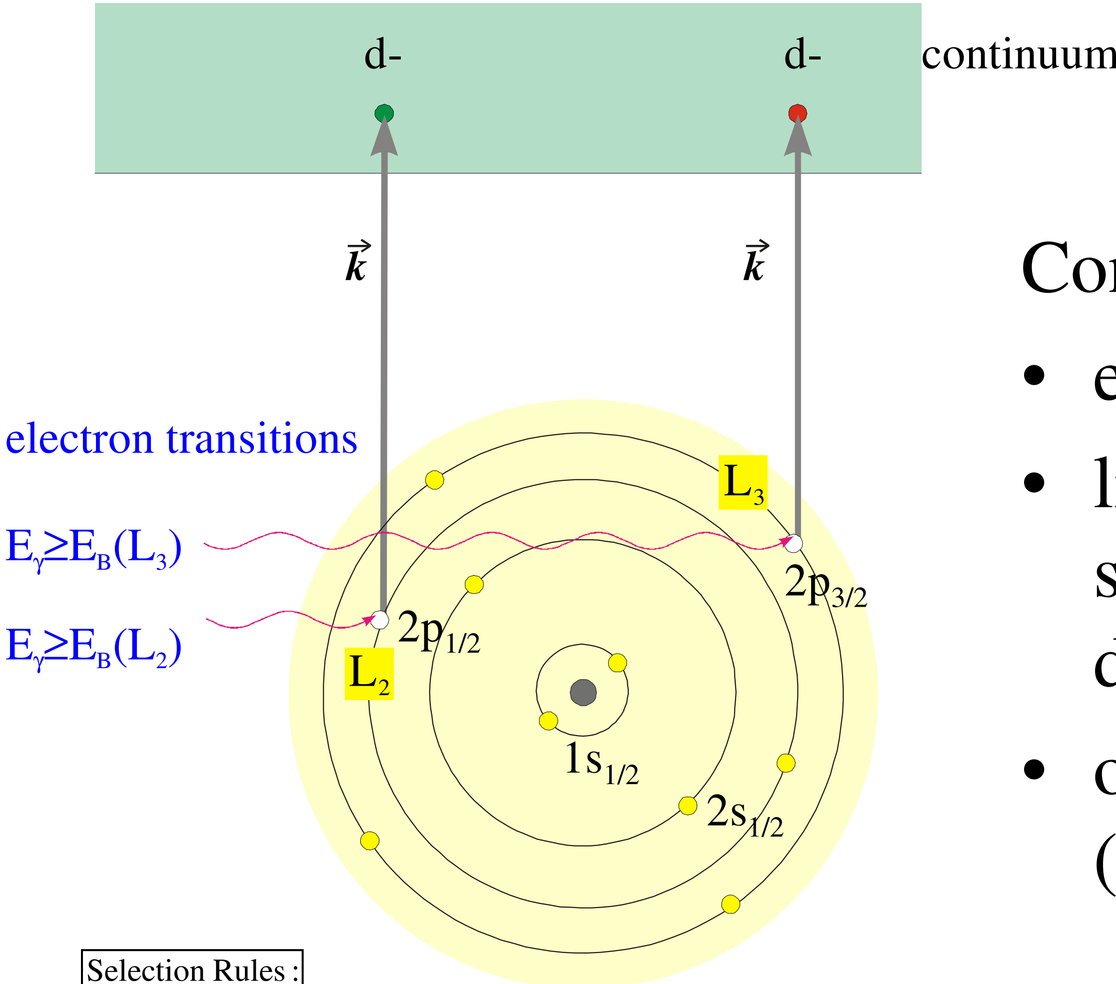
Dichroic signal





dichroic signal $\mu^+ - \mu^- = \Delta\mu$

X-ray Absorption



Selection Rules :

