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Joint INFM - the Abdus Salam ICTP School on
"Magnetic Properties of Condensed Matter Investigated by Neutron Scattering and Synchrotron Radiation Techniques"

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THE THEORY OF MAGNETIC X-RAY SCATTERING

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These are preliminary lecture notes, intended only for distribution to participants.

THE THEORY OF MAGNETIC X-RAY SCATTERING

M. Blume

The American Physical Society

and

Brookhaven National Laboratory

Introduction - neutrons
and x-rays.

General Theory of x-ray scattering

Big Formula for all phenomena

Bragg scattering (charge and
magnetic)

Correlation functions

Resonant scattering.

HEWLETT PACKARD ↑ ↑ ↑ ↑ ↑ ↑ HEWLETT PACKARD ↑ ↑ ↑ ↑ ↑ ↑ HEWLETT PACKARD ↑ ↑ ↑ ↑ ↑ ↑ HEWLETT PACKARD ↑ ↑ ↑ ↑

	Synchrotron Radiation	Neutrons
Energy	$h\nu = hc/\lambda$	$p^2/2m_0 = h^2/2m_0\lambda^2$
	$=1.44\times10^8(K)/\lambda(Å)$	$= 0.95\times10^3(K)/\lambda^2(Å^2)$
kT/E *($T \doteq 100K$, $\lambda = 1.5Å$)*	$=1.04\times10^{-6}$	$= 0.237$
Interactions	Electromagnetic	Nuclear, Magnetic
Cross section/atom	10^{-6} - 500 barns	≈ 1 barn

Neutron Magnetic Moment

$= -1.91\ eh/4\pi m_0 c$

HEWLETT PACKARD HEWLETT PACKARD HEWLETT PACKARD HEWLETT PACKARD

MAGNETIC PROPERTIES

- Magnetic Structure
- Magnetization Density (orbital and spin)
- Temperature Dependence of Sublattice Magnetization
-
- Magnetic Excitations (crystal fields and spin-waves)
- Magnetic Short Range Order and Critical Scattering (elastic and inelastic)
- Excited and Filled Magnetic Electronic States
- Momentum Density of Magnetic Electrons
- Magnetic Domain Structure

Polarization effects are especially important in magnetic scattering

Magnetic moments are vector quantities

TECHNIQUES FOR THE STUDY OF MAGNETISM

Synchrotron Radiation

Magnetic X-ray Scattering

Resonant Magnetic Scattering

Reflectometry

Magnetic Circular and Linear Dichroism

Faraday Effect

Spin Polarized Photoemission

Magnetic Compton Scattering

Neutrons

Magnetic Neutron Scattering (elastic and inelastic)

Magnetic Diffuse Scattering

Neutron Reflectometry

Polarization Dependent Transmission

"In the 1920's we used to joke that good physicists, once passed to their heavenly rewards, would find apparatus in paradise which, with a twist of some knobs, would give electromagnetic radiation of any desired frequency, intensity, polarization, and direction of propagation."

E. Segrè - "From X-Rays to Quarks"

MAGNETIC X-RAY SCATTERING

BRAGG SCATTERING

X-RAY SCATTERING MEASURES THE FOURIER TRANSFORM OF

$$\rho(r) = \Sigma < \delta(r - r_i) >,$$

the electronic charge density.

NEUTRON SCATTERING MEASURES THE FOURIER TRANSFORM OF

$$S(r) = \Sigma < \underline{s}_i \, \delta(r - r_i) >,$$

the electronic spin magnetization density,

as well as L(r), the orbital magnetization density.

There are smaller terms in the x-ray cross section that depend on S(r) and L(r).

MAGNETIC X-RAY SCATTERING

GELL-MANN and GOLDBERGER; LOW

PLATZMAN and TZOAR

deBERGEVIN and BRUNEL

KRIPLOVICH and ZHIZHIMOV

GIBBS, D'AMICO, MONCTON, BOHR

MB; GIBBS and MB; HANNON, TRAMMELL, MB, GIBBS

NAMIKAWA, ANDO

MCWHAN, VETTIER, ISAACS, HARSHMAN, MILLS

DURBIN

....

SPIN DEPENDENT COMPTON SCATTERING (CIRCULAR POLARIZATION)

GOLDHABER

SAKAI and ONO

COOPER

MILLS

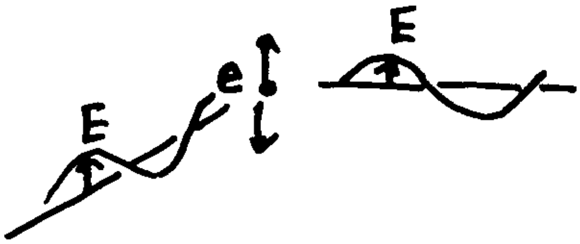
.....

ABSORPTION

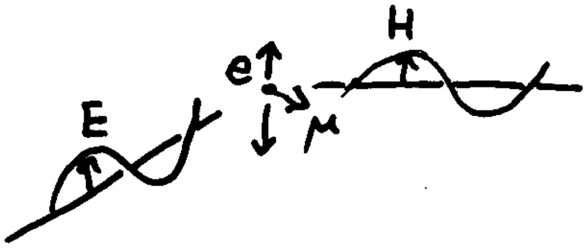
STERN

SCHÜTZ

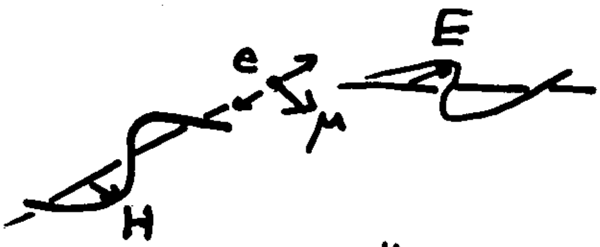
.....



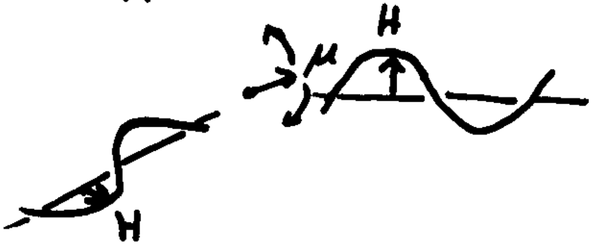
Electric Dipole -
Electric Dipole
(Charge Scattering)



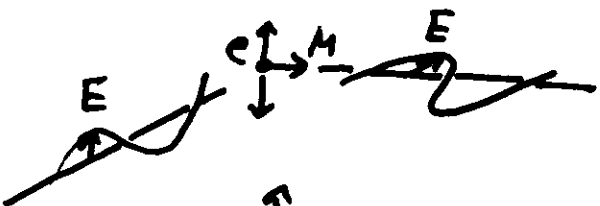
Electric Dipole -
Magnetic Quadrupole



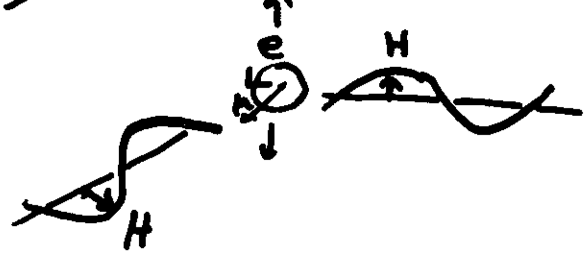
Magnetic Quadrupole -
Electric Dipole



Magnetic Dipole -
Magnetic Dipole



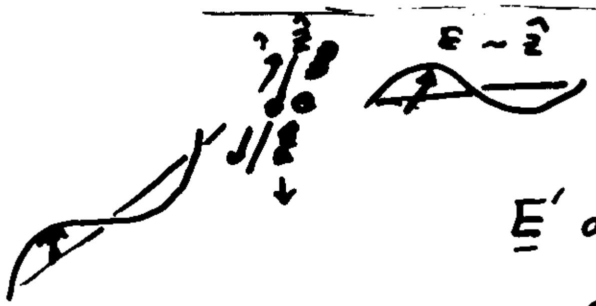
Electric Dipole -
Electric Dipole
(Moving magnetic dipole)



Lorentz Force
(Moving Mag. dipole -
orbital scattering)

(After de Bergevin and Brunel)

Bound Electrons



electric dipole
- electric dipole.