$\Delta l = \pm 1$

$\Delta m_l = \sigma = 0, \pm 1$

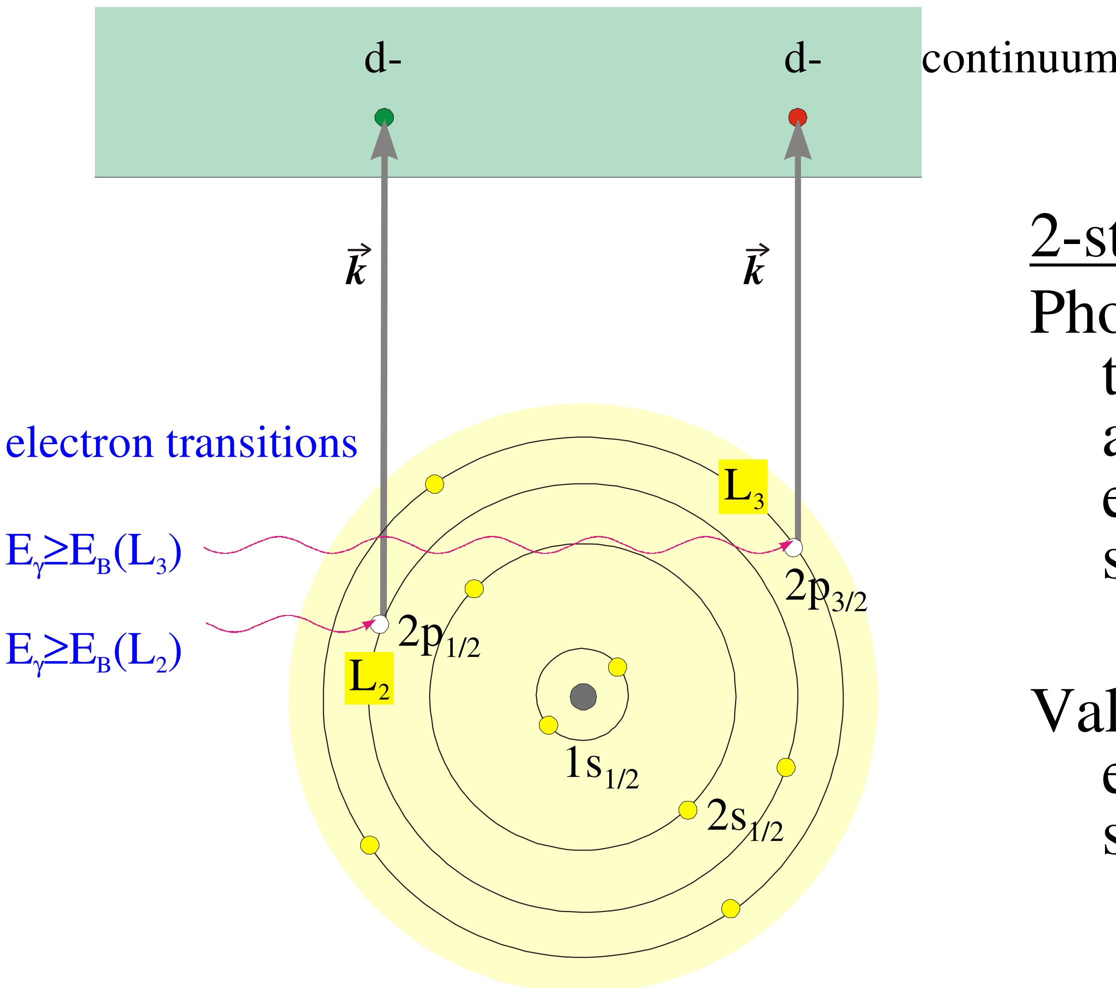
$\Delta s = 0$

$\Delta m_s = 0$

Conservation laws

- energy $E_{ph} = E_f - E_i$
- linear momentum (for small e-energies) in direction of E-vector
- orbital momentum (symmetry)

Absorption of Circularly Polarized X-rays



<u>2-step description:</u>

Photon "polarizes" electron through the transfer of angular momentum to electron spin through spin-orbit coupling

Valence band only takes electrons of appropriate spin

Origin of Spin and Orbital Polarisation

L_2 absorption of a right pol. photon

d continuum $|m_l m_s>$: $|2, \downarrow>$ $|0, \uparrow>$ $|1, \uparrow>$ $|1, \downarrow>$

$\Delta m_l = +1$

$\Delta m_s = 0$

$p_{1/2}$

$m_j = +1/2$ $\quad 2/3|\, m_l=1, \downarrow>$ $\quad 1/3|\, m_l=0, \uparrow>$

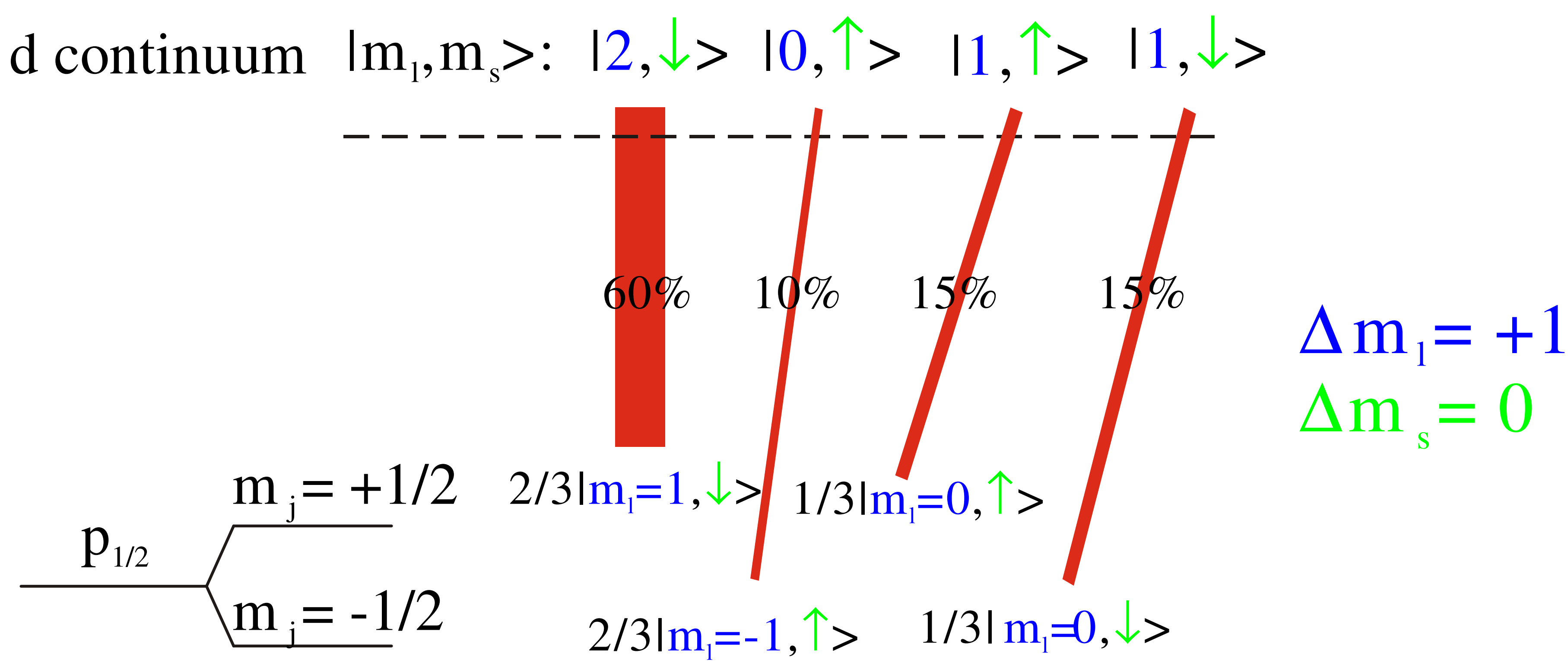
$m_j = -1/2$ $\quad 2/3|\, m_l=-1, \uparrow>$ $\quad 1/3|\, m_l=0, \downarrow>$

Not all transitions are allowed, considering the allowed transitions, we calculate average spin and angular momentum of the excited electrons

	L_2	L_3
$\langle \sigma_z \rangle$	-1/2	+1/4
$\langle l_z \rangle$	+3/4	+3/4

Transition Probabilities

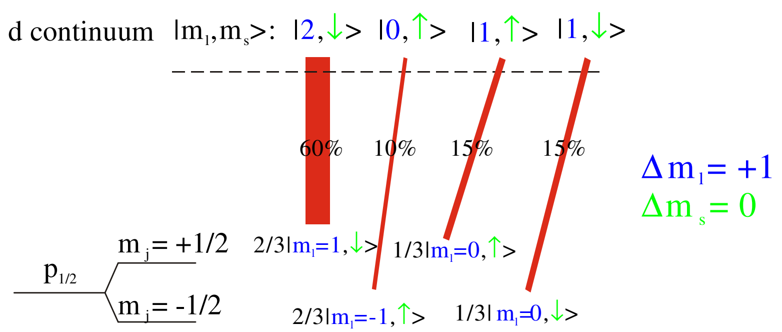
L_2 absorption of a right pol. photon



Spin and Orbital Polarizations

$\langle\sigma_z\rangle$ for $p_{1/2}$ to d	Initial State	(Clebsh-Gordon)^2	Transition Probability	Relative Transition Probability	Spin up or down (+ or -)	Weighted Spin
	ml=1, spin down	2/3	60%	1	-	-2/3
	ml=0, spin up	1/3	15%	½	+	1/6
	ml=1, spin up	2/3	10%	1/6	+	1/9
	ml=0, spin down	1/3	15%	½	-	-1/6