$\underline{E}'$ along $\hat{z}$

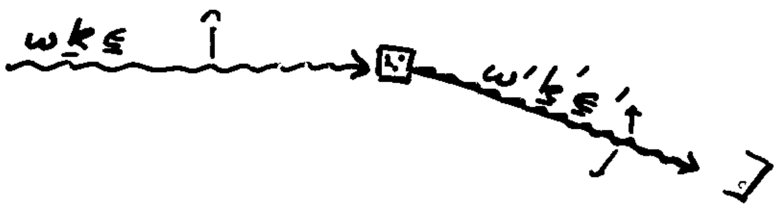
$$E' \sim \hat{z}\,\frac{e^{i\underline{k}'\cdot\underline{r}-\omega' t}}{\omega-\omega_r+i\frac{\Gamma}{2}}\,(\underline{\epsilon}\cdot\hat{z})$$

$$\longrightarrow (\epsilon'\cdot\hat{z})(\epsilon\cdot\hat{z})$$

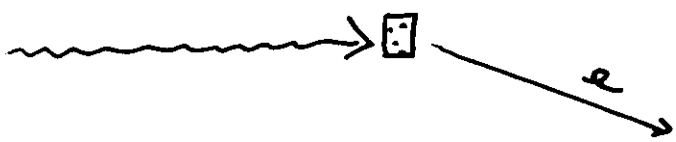
Interactions of X-Rays with Matter

Absorption

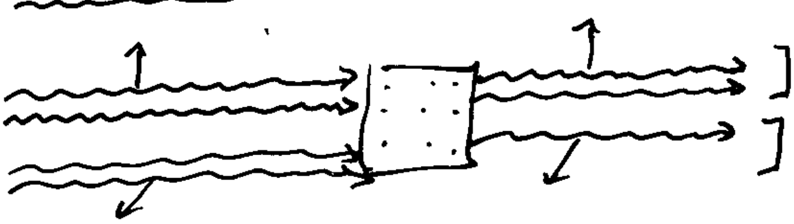
Scattering (Fluorescence)



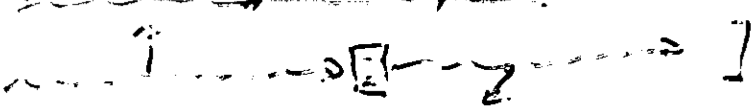
Photoeffect



Dichroism:



Faraday; Kerr; etc.



We use the Golden Rule to calculate the scattering cross-section:

The transition probability per unit time is

$$W = \frac{2\pi}{\hbar} \left| \langle \underline{k}'\lambda'; b \,|\, \mathcal{H}' \,|\, \underline{k}\lambda; a \rangle + \sum_n \frac{\langle \underline{k}'\lambda'; b \,|\, \mathcal{H}' \,|\, n \rangle \langle n \,|\, \mathcal{H}' \,|\, \underline{k}\lambda; a \rangle}{E_a + \hbar\omega_{\underline{k}} - E_n} \right|^2$$

$$\times\, \delta\left(E_b - E_a + \hbar\omega' - \hbar\omega\right).$$

$\mathcal{H}'$ is the interaction between the photons and the electrons in the scatterer.

References:

- M Blume, Magnetic Scattering of X-Rays, J. Appl. Phys. 57, 3615 (1985)
- M. Blume, Magnetic Effects in Anomalous Dispersion, in "Resonant Anomalous X-Ray Scattering," G Materlik, C. J Sparks, and K Fischer, eds., Elsevier (1994) p. 495.

Index of Refraction

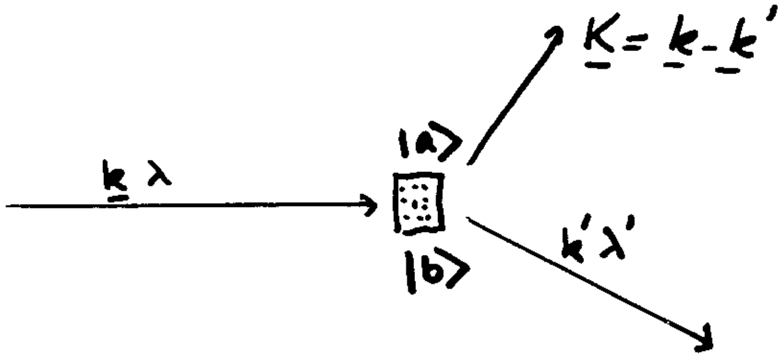
$$n = 1 + \frac{2\pi}{k^2}\frac{1}{V} A(k_0\lambda_1, k_0\lambda_0)$$

"Forward" scattering

Imaginary part gives absorption; real part gives rotation of polarization, etc.

Dichroism

Faraday Rotation



Electrons in Condensed Matter:

$$\mathcal{H}_0 = \frac{1}{2m}\sum_j p_j^2 + \sum_{i\neq j}\varphi(\underline{r}_i,\underline{r}_j) - \frac{e\hbar}{2(mc)^2}\sum_j\left(\underline{E}(\underline{r}_j)\times \underline{p}_j\right)\cdot\underline{S}_j$$

$$\underline{E}(\underline{r}_j) = -\nabla_j\,\varphi.$$

Introducing the Vector Potential $\underline{A}$ of the Electromagnetic Field:

$$\mathcal{H}_0 \longrightarrow \mathcal{H}$$

$$= \frac{1}{2m}\sum_j\left(p_j - \frac{e}{c}\underline{A}(\underline{r}_j)\right)^2 + \sum_{i\neq j}\varphi(\underline{r}_i,\underline{r}_j) - \frac{e\hbar}{2mc}\sum_j \underline{S}_j\cdot\nabla\times A(\underline{r}_j)$$

$$- \frac{e\hbar}{2(mc)^2}\sum_j\left(\underline{E}(\underline{r}_j)\times\left(p_j - \frac{e}{c}A(\underline{r}_j)\right)\right)\cdot\underline{S}_j$$

$$+ \sum_{q\lambda} c^{\dagger}(q\lambda)\,c(q\lambda)\cdot\hbar\omega_q$$

$$E(\underline{r}_j) = -\nabla_j\varphi - \frac{1}{c}\dot{\underline{A}}(\underline{r}_j)$$

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_R + \mathcal{H}'$$

$$\mathcal{H}_0 = \frac{1}{2m}\sum_i p_i^2 + \sum_{i \neq j} \varphi(\underline{r}_i, \underline{r}_j) - \frac{e\hbar}{2(mc^2)^2}\sum_i \left(-\nabla\varphi_i \times p_i\right)$$

$$\mathcal{H}_R = \sum_{q\lambda} \hbar\omega_q \left(c^{\dagger}(q\lambda)\, c(q\lambda)\right)$$

$$\mathcal{H}' = \frac{e^2}{2mc^2}\sum_i A^2(r_i) - \frac{e}{mc}\sum_i \underline{A}(r_i)\cdot p_i$$

$$- \frac{e\hbar}{mc}\sum_i \underline{s}_i \cdot \left(\nabla_i \times \underline{A}(\underline{r}_i)\right)$$

$$- \frac{e^2}{2mc^2}\,\frac{\hbar}{mc^2}\sum_i \underline{s}_i \cdot \left(\dot{\underline{A}}(r_i) \times \underline{A}(\underline{r}_i)\right)$$

$$\underline{A}(\underline{r}_i) = \sum_{q\lambda}\left(\frac{2\pi\hbar c^2}{V\omega_q}\right)^{1/2}\left(\epsilon_{q\lambda} c_{q\lambda} e^{iq\cdot \underline{r}_j} + \epsilon^*_{q\lambda} c^{\dagger}_{q\lambda} e^{-iq\cdot\underline{r}_j}\right)$$

$$\frac{d^2\sigma}{d\Omega\, dE} = W(k\lambda \rightarrow k'\lambda') \times \rho(E_f)$$

density of states

$$\frac{d^2\sigma}{d\Omega_1 d\omega_1} = \left(\frac{e^2}{mc^2}\right)^2 \frac{\omega_1}{\omega_0} \Big| \epsilon_1^{\alpha *} \epsilon_0^{\beta} \Big\{ \langle b | \sum_j e^{i\underline{K}\cdot\underline{r}_j} | a \rangle \delta^{\alpha\beta}$$

$$- i \frac{\hbar \bar{\omega}}{mc^2} \langle b | \sum_j e^{i\underline{K}\cdot\underline{r}_j} s_j^{\gamma} | a \rangle \varepsilon^{\alpha\beta\gamma}$$

$$+ \frac{1}{m} \sum_c \frac{\langle b | O^{\alpha +}(k_1) | c \rangle \langle c | O^{\beta}(k_0) | a \rangle}{E_a - E_c + \hbar\omega_0 + i \frac{\hbar \Gamma_c}{2}}$$

$$+ \frac{1}{m} \sum_c \frac{\langle b | O^{\beta}(k_0) | c \rangle \langle c | O^{\alpha +}(k_1) | a \rangle}{E_b - E_c - \hbar\omega_0 + i \frac{\hbar \Gamma}{2}} \Big|^2$$

$$\times \delta\left(\frac{E_b - E_a}{\hbar} + \omega_1 - \omega_0\right)$$

$$\bar{\omega} = \frac{\omega_1 + \omega_2}{2}$$

$$O^{\beta}(\underline{k}_0) = \sum_i e^{i\underline{k}_0\cdot\underline{r}_i} \left(p_i^{\beta} + i\,(k_0 \times \underline{s}_i)^{\beta}\right)$$

$$\equiv \frac{m}{e} \underline{j}^{\beta}(\underline{k}_0)$$ [current density]

$$\sum_j e^{i\underline{K}\cdot\underline{r}_j} \equiv \frac{1}{e} \rho(\underline{K})$$ [charge density]

$$\sum_j e^{i\underline{K}\cdot\underline{r}_j} \underline{s}_j \equiv \frac{mc}{e} M_s(\underline{K})$$ [spin magnetization density]