L_2 absorption of a right pol. photon



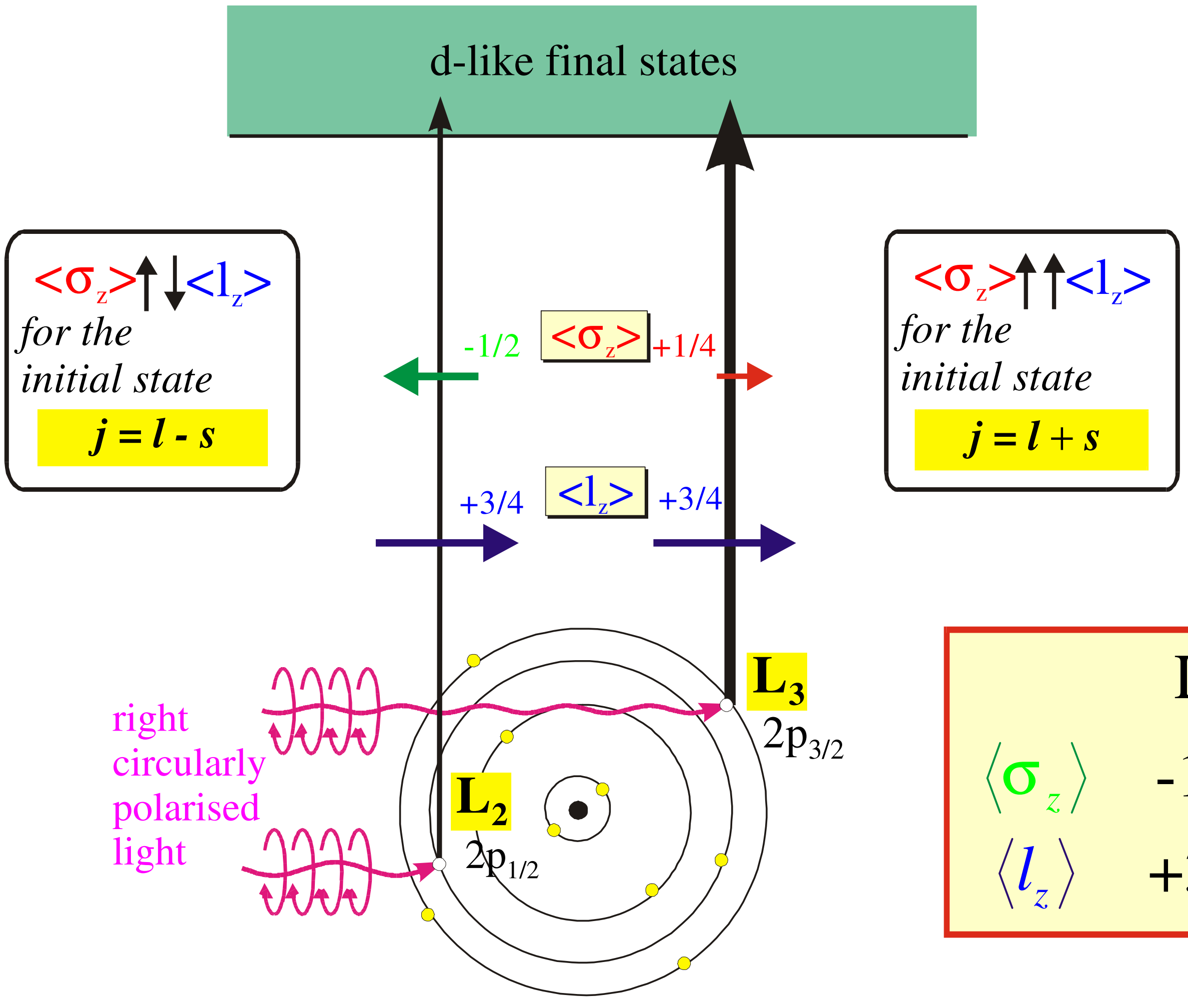
$\langle\sigma_z\rangle$=average spin

$= (-2/3+1/6+ 1/9-1/6)/ (2/3+1/6+ 1/9+1/6)$

$=(-5/9) \; / \; (10/9)$

$=-1/2$

Spin-orbit coupling



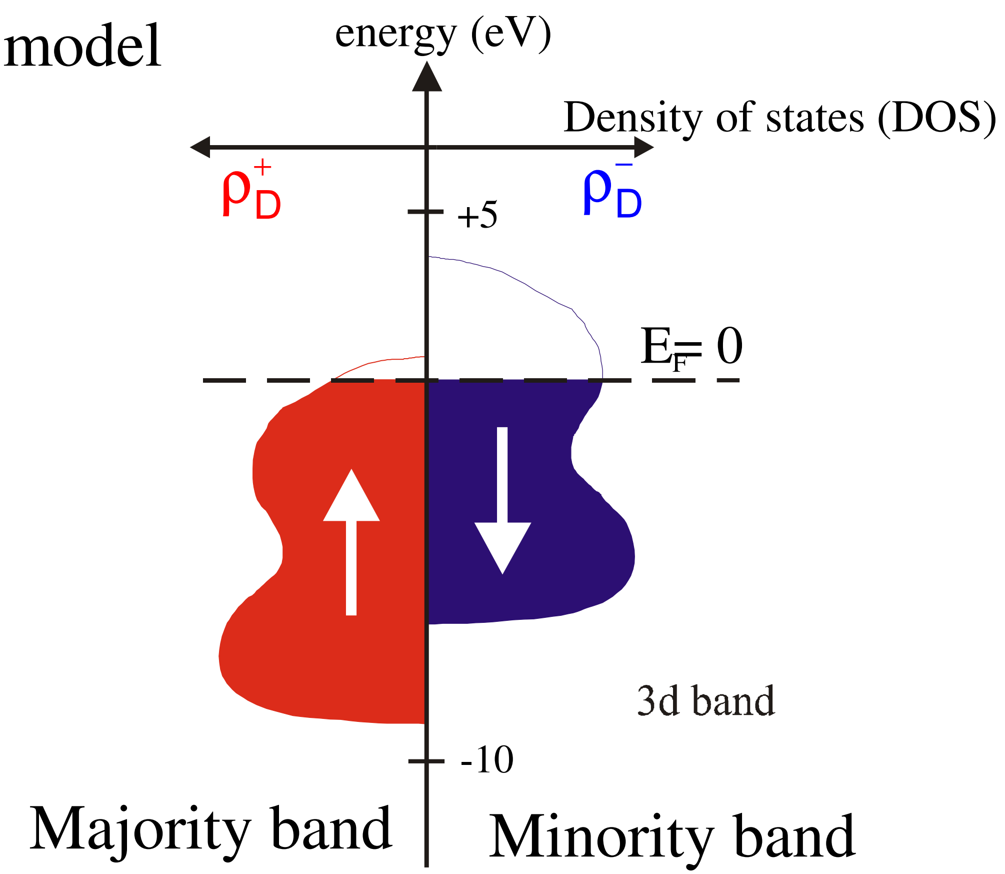
	L_2	L_3
$\langle \sigma_z \rangle$	-1/2	+1/4
$\langle l_z \rangle$	+3/4	+3/4

Photoelectron achieves Spin and Orbital polarization $<\sigma_z>$, $<l_z>$ in photon propagation direction z.

Ferromagnetic state of a 3d transition metal

Stoner model

Net magnetic moment in material caused by exchange splitting



The "hole-moment"

Splitting of "spin-up" and "spin-down" bands at the Fermi level

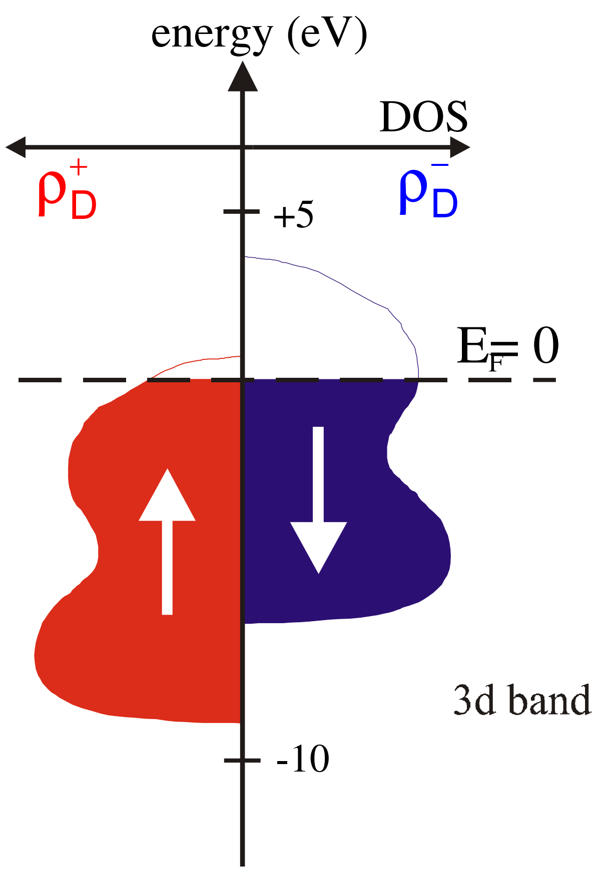
↓

Formation of a magnetic Spin moment

$$m_s / \mu_B = \int_{\sim -10\ \text{eV}}^{0} (\rho^{+} - \rho^{-})(E)\, dE$$

Define a "hole"-moment...

$$m_s / \mu_B^{\ unocc.} = \int_{0}^{\sim 5\ \text{eV}} (\rho^{+} - \rho^{-})(E)\, dE = - m_s / \mu_B^{\ occ.}$$

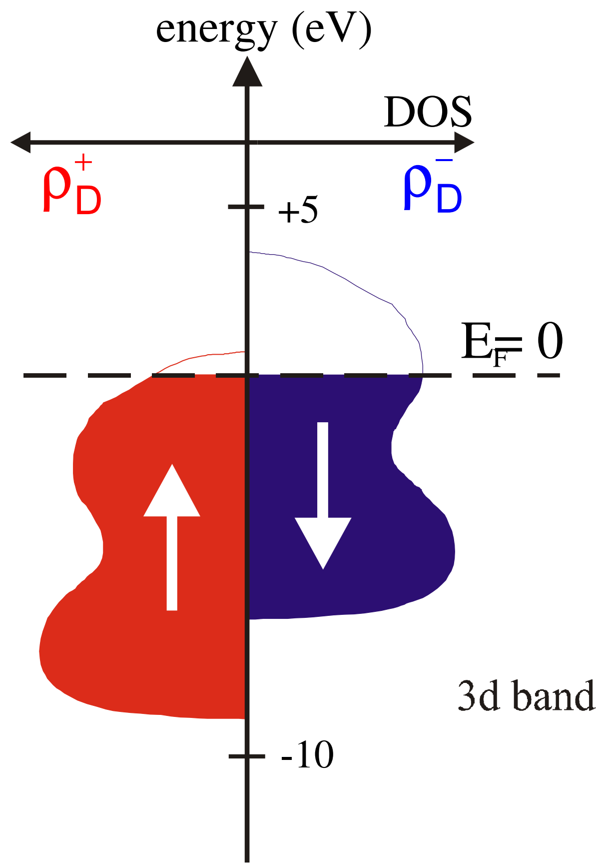


What does XMCD probe?

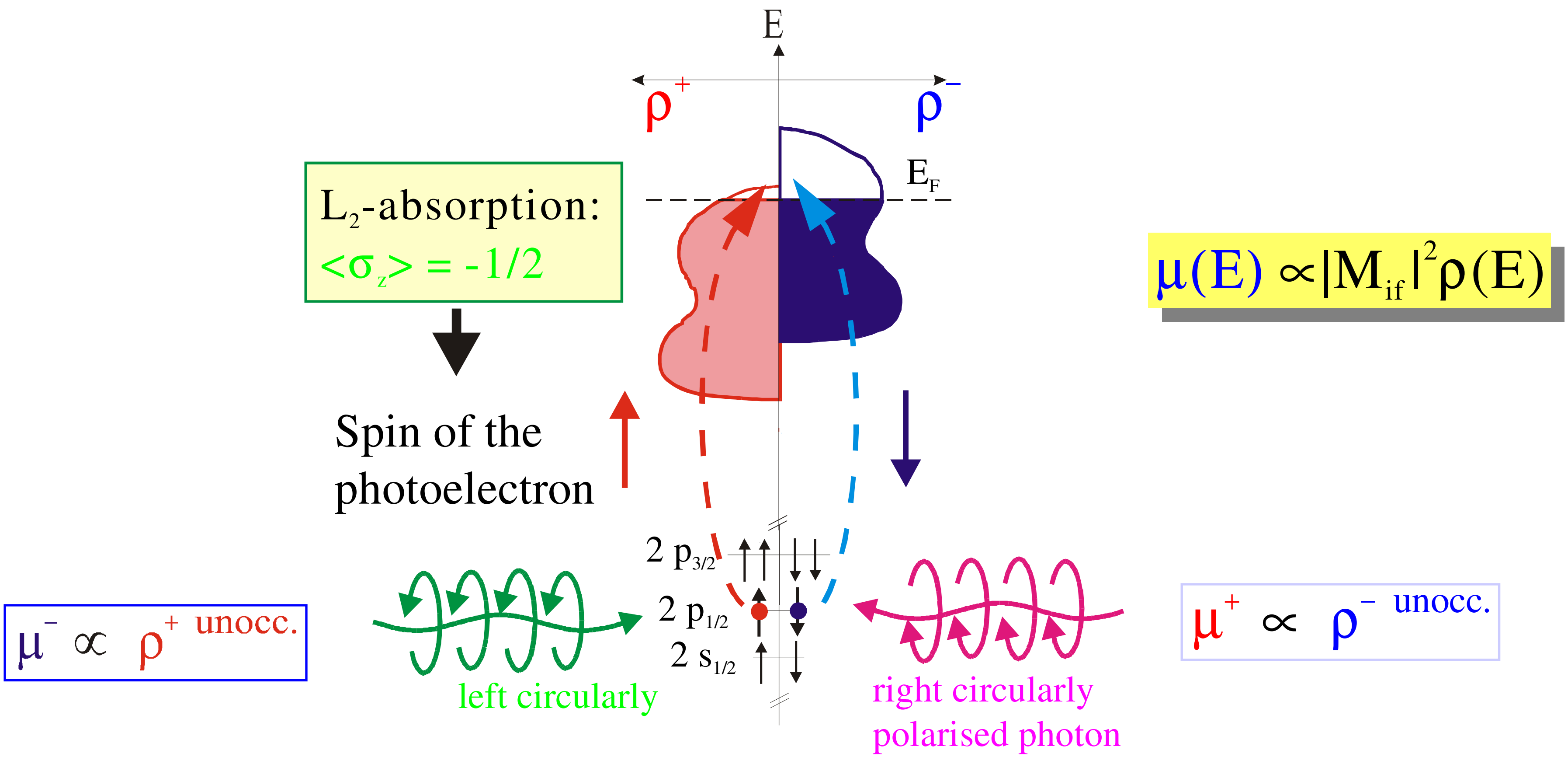
Absorption probes the density of valence states