Using the Golden Rule, we obtain for the <u>amplitude</u> of Scattering

$$- \frac{e^2}{mc^2} \epsilon_{\lambda'}^{\prime\alpha *} \epsilon_{\lambda}^{\beta} \Big\{ \langle b | \sum_i e^{i\underline{K}\cdot\underline{r}_i} | a \rangle$$

$$- i \varepsilon^{\alpha\beta\gamma} \frac{\hbar\omega}{mc^2} \langle b | \sum_i s_i^{\alpha} e^{i\underline{K}\cdot\underline{r}_i} | a \rangle$$

$$+ \frac{1}{m} \sum_c \frac{\langle b | O^{\alpha\dagger}(\underline{k}') | c \rangle \langle c | O^{\beta}(\underline{k}) | a \rangle}{E_a - E_c + \hbar\omega - i\frac{\Gamma}{2}}$$

$$+ \frac{1}{m} \sum_c \frac{\langle b | O^{\beta}(\underline{k}) | c \rangle \langle c | O^{\alpha\dagger}(\underline{k}' | a \rangle}{E_a - E_c - \hbar\omega} \Big\}$$

$$O^{\beta}(\underline{k}) = \sum_i e^{i\underline{k}\cdot\underline{r}_i} (p_i^{\beta} - i\hbar (\underline{k} \times \underline{s}_i)^{\beta})$$

$$\frac{1}{E_a - E_c + \hbar\omega} = \left(\frac{1}{E_a - E_c + \hbar\omega} - \frac{1}{\hbar\omega} \right) + \frac{1}{\hbar\omega}$$

$$= -\left(\frac{E_a - E_c}{\hbar\omega} \cdot \frac{1}{E_a - E_c + \hbar\omega} \right) + \frac{1}{\hbar\omega}$$

Combining these terms gives the magic expression for x-ray scattering:

$$\frac{d^2\sigma}{d\Omega' dE'} = \left(\frac{e^2}{mc^2}\right)^2 \frac{k'}{k} \Bigg| \epsilon'^{*\alpha} \epsilon^{\beta} \Bigg\{ \langle b | \sum_j e^{i\underline{K}\cdot\underline{r}_j} | a \rangle \delta^{\alpha\beta}$$

$$- i \frac{\hbar\omega_0}{mc^2} \langle b | \sum_i e^{i\underline{K}\cdot\underline{r}_i} \left(-i \frac{(\underline{K}\times\underline{p}_i)^{\gamma}}{K^2} A^{\alpha\beta\gamma} + s_i^{\gamma} B^{\alpha\beta\gamma} \right) | a \rangle$$

$$+ \frac{1}{m} \sum_c \Bigg(\left(\frac{E_c - E_a}{\hbar\omega_0} \right) \frac{\langle b | O^{+\alpha}(\underline{k}') | c \rangle \langle c | O^{\beta}(\underline{k}) | a \rangle}{E_a - E_c + \hbar\omega_0 + i\Gamma/2}$$

$$+ \left(\frac{E_b - E_c}{\hbar\omega_0} \right) \frac{\langle b | O^{\beta}(\underline{k}) | c \rangle \langle c | O^{\alpha +}(\underline{k}') | a \rangle}{E_b - E_c - \hbar\omega_0 + i\Gamma/2} \Bigg\} \Bigg|^2$$

$$\times \; \delta(E_b - E_a + \hbar\omega_1' - \hbar\omega_0)$$

$$O^{\beta}(\underline{k}_0) = \sum_i e^{i\underline{k}_0\cdot\underline{r}_i} \left(p_i^{\beta} + i\hbar (\underline{k}_0 \times \underline{s}_i)^{\beta} \right)$$

$A^{\alpha\beta\gamma}$, $B^{\alpha\beta\gamma}$ are <u>polarization</u> factors.

Bragg, Thermal Diffuse (phonon), Compton, Thompson

Orbital magnetic, spin magnetic, spin-dependent Compton

Anomalous Dispersion, Resonant Magnetic (resonant exchange)

Raman, Resonant Raman, Rayleigh

+ "Index of refraction" for elastic scattering

$$n_{\lambda'\lambda} = \delta_{\lambda'\lambda} + \frac{2\pi}{k^2}\frac{1}{V} A(\underline{k}_0\lambda', \underline{k}_0\lambda)$$

A "Dynamical" Scattering effect!

note forward scattering

(gives absorption, dichroism, Faraday rotation, optical activity, Cotton-Mouton ~~effect~~, Kerr ~~effect~~, etc., all as a fn. of initial polarization

$n = n' + in''$:

Example: dichroism

$$D = \frac{e^{-n''_{++}kz} - e^{-n''_{--}kz}}{e^{-n''_{++}kz} + e^{-n''_{--}kz}}$$

Magnetic Bragg Scattering

X-Rays

charge scattering

orbital mag.

spin mag.

$$\frac{d\sigma}{d\Omega} = \left(\frac{e^2}{mc^2}\right)^2 \Bigg| \langle a| \sum_j e^{i\underline{K}\cdot \underline{r}_j} |a\rangle\, \epsilon^*_{\lambda_1}\cdot\epsilon_{\lambda_0}$$

$$- i\,\frac{\hbar\omega_0}{mc^2}\,\langle a|\sum_j e^{i\underline{K}\cdot\underline{r}_j}\left(-i\,\frac{\underline{K}\times\underline{p}_j}{K^2}\cdot\underline{A}_{\lambda_1\lambda_0} + \underline{s}_j\cdot\underline{B}_{\lambda_1\lambda_0}\right)|a\rangle$$

$$+\frac{1}{m}\sum_c\left(\frac{E_c-E_a}{\hbar\omega_0}\right)\frac{\langle a|\sum_i e^{-i\underline{k}_1\cdot\underline{r}_i}\, p_i\cdot\epsilon^*_{\lambda_1}|c\rangle\langle c|\sum_j e^{i\underline{k}_0\cdot\underline{r}_j}(p_j\cdot\epsilon_{\lambda_0})|a\rangle}{E_a-E_c+\hbar\omega_0 - i\frac{\Gamma}{2}}\Bigg|^2$$

resonance: anamalous dispersion and magnetic.

$$\underline{A}_{\lambda_1\lambda_0} = -\frac{K^2}{k_0^2}\,\epsilon^*_{\lambda_1}\times\epsilon_{\lambda_0}\ ;$$

$$\underline{B}_{\lambda_1\lambda_0} = \epsilon^*_{\lambda_1}\times\epsilon_{\lambda_0} + (\hat{k}_1\times\epsilon^*_{\lambda_1})(\hat{k}_1\cdot\epsilon_{\lambda_0}) - (\hat{k}_0\times\epsilon_{\lambda_0})\,\hat{k}_0\cdot\epsilon^*_{\lambda_1}$$
$$- (\hat{k}_1\times\underline{\epsilon}^*_{\lambda_1})\times(\hat{k}_0\times\underline{\epsilon}_{\lambda_0})$$

Neutrons:

orbit

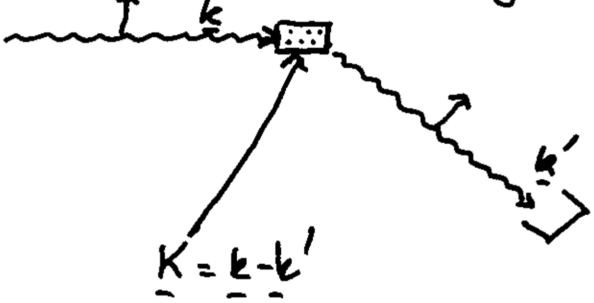
spin

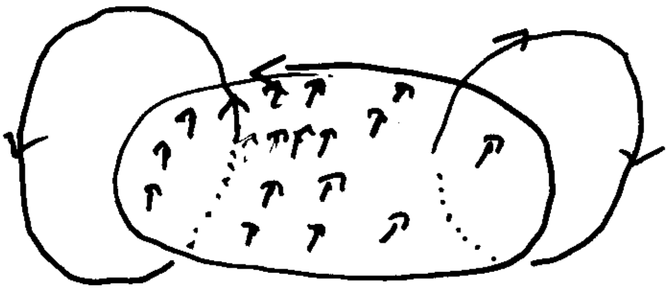
$$\frac{d\sigma}{d\Omega} = \left(\gamma\frac{e^2}{mc^2}\right)^2 \Big|\langle a\lambda_1|\sum_j e^{i\underline{K}\cdot\underline{r}_j}\left(-i\,\frac{\underline{K}\times\underline{p}_j}{K^2} + \underline{s}_j\right)\hat{K}\times(\underline{s}_N\times\hat{K})\,|a\lambda_0\rangle\Big|^2$$

$\underline{s}_N$ = neutron spin

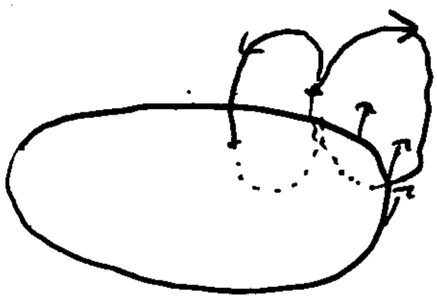
Note! Same polarization factor multiplies orbit and spin for neutrons, but <u>not</u> for x-rays

Scattering



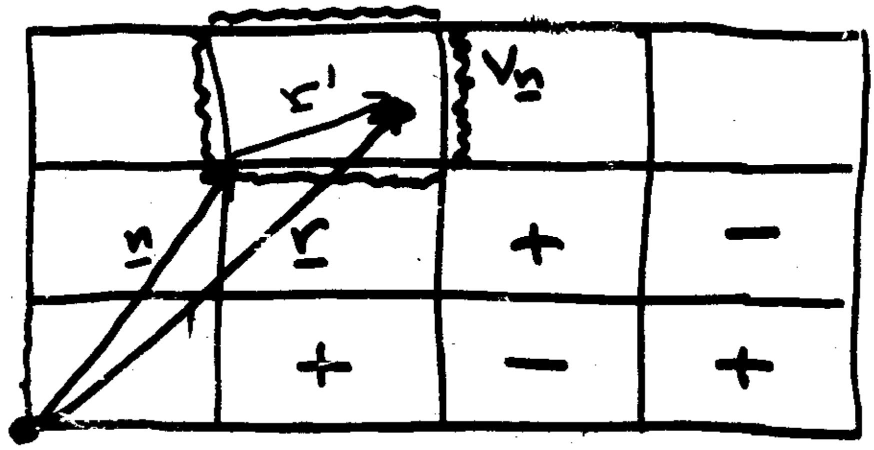


Orbital Magnetization Density



Spin Magnetization Density

Bragg Scattering



$$\langle \rho(\underline{r}+\underline{n})\rangle = \langle \rho(\underline{r})\rangle$$

$$\int_V e^{i\underline{K}\cdot\underline{r}} \langle \rho(\underline{r})\rangle d\underline{r} = \sum_{\underline{n}} \int_{V_{\underline{n}}} e^{i\underline{K}\cdot\underline{r}} \langle \rho(\underline{r})\rangle d\underline{r}$$

$$= \sum_{\underline{n}} \int_{V_{\underline{n}}} e^{i\underline{K}\cdot\underline{n}} e^{i\underline{K}\cdot\underline{r}'} \langle \rho(\underline{r}')\rangle d\underline{r}'$$

$$= \sum_{\underline{n}} e^{i\underline{K}\cdot\underline{n}} F(\underline{K})$$

$$\left[F(\underline{K}) = \int_{V_0} e^{i\underline{K}\cdot\underline{r}'} \langle \rho(\underline{r}')\rangle d\underline{r}' \right]$$

(Structure Factor)

$\sum_{\underline{n}} e^{i\underline{K}\cdot\underline{n}} = 0$ unless $\underline{K}\cdot\underline{n} = 2\pi \times$ integer

[$\underline{K} = 2\pi \times$ reciprocal lattice vector]

How large is (non-resonant) magnetic scattering?

$$\frac{d\sigma}{d\Omega} = \left(\frac{e^2}{mc^2}\right)^2 \left| \rho(\underline{K})\underline{\epsilon}'\cdot\underline{\epsilon} - i\,\frac{\hbar\omega}{mc^2}\left(M_L(\underline{K})\cdot\underline{A} + M_S(\underline{K})\cdot\underline{B}\right)\right|^2$$

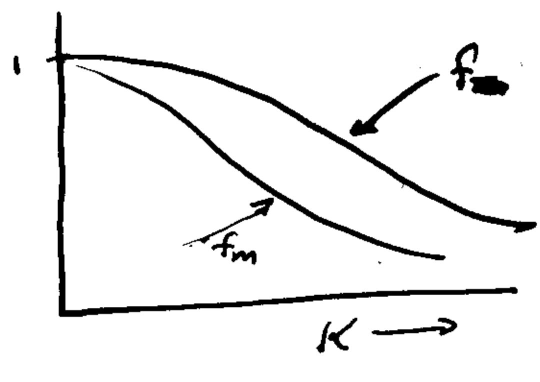
Ratio of scattering probabilities:

$$\frac{\sigma_{mag}}{\sigma_{charge}} \approx \left| \frac{\hbar\omega}{mc^2}\,\frac{M_S(\underline{K})}{\rho(K)} \right|^2$$

$$\approx \left| \frac{\hbar\omega}{mc^2}\,\frac{N_m}{N}\cdot\frac{f_m}{f}\,\langle S\rangle \right|^2$$

For 10 keV x-rays: $\frac{\hbar\omega}{mc^2} \approx .02$

For Iron : $\frac{N_m}{N} \approx \frac{2}{26}$



$$\boxed{\frac{\sigma_{mag}}{\sigma_{charge}} \approx 10^{-6}}$$

Comparison of x-rays and neutrons:

x-rays:

$$\frac{d\sigma}{d\Omega} = \left(\frac{e^2}{mc^2}\right)^2 \left| \rho(\underline{K})\, \underline{\epsilon}' \cdot \underline{\epsilon} - i\frac{\hbar\omega}{mc^2}\left(\underline{M}_L(\underline{K})\cdot\underline{A} + \underline{M}_S(\underline{K})\cdot\underline{B}\right)\right|^2$$

Neutrons

$$\frac{d\sigma}{d\Omega} = \left(\frac{\gamma e^2}{mc^2}\right)^2 \left| \left(\underline{M}_L(\underline{K}) + \underline{M}_S(\underline{K})\right)\cdot \underline{C} \right|^2$$

neutron polarization

Ratio of magnetic x-ray and neutron scattering:

$$\frac{I_0^x\, \sigma_{mag}^x}{I_0^n\, \sigma_{mag}^n} \approx \frac{1}{4}\left(\frac{\hbar\omega}{mc^2}\right)^2 \frac{I_0^x}{I_0^n} \approx 10^{-4}\,\frac{I_0^x}{I_0^n}$$

"Charge, Spin, and Momentum Densities"

(orbit + spin) (polarization)

Charge, Magnetization, Current, Momentum Densities

WHY AND HOW DO WE MEASURE AND CALCULATE THEM?

$$\rho(\underline{r}) = e\sum_i{}' \delta(\underline{r}-r_i) \qquad e = -|e| < 0 \text{ for electrons}$$

$$\tilde{\rho}(\underline{K}) = \int d\underline{r}\, e^{i\underline{K}\cdot\underline{r}} \rho(\underline{r}) = e\sum_i e^{i\underline{K}\cdot\underline{r}_i}$$

$$(\langle\rho(r)\rangle \to e|\psi(r)|^2)$$

$$\underline{M}_s(\underline{r}) = \frac{e\hbar}{mc}\sum_i \underline{s}_i\,\delta(\underline{r}-\underline{r}_i)$$

$$\tilde{\underline{M}}_s(K) = \frac{e\hbar}{mc}\sum_i \underline{s}_i\, e^{i\underline{K}\cdot\underline{r}_i}$$

$$\underline{j}(\underline{r}) = \frac{e}{m}\sum_i \underline{p}_i\,\delta(\underline{r}-\underline{r}_i) + c\,\nabla\times\underline{M}_s(\underline{r})$$

$$\underline{j}(\underline{r}) = \dot{\underline{P}}(\underline{r}) + c\,\nabla\times\left(\underline{M}_L(\underline{r}) + \underline{M}_s(\underline{r})\right)$$

"Polarization" orbital Magnetization

$$\tilde{\underline{J}}(\underline{K}) = \tilde{\dot{\underline{P}}}(\underline{K}) - ic\,\underline{K}\times\left(\tilde{\underline{M}}_L(\underline{K}) + \tilde{\underline{M}}_s(\underline{K})\right)$$

$$f(\underline{p}) = \sum_i \delta(\underline{p}-\underline{p}_i); \quad \text{(momentum density)}$$

$$\underline{f}_s(\underline{p}) = \sum_i \underline{s}_i\,\delta(\underline{p}-\underline{p}_i)$$

Orbital Magnetization and Polarization Densities

$$\frac{e}{m}\sum_i p_i e^{i\underline{K}\cdot\underline{r}_i} = \tilde{\underline{P}}(\underline{K}) - ic\,\underline{K}\times\tilde{\underline{M}}_L(\underline{K})$$

consider:
$$e\frac{d}{dt}\sum_i \underline{r}_i \frac{e^{i\underline{K}\cdot\underline{r}_i}-1}{i\underline{K}\cdot\underline{r}_i} = e\frac{d}{dt}\sum_i \int_0^1 d\lambda\, \underline{r}_i e^{i\underline{K}\cdot\underline{r}_i\lambda}$$

$$= \frac{e}{m}\sum_i \int_0^1 d\lambda \left\{ \underline{p}_i e^{i\underline{K}\cdot\underline{r}_i\lambda} + \lambda \underline{r}_i e^{i\underline{K}\cdot\underline{r}_i\lambda}(i\underline{K}\cdot\underline{p}_i)\right\}$$

integrate by parts

$$= \frac{e}{m}\sum_i \left\{ \lambda p_i e^{i\underline{K}\cdot\underline{r}_i\lambda}\Big|_0^1 + \lambda\underbrace{\left(-p_i(i\underline{K}\cdot\underline{r}_i) + \underline{r}_i(i\underline{K}\cdot\underline{p}_i)\right)}_{+i\underline{K}\times(r_i\times p_i)} e^{i\underline{K}\cdot\underline{r}_i\lambda}\right\}$$

$$= \frac{e}{m}\sum_i \underline{p}_i e^{i\underline{K}\cdot\underline{r}_i} + i\underline{K}\times\int_0^1 d\lambda\, \lambda\, \underline{l}_i e^{i\underline{K}\cdot\underline{r}_i\lambda}$$

$$\frac{e}{m}\sum_i p_i e^{i\underline{K}\cdot r_i} = e\frac{d}{dt}\sum_i \int_0^1 d\lambda\, e^{i\underline{K}\cdot\underline{r}_i\lambda}\underline{r}_i$$
$$- ic\,\underline{K}\times\frac{e}{mc}\int_0^1 d\lambda \sum_i \underline{l}_i e^{i\underline{K}\cdot\underline{r}_i\lambda}\lambda$$