$$m_s / \mu_B^{unocc.} = \int_0^{\sim 5\,eV} (\rho^+ - \rho^-)(E)\, dE = -\, m_s / \mu_B^{occ.}$$

…or the “hole moment”
→ Electron moments



Fermi´s Golden Rule

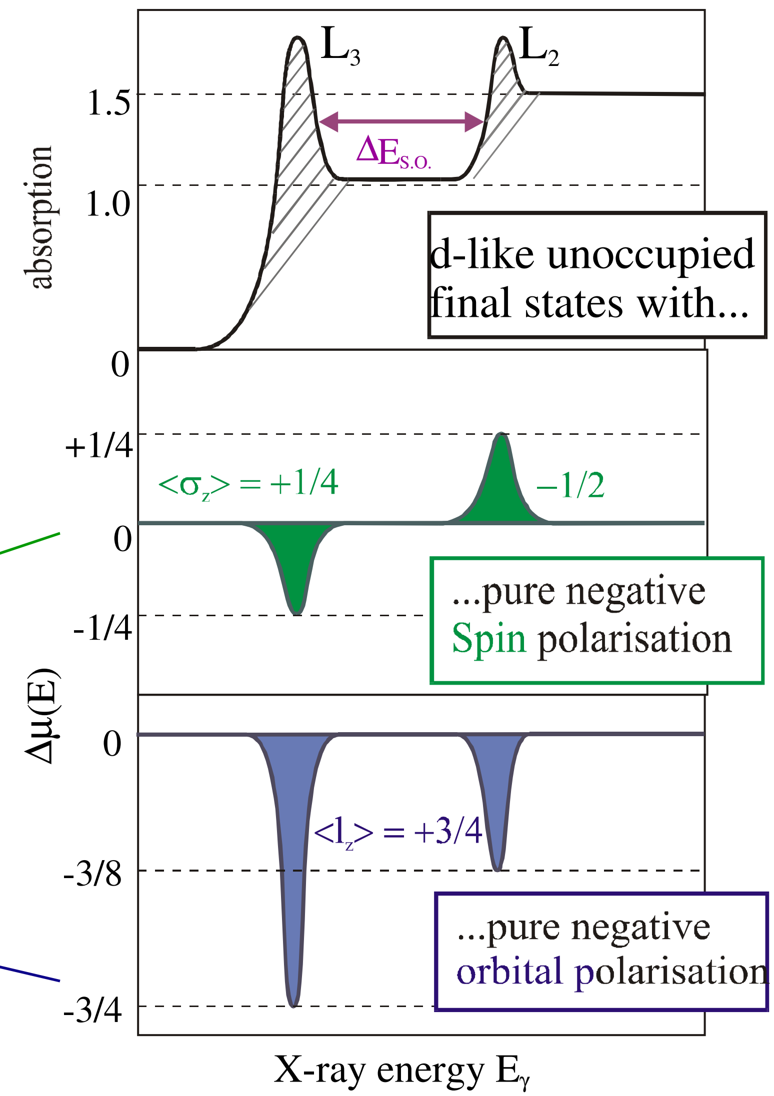


$$Transition\ \ probability = T_{if} \propto \left|\left\langle f \left| H_{\text{int}} \right| i \right\rangle\right|^2 \delta(Ei - Ef)\rho(Ef)$$

The sum rules of XMCD

difference spectra
$\Delta\mu = \mu^+ - \mu^-$
schematically

Sum rules:
Fit the experimental spectrum by a weighted addition of both contributions to obtain spin and orbital moments



Orbital and Spin Moments

Orbital Magnetic Moment :

$$\left\langle m_o^z \right\rangle = \frac{-\mu_B}{\hbar}\left\langle l_z \right\rangle$$

Magnetic moment from motion of electron in orbit

Spin Magnetic Moment :

$$\left\langle m_s^z \right\rangle = \frac{-2\mu_B}{\hbar}\left\langle s_z \right\rangle$$

Intrinsic magnetic moment of the electron

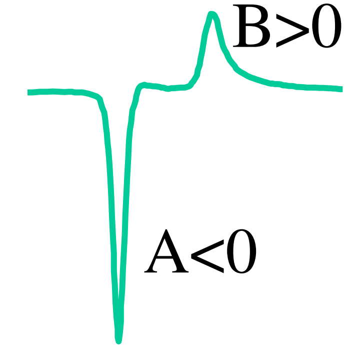
where :

$$BohrMagneton: \quad \mu_B = \frac{e\mu_o\hbar}{2m} = 1.17\times 10^{-29}\,V\,m\,s$$

$\left\langle l_z \right\rangle, \left\langle s_z \right\rangle$ expectation values of the angular and spin momentum operators

J. Stohr and H. C. Siegmann, Magnetism, From Fundamentals to Nanoscale Dynamics, (Springer-Verlag Berlin Heidelberg, 2006).