Trammell (1953)

$$\boxed{\begin{aligned}\underline{P}(\underline{r}) &= e\sum_i \underline{r}_i \int_0^1 d\lambda\, \delta(\underline{r}-\underline{r}_i\lambda)\\ \underline{M}_L(\underline{r}) &= \frac{e}{mc}\sum_i \underline{l}_i \int_0^1 d\lambda\, \lambda\, \delta(\underline{r}-\underline{r}_i\lambda)\end{aligned}}$$

classical calculation of $\langle \underline{M}_L(\underline{r}) \rangle$ for a charge moving in a circle:

$$\langle \underline{M}_L(\underline{r}) \rangle = \frac{e}{mc} \left\langle r_j(t) \times p_j(t) \int_0^1 d\lambda\, \lambda\, \delta(r - r_j(t)\lambda) \right\rangle$$

$$r_j(t) \times p_j(t) = m r_0^2 (\hat{e}_x \cos\omega t + \hat{e}_y \sin\omega t) \times (-\hat{e}_x \omega \sin\omega t + \hat{e}_y \omega \cos\omega t)$$

$$= m\omega r_0^2 \hat{e}_z = l_0 \hat{e}_z$$

$$\langle \underline{M}_L(\underline{r}) \rangle = \frac{e}{mc} l_0 \hat{e}_z \int_0^1 d\lambda\, \lambda \left\langle \frac{\delta(r - \lambda r_0)}{\lambda r_0} \delta(z)\, \delta(\vartheta - \omega t) \right\rangle$$

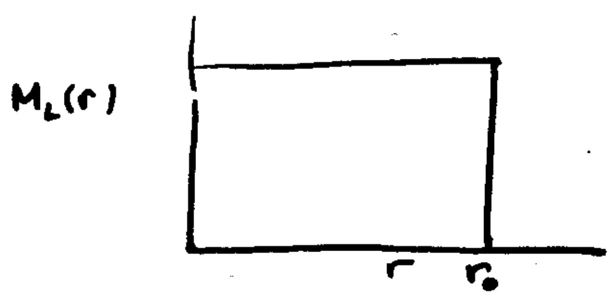
$$\langle \delta(\vartheta - \omega t) \rangle = \frac{\omega}{2\pi} \cdot \int_0^{2\pi/\omega} \delta(\vartheta - \omega t)\, dt = \frac{1}{2\pi}$$

$$\langle \underline{M}_L(\underline{r}) \rangle = \frac{e}{mc} l_0 \hat{e}_z \cdot \frac{1}{2\pi r_0^2} \delta(z)\, \epsilon\left(1 - \frac{r}{r_0}\right)$$

$$\left\langle \int_V M_L(\underline{r})\, d\underline{r} \right\rangle = \frac{e}{2mc} l_0 \hat{e}_z$$

why not

$\underline{M}_L(\underline{r}) \propto \sum_i \underline{l}_i\, \delta(\underline{r} - \underline{r}_i)$?



Charge Density

$$\frac{d\sigma}{d\Omega} = \left(\frac{e^2}{mc^2}\right)^2 \Bigg| \rho(\underline{K})\, \underline{\epsilon}'\cdot\underline{\epsilon}$$

Orbital mag. density

$$- i\,\frac{\hbar\omega}{mc^2}\left(\underline{M}_L(\underline{K})\cdot\underline{A}' + \underline{M}_S\cdot\underline{B}\right)$$

Charge Polarization density

$$+ \sum_c \frac{\underline{\epsilon}'\cdot\underline{P}_{ac}(\underline{k}')\ \underline{\epsilon}\cdot\underline{P}_{ac}(\underline{k})}{\omega - \omega_{ac} - i\frac{\Gamma}{2}} \Bigg|^2$$

Spin mag. density

Resonant scattering; anomalous dispersion and magnetic

$$\tilde{\rho}(\underline{K}) = e\sum_i e^{i\underline{K}\cdot\underline{r}_i}$$

$$= e\sum_i \; 4\pi\sum_{lm} i^l j_l(Kr_i)\, Y^*_{lm}(\hat{r}_i)\, Y_{lm}(\hat{K})$$

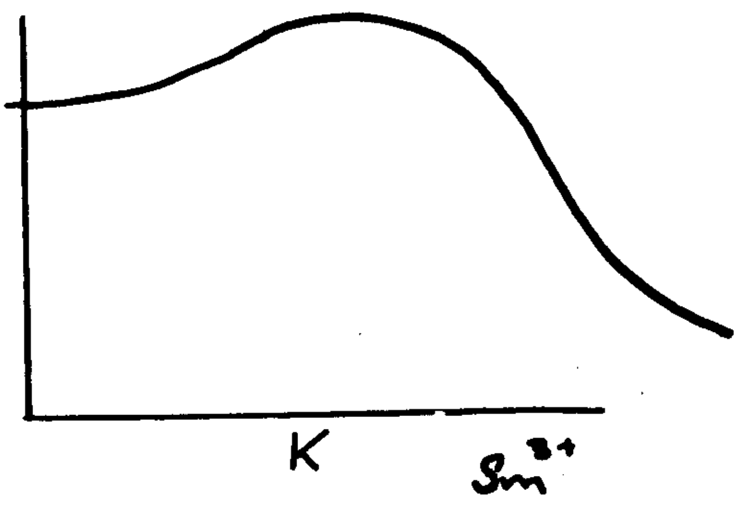
$$\tilde{\underline{M}}_S(\underline{K}) = \frac{e\hbar}{mc}\sum_i \underline{s}_i\, e^{i\underline{K}\cdot\underline{r}_i}$$

$$= \frac{e\hbar}{mc}\sum_i \; 4\pi\sum_{lm} i^l\, \underline{s}_i\, j_l(Kr_i)\, Y^*_{lm}(\hat{r}_i)\, Y_{lm}(\hat{K})$$

$$\tilde{\underline{M}}_L(\underline{K}) = \frac{e}{mc}\sum_i \underline{l}_i \int_0^1 d\lambda\, \lambda\, e^{i\underline{K}\cdot\underline{r}_i\lambda}$$

$$= \frac{e}{mc}\sum_i \; 4\pi\sum_{lm} i^l\, \underline{l}_i \underbrace{\left[\int_0^1 d\lambda\,\lambda\, j_l(Kr_i\lambda)\right]}_{\equiv\, g_l(Kr_i)} Y^*_{lm}(\hat{r}_i)\, Y_{lm}(\hat{K})$$

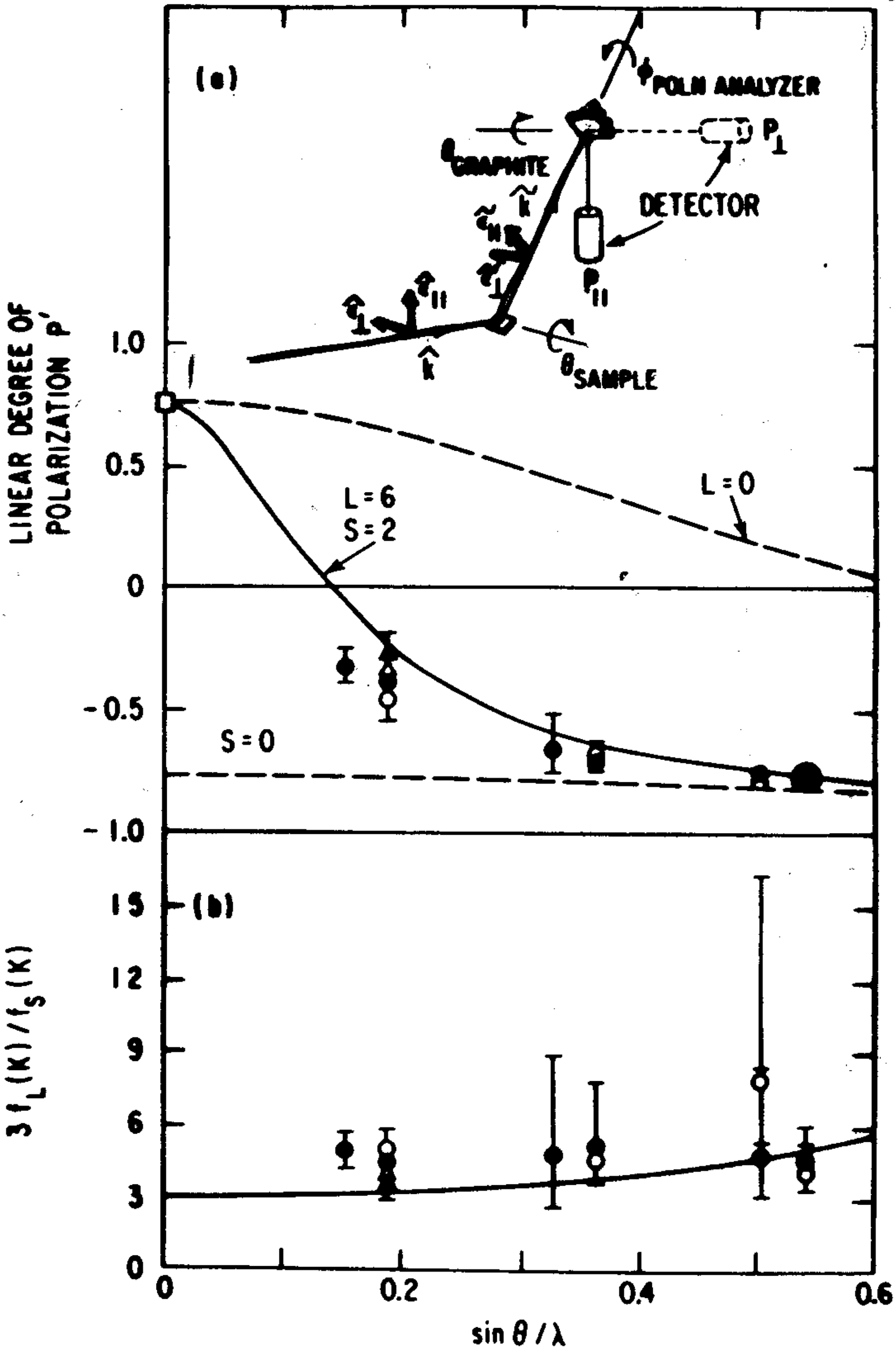
$\tilde{M}(\underline{K})$



K

Sm^{3+}

$\tilde{M}_L(\underline{K})$ and $\tilde{\underline{M}}_S(\underline{K})$ are of opposite sign.



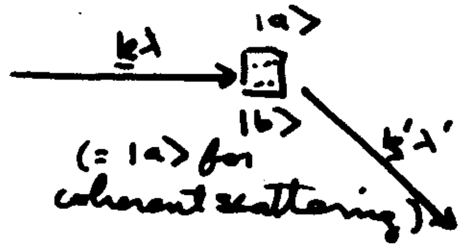
(a)
$\phi_{POLN\ ANALYZER}$
$\theta_{GRAPHITE}$
$P_\perp$
DETECTOR
$P_\parallel$
θ_{SAMPLE}
LINEAR DEGREE OF POLARIZATION P'
L = 6
S = 2
L = 0
S = 0
(b)
$3 f_L(K)/f_S(K)$
$\sin\theta/\lambda$

Charge Density

Best measured by elastic x-ray scattering

$$\frac{d\sigma}{d\Omega} = \left(\frac{e^2}{mc^2}\right)^2 \left|\underline{\epsilon}_{\lambda'}^{\prime *} \cdot \underline{\epsilon}_{\lambda}\right|^2 \left|\left\langle \sum_i e^{i\underline{K}\cdot\underline{r}_i} \right\rangle\right|^2$$

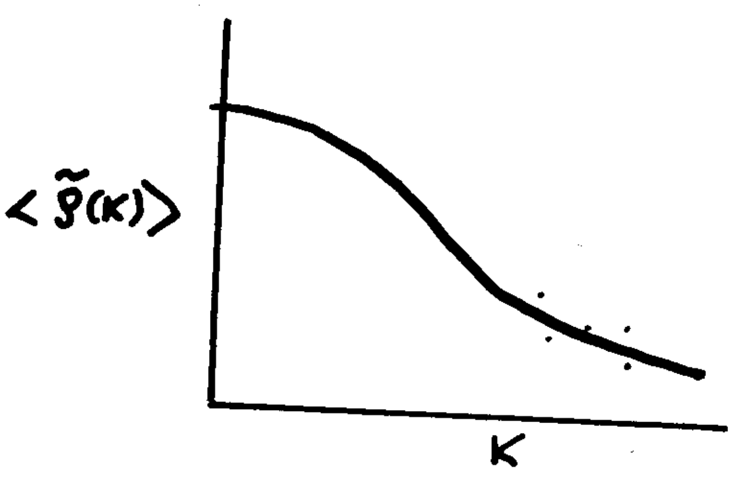
$$\underline{K} = \underline{k} - \underline{k}'$$



$$\langle \cdots \rangle = \sum_a \rho_a \langle a | \cdots | a \rangle$$

$$\frac{d\sigma}{d\Omega} = \left(\frac{e^2}{mc^2}\right)^2 \left|\underline{\epsilon}_{\lambda'}^{\prime *} \cdot \underline{\epsilon}_{\lambda}\right|^2 \left|\langle \tilde{\rho}(\underline{K}) \rangle\right|^2$$

measured by Bragg scattering



Aspects of Charge density:

a) Core electrons unaffected by bonding. Anisotropy of form factor contains this information.

b) $\langle \tilde{\rho}(\underline{K}) \rangle$ decreases with $|K|$:

$$|\langle \tilde{\rho}(\underline{K}) \rangle| = \left| \int e^{i\underline{K}\cdot\underline{r}} \langle \bar{\rho}(\underline{r}) \rangle \, d\underline{r} \right|$$

$$\leq \int |\langle \bar{\rho}(\underline{r}) \rangle| \, d\underline{r} = |\langle \tilde{\rho}(0) \rangle|$$

<u>provided</u> $|\langle \rho(\underline{r}) \rangle| = \langle \rho(r) \rangle$

also <u>provided</u> only electronic + <u>not</u> nuclear charge densities

(This is particularly important for ~~[illegible]~~ magnetization densities)

Observation of Magnetic Scattering

a) Interference with charge scattering

b) Observation of Bragg peak at a point where no charge scattering occurs:!
   spiral structures and antiferromagnets

c) Resonance effects (taking electron binding into account.

Advantages

High resolution of synchrotron experiments

Use of polarization to distinguish orbital, spin, and charge scattering

Elemental specificity thru resonance; surface magnetism.

Correlation Functions

$$\frac{d^2\sigma}{d\Omega_1 d\omega_1} = \left(\frac{e^2}{mc^2}\right)^2 \frac{\omega_1}{\omega_0} \left|\langle b | U | a \rangle\right|^2 \delta\left(\frac{E_b - E_a}{\hbar} + \omega_1 - \omega_0\right)$$

$a \to b$
$\omega_0 \to \omega_1$
$\lambda_0 \to \lambda_1$
$\underline{k}_0 \to \underline{k}_1$

$$U = \frac{1}{e} \rho(\underline{K})' \underline{\epsilon}_1^* \cdot \underline{\epsilon}_0 - \frac{i\hbar\bar{\omega}}{mc^2} \cdot \frac{mc}{e} \underline{M}_s(\underline{K}) \cdot (\underline{\epsilon}_1^* \times \underline{\epsilon}_0)$$

$$+ \frac{m}{e^2} \sum_c \frac{(\epsilon_1^* \cdot \underline{j}^+(\underline{k}_1)) |c\rangle\langle c| (\underline{\epsilon}_0 \cdot \underline{j}(\underline{k}_0))}{E_a - E_c + \hbar\omega_0 + i\frac{\hbar\Gamma}{2}}$$

$$+ \frac{m}{e^2} \sum_c \frac{(\underline{\epsilon}_0 \cdot \underline{j}(\underline{k}_0)) |c\rangle\langle c| (\epsilon_1^* \cdot \underline{j}^+(\underline{k}_1))}{E_b - E_c - \hbar\omega_0 + i\frac{\hbar\Gamma}{2}}$$

$$\delta\left(\frac{E_b - E_a}{\hbar} + \omega_1 - \omega_0\right) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt\, e^{i\frac{E_b - E_a}{\hbar}t + i(\omega_1 - \omega_0)t}$$

$$\frac{d^2\sigma}{d\Omega_1 d\omega_1} = \left(\frac{e^2}{mc^2}\right)^2 \frac{\omega_1}{\omega_0} \langle a | U^+ | b \rangle \langle b | U | a \rangle \delta\left(\frac{E_b - E_a}{\hbar} + \omega_1 - \omega_0\right)$$

Sum over final states $|b\rangle$; average over initial states $|a\rangle$.

$$\frac{d^2\sigma}{d\Omega_1 d\omega_1} = \sum_{ab} p_a \left.\frac{d^2\sigma}{d\Omega_1 d\omega_1}\right|_{\cancel{b \leftarrow} a}$$

$$\frac{d^2\sigma}{d\Omega_1 d\omega_1} = \left(\frac{e^2}{mc^2}\right)^2 \frac{\omega_1}{\omega_0} \sum_{ab} p_a \int_{-\infty}^{\infty} dt \frac{e^{i(\omega_1-\omega_0)t}}{2\pi}$$

$$\times \langle a | U^+ | b \rangle \langle b | \underbrace{e^{\frac{i}{\hbar}(E_b - E_a)t} U | a \rangle}_{= U(t)}$$

$$\frac{d^2\sigma}{d\Omega_1 d\omega_1} = \left(\frac{e^2}{mc^2}\right)^2 \frac{\omega_1}{\omega_0} \cdot \frac{1}{2\pi} \int_{-\infty}^{\infty} dt\, e^{-i(\omega_0-\omega_1)t} \langle U^+ U(t) \rangle$$

where $\langle \cdots \rangle = \sum_a p_a \langle a | \cdots | a \rangle$

Example: $\rho(\underline{K})$ term in U:

$$\frac{d^2\sigma}{d\Omega_1 d\omega_1} = \left(\frac{e^2}{mc^2}\right)^2 \frac{\omega_1}{\omega_0} \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i\omega t} \langle \rho^+(\underline{K})\, \rho(\underline{K}t) \rangle |\epsilon_1^* \cdot \epsilon_0|$$

$\hbar\omega = \hbar(\omega_0 - \omega_1) \equiv$ energy transfer.