Sum rules



From

$$\Delta I_{XMCD} = QR^2 \sum_{states} \left| \left\langle C_{-1}^{(1)} \right\rangle \right|^2 - \left| \left\langle C_{+1}^{(1)} \right\rangle \right|^2$$

$$\Rightarrow SUM\ \ RULES:$$

$$\left\langle -A + 2B \right\rangle = \frac{C}{\mu_B} m_s \qquad \textit{spin sum rule}$$

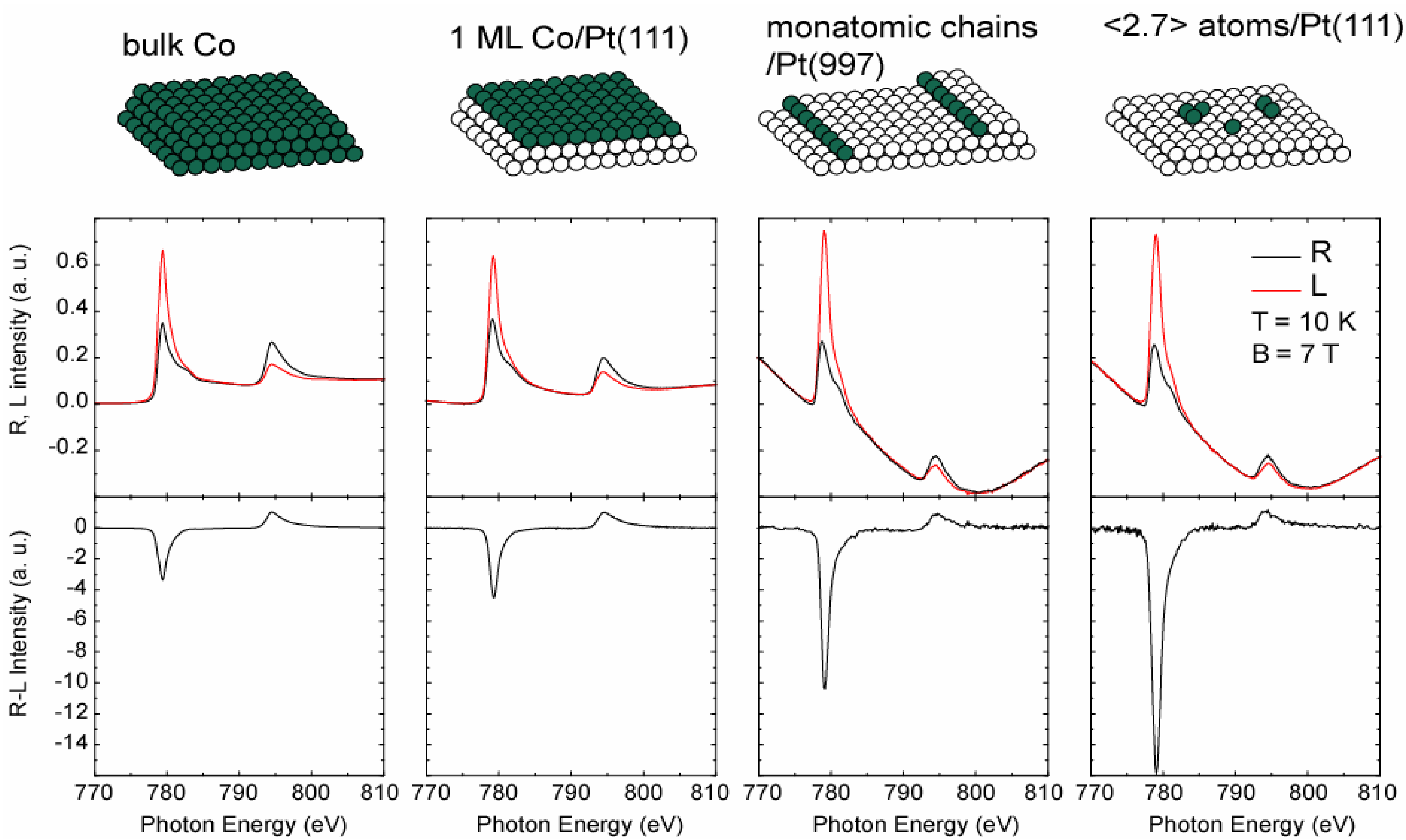
$$\left\langle -A + B \right\rangle = \frac{3C}{2\mu_B} m_o \qquad \textit{orbital moment sum rule}$$

$$where\ C = QR^2 \frac{L}{3(2L+1)}$$

J. Stohr and H. C. Siegmann, Magnetism, From Fundamentals to Nanoscale Dynamics, (Springer-Verlag Berlin Heidelberg, 2006).

XMCD of Co structures: 3D ⇒ 0D

*P. Gambardella et al., NATURE **416**, 301 (2002)*



XMCD

Following Stöhr and Siegmann:

$$\sigma_{abs} = \mathrm{Q}\ \hbar\omega \left|\langle b|\vec{\varepsilon}\cdot\vec{r}|a\rangle\right|^2 \delta(\hbar\omega-(Eb-Ea))\rho(Eb)$$

$\Rightarrow$ int*egrate over energy* $\quad I_{res} = \mathrm{Q}\hbar \quad \left|\langle b|\vec{\varepsilon}\cdot\vec{r}|a\rangle\right|^2$

where $\mathrm{Q} = \text{constant}$

now, using the dipole operators $\mathrm{P}_{\alpha}^{\mathrm{q}} = \vec{\varepsilon}\cdot\vec{r}$

$$I_{res} = \mathrm{Q} \quad \left|\langle b|\mathrm{P}_{\alpha}^{\mathrm{q}}|a\rangle\right|^2$$

J. Stohr and H. C. Siegmann, Magnetism, From Fundamentals to Nanoscale Dynamics, (Springer-Verlag Berlin Heidelberg, 2006).