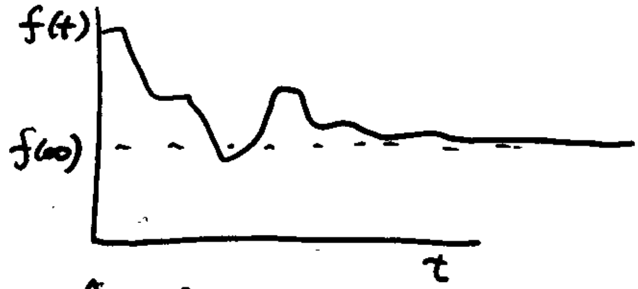
Just like Van Hove correlation fn. for neutron scattering, but with electron density instead of nuclear scattering density!

Separation of "fluctuation" (inelastic) scattering from "coherent" scattering:

$$\frac{d^2\sigma}{d\Omega_1\, d\omega_1} = \left(\frac{e^2}{mc^2}\right)^2 \frac{\omega_1}{\omega_0} \cdot \frac{1}{2\pi}\int_{-\infty}^{\infty} dt\, e^{-i\omega t} \langle U^+(0)\, U(t)\rangle$$

Consider

$$\frac{1}{2\pi}\int_{-\infty}^{\infty} dt\, f(t)\, e^{-i\omega_0 t}$$



write $f(t) \equiv \underbrace{\big(f(t) - f(\infty)\big)}_{\to 0 \text{ as } t \to \infty} + f(\infty)$

$$\frac{1}{2\pi}\int_{-\infty}^{\infty} dt\, f(t)\, e^{-i\omega t} \equiv \frac{1}{2\pi}\int_{-\infty}^{\infty} dt\, e^{-i\omega t}\big(f(t) - f(\infty)\big)$$

$$+ \frac{1}{2\pi} f(\infty) \int_{-\infty}^{\infty} dt\, e^{-i\omega t}$$

$$= \frac{1}{2\pi}\int_{-\infty}^{\infty} dt\, e^{-i\omega t}\big(f(t) - f(\infty)\big) \quad + f(\infty)\,\delta(\omega)$$

"coherent" elastic scattering

For $f(t) = \langle U^+(0)\, U(t)\rangle$,

$$f(\infty) \longrightarrow \langle U^+(0)\, U(\infty)\rangle \longrightarrow \langle U^+\rangle\langle U\rangle$$

$$f(t) - f(\infty) \longrightarrow \langle u^+(0)\, u(t) \rangle - \langle u^+ \rangle \langle u \rangle$$

$$\equiv \langle (u^+ - \langle u^+ \rangle)(u(t) - \langle u \rangle) \rangle$$

$$\equiv \langle \delta u^+(0)\, \delta u(t) \rangle$$

$$\frac{d^2\sigma}{d\Omega_1 d\omega_1} = \left(\frac{e^2}{mc^2}\right)^2 \frac{\omega_1}{\omega_0} \cdot \frac{1}{2\pi} \int_{-\infty}^{\infty} dt\, e^{-i\omega t} \langle \delta u^+(0)\, \delta u(t) \rangle$$

$$+ \left(\frac{e^2}{mc^2}\right)^2 \underset{\to 1}{\frac{\omega_1}{\omega_0}} \cdot |\langle u \rangle|^2 \underbrace{\delta(\omega)}_{\equiv \delta(\omega_1 - \omega_0)}$$

Back to example of $\rho(\underline{K})$ term:

$$\frac{d^2\sigma}{d\Omega_1 d\omega_1} = \left(\frac{e^2}{mc^2}\right)^2 \frac{\omega_1}{\omega_0} \cdot \frac{1}{2\pi e^2} \int_{-\infty}^{\infty} dt\, e^{-i\omega t} \langle \delta\rho^+(\underline{K})\, \delta\rho(\underline{K}, t) \rangle$$

$$|\epsilon_1^* \cdot \epsilon_0|^2$$

$$+ \left(\frac{e^2}{mc^2}\right)^2 \frac{1}{e^2} |\langle \rho^*(\underline{K}) \rangle|^2 |\epsilon_1^* \cdot \epsilon_0|^2 \delta(\omega)$$

Bragg Scattering

Coherent elastic scattering:

$$\langle U\rangle = \frac{1}{e}\langle \rho(K)\rangle\, \epsilon_1^*\cdot\epsilon_0 \;-\; i\,\frac{\hbar\omega_0}{mc^2}\,\frac{mc}{e}\,M_s(\underline{K})\cdot(\epsilon_1^*\times\epsilon_0)$$

$$+\;\frac{1}{me^2\hbar\omega_0}\left\langle [\,j^{\alpha+}(k_1),\, j^{\beta}(k_0)]\right\rangle$$ contributes to mag. scatt

$$-\;\frac{1}{me^2\hbar\omega_0}\int_{-\infty}^{\infty}dt_1\, e^{-i\omega_0 t_1-\frac{\Gamma}{2}|t_1|}$$

$$\left\langle T\left(j^{\alpha+}(k_1)\, j^{\beta}(k_0 t_1)\right)\right\rangle\, \epsilon_1^{\alpha *}\epsilon_0^{\beta}$$

$$\left.\frac{d\sigma}{d\Omega_1}\right|_{coh} = \left(\frac{e^2}{mc^2}\right)^2\,|\langle U\rangle|^2$$

$$\frac{d^2\sigma}{d\Omega_1 d\omega_1} = \left(\frac{e^2}{mc^2}\right)^2 \frac{\omega_1}{\omega_0} \Big| \epsilon_1^{\alpha *}\epsilon_0^{\beta} \Big\{ \langle b| \sum_j e^{i\underline{K}\cdot\underline{r}_j} |a\rangle \delta^{\alpha\beta}$$

$$- i\frac{\hbar\omega}{mc^2} \langle b| \sum_j e^{i\underline{K}\cdot\underline{r}_j} \underline{s}_j^{\gamma} |a\rangle \varepsilon^{\alpha\beta\gamma}$$

$$+ \frac{1}{m}\sum_c \frac{\langle b| O^{\alpha\dagger}(\underline{k}_1)|c\rangle\langle c|O^{\beta}(\underline{k}_0)|a\rangle}{E_a - E_c + \hbar\omega_0 + i\frac{\hbar\Gamma}{2}}$$

$$+ \frac{1}{m}\sum_c \frac{\langle b| O^{\beta}(k_0)|c\rangle\langle c|O^{\alpha\dagger}(k_1)|a\rangle}{E_b - E_c - \hbar\omega_0 + i\frac{\hbar\Gamma}{2}} \Big\} \Big|^2 \delta\left(\frac{E_a - E_b}{\hbar} + \omega_0 - \omega_1\right)$$

Resonant Elastic Scattering

$$\propto \left| \sum_{\underline{n}s} e^{i\underline{K}\cdot\underline{n}} \epsilon_1'^{\alpha *} \epsilon_0^{\beta} \underbrace{\sum_c \frac{\langle a | \underline{R}^{\alpha}_{ns} | c\rangle \langle c | \underline{R}^{\beta}_{ns} | a\rangle}{E_a - E_c + \hbar\omega_0 - i\frac{\Gamma}{2}}}_{\equiv C^{\alpha\beta} \text{ (a tensor)}} \right|^2$$

$$C^{\alpha\beta} = C_0 \delta^{\alpha\beta} + C_a^{\alpha\beta} + C_s^{\alpha\beta}$$ (all have resonance energy denominators)

$$\begin{cases} C_0^{\alpha\beta} = \frac{1}{3}\mathrm{Tr}\, C\, \delta^{\alpha\beta} \\ C_a^{\alpha\beta} = \frac{1}{2}(C^{\alpha\beta} - C^{\beta\alpha}) \\ C_s^{\alpha\beta} = \frac{1}{2}(C^{\alpha\beta} + C^{\beta\alpha}) - \frac{1}{3}\mathrm{Tr}\, C\, \delta^{\alpha\beta} \end{cases}$$

If there is only a magnetic moment,

$$C_0^{\alpha\beta} \propto \delta^{\alpha\beta} \longrightarrow \underline{\epsilon}' \cdot \underline{\epsilon}$$

$$C_a^{\alpha\beta} \propto i\,\varepsilon^{\alpha\beta\gamma} m^{\gamma} \longrightarrow i\,\underline{\epsilon}' \times \underline{\epsilon} \cdot \underline{m}$$

$$C_s^{\alpha\beta} \propto m^{\alpha} m^{\beta} - \frac{1}{3} m^2 \delta^{\alpha\beta} \longrightarrow (\underline{\epsilon}' \cdot \underline{m})(\underline{\epsilon} \cdot \underline{m}) - \frac{1}{3} m^2\, \underline{\epsilon}' \cdot \underline{\epsilon}$$