XMCD

$$I_{res} = \mathrm{Q} \quad \left| \langle b | \mathrm{P}_{\alpha}^{\mathrm{q}} | a \rangle \right|^2$$

$$= \left| \delta(m_s', m_s) \quad \langle R_{n',l}(r) | r | R_{n,c}(r) \rangle \quad \sum_{mc,ml,p} e_{\alpha,p}^{q} \langle l, m_l | C_p^{(1)} | c, m_c \rangle \right|^2$$

spin *radial* *angular*

$$\frac{\mathrm{P}_{\alpha}^{\mathrm{q}}}{r} = \sum_{p=0,\pm 1} e_{\alpha,p}^{q} C_p^{(1)}$$

$$where\ C_m^{(l)} = Racah'\ spherical tensor operators = \sqrt{\frac{4\pi}{2l+1}} Y_{l,m}(\theta, \psi)$$

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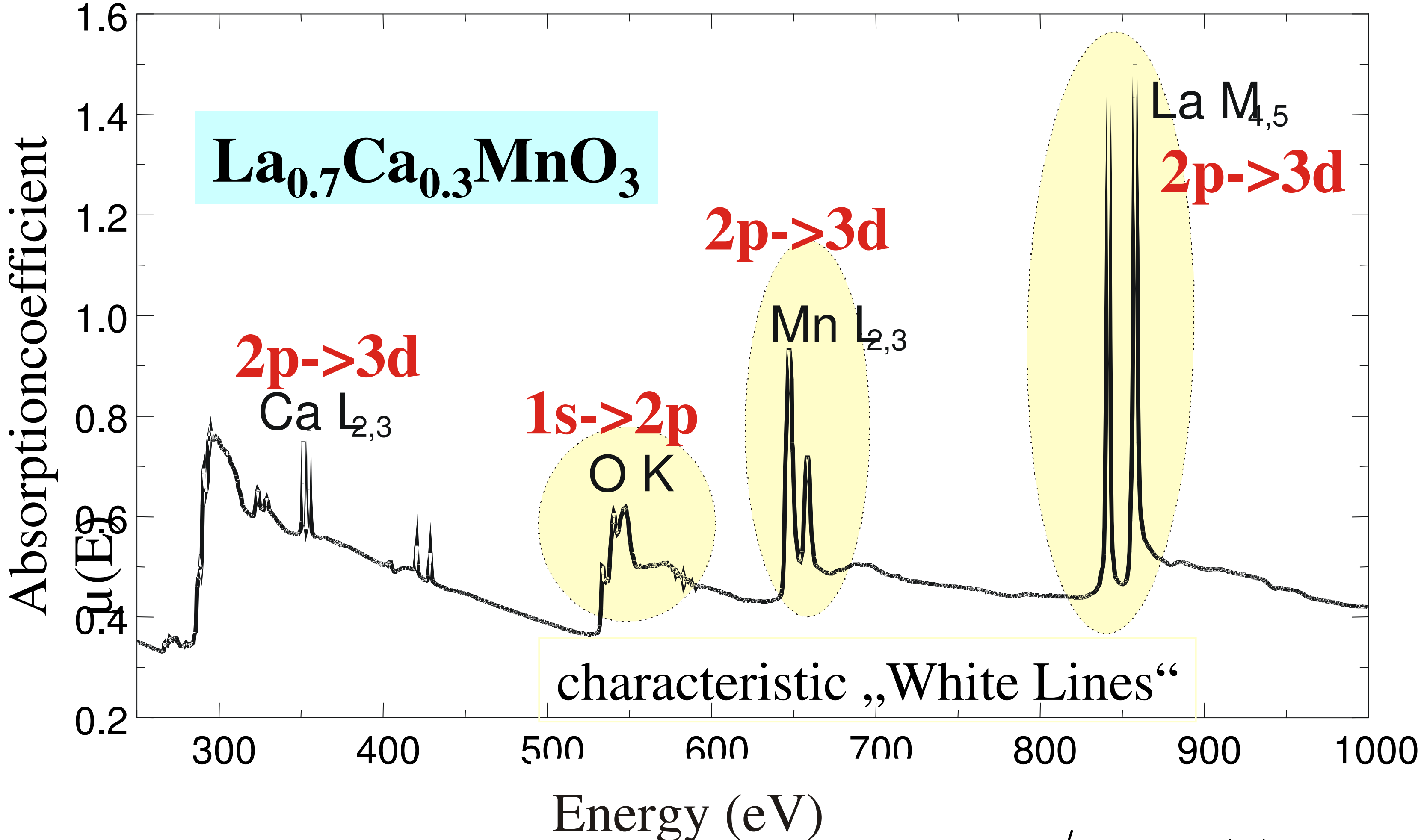
Element Specificity



Element specificity caused by radial term: $\left\langle R_{n',l}(r) \middle| r \middle| R_{n,c}(r) \right\rangle$

Radial component of core levels is highly localized

XMCD difference signal

$$\Delta I \equiv I^{\uparrow\downarrow} - I^{\uparrow\uparrow}$$

$$for\ \ fixed\ magnetization \Rightarrow \Delta I = I^{-} - I^{+}$$

$$\Delta I_{XMCD} = QR^2 \sum_{states} \left|\left\langle C_{-1}^{(1)} \right\rangle\right|^2 - \left|\left\langle C_{+1}^{(1)} \right\rangle\right|^2$$

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References and Acknowledgements

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