With no magnetic moment, but uniaxial local symmetry

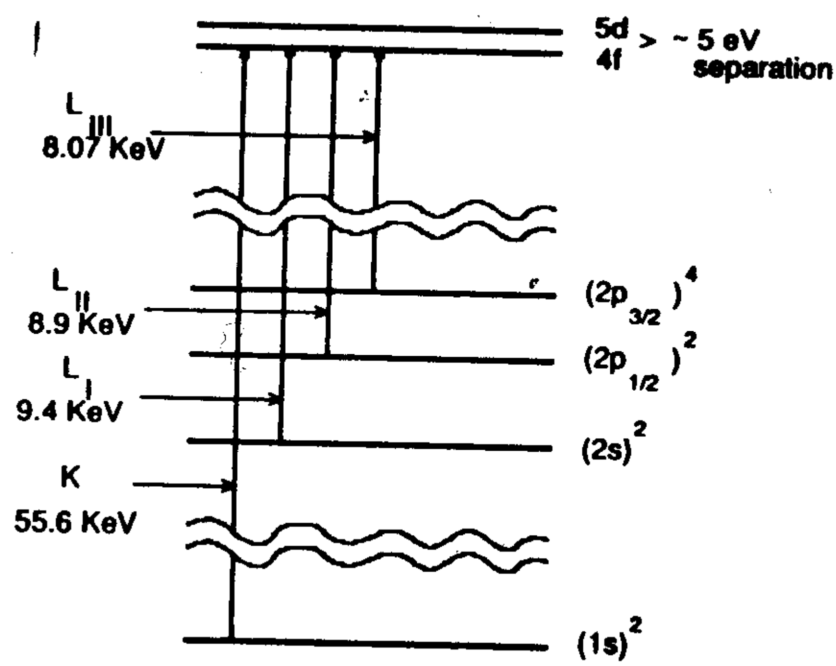
$$C_0^{\alpha\beta} \propto \delta^{\alpha\beta} \longrightarrow \underline{\epsilon}' \cdot \underline{\epsilon}$$

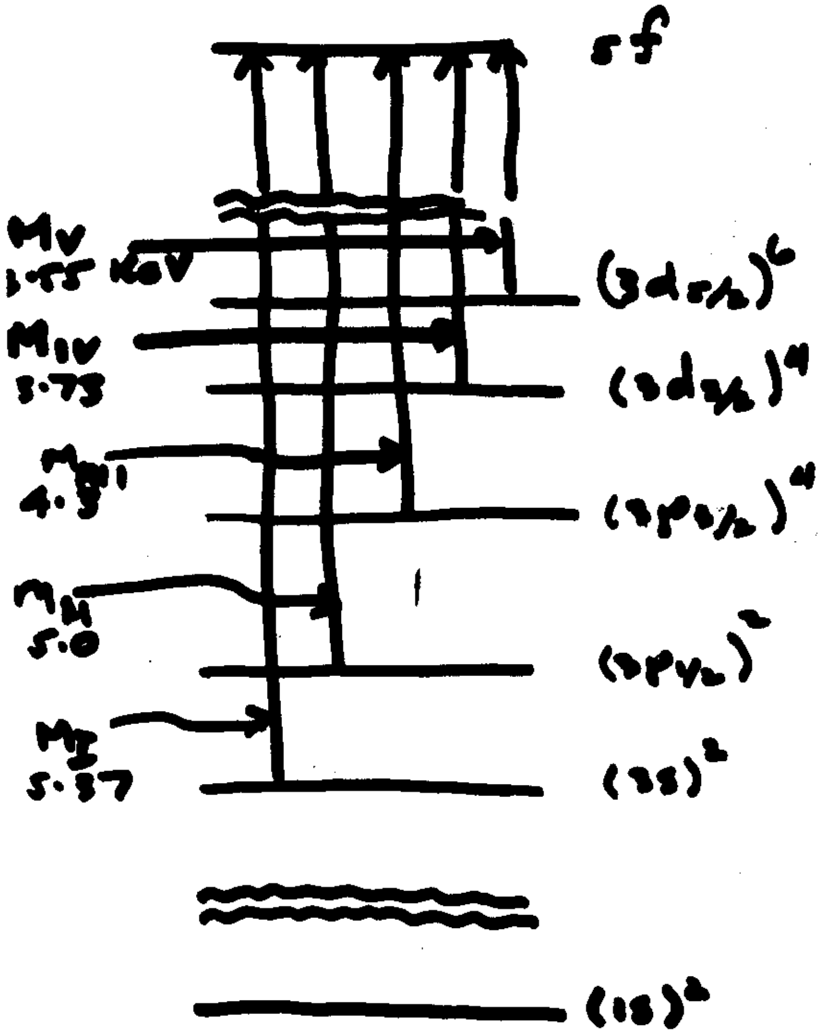
$$C_a^{\alpha\beta} = 0$$ (Templeton scattering)

$$C_s^{\alpha\beta} = \hat{n}^{\alpha}\hat{n}^{\beta} - \frac{1}{3}\delta^{\alpha\beta} \longrightarrow (\underline{\epsilon}' \cdot \hat{n})(\underline{\epsilon} \cdot \hat{n}) - \frac{1}{3}\underline{\epsilon}' \cdot \underline{\epsilon}$$

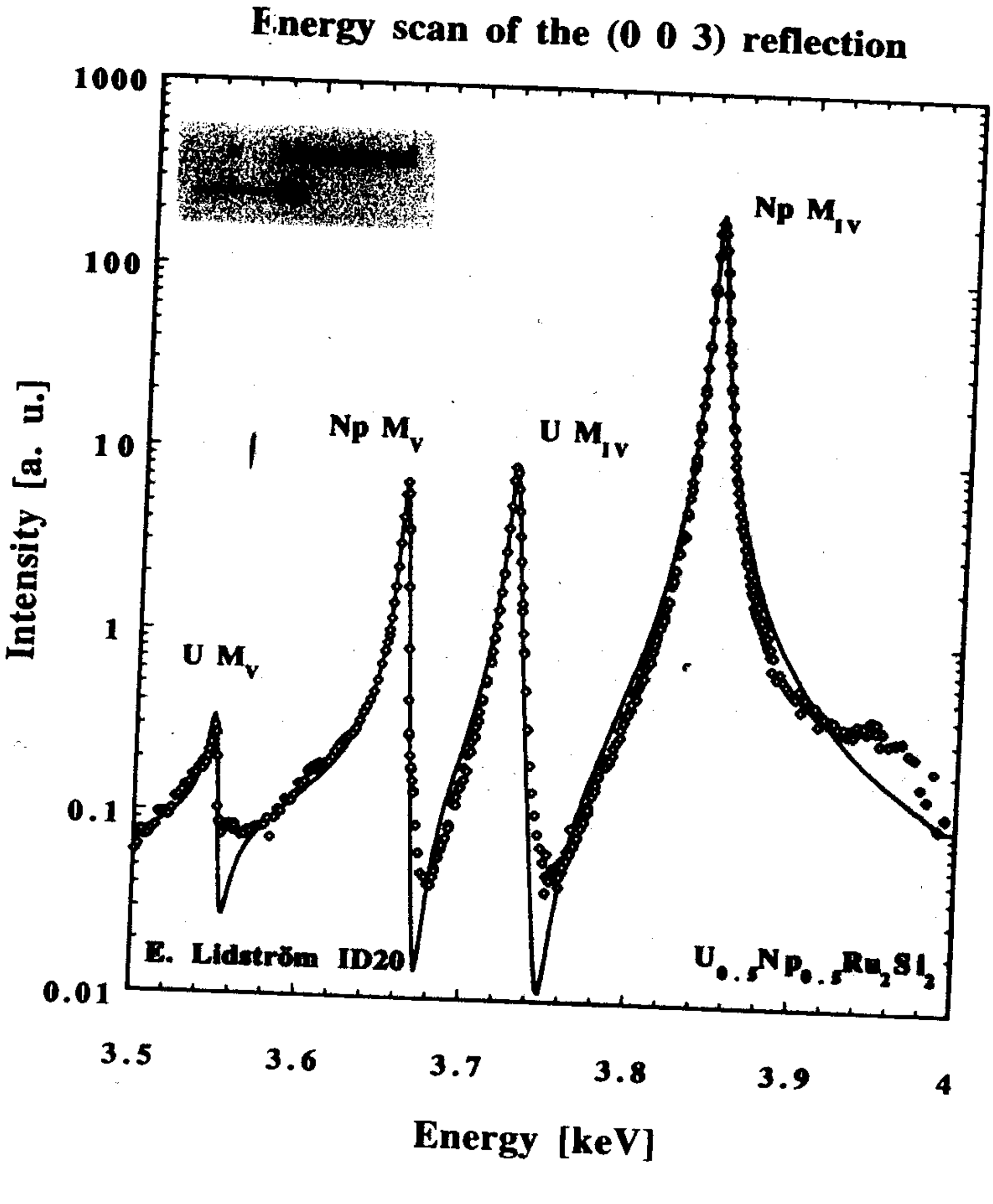
Resonant Scattering

HOLMIUM




5f
Mv
3.55 KeV
$(3d_{5/2})^6$
MIV
3.73
$(3d_{3/2})^4$
MIII
4.3
$(3p_{3/2})^4$
MII
5.0
$(3p_{1/2})^2$
MI
5.57
$(3s)^2$
$(1s)^2$

URANIUM



Energy scan of the (0 0 3) reflection
1000
100
10
1
0.1
0.01
Intensity [a. u.]
U M$_V$
Np M$_V$
U M$_{IV}$
Np M$_{IV}$
E. Lidström ID20
U$_{0.5}$Np$_{0.5}$Ru$_2$Si$_2$
3.5
3.6
3.7
3.8
3.9
4
Energy [keV]

CONCLUSIONS

- Synchrotron radiation has made magnetic x-ray scattering, absorption, and photoemission important tools for the study of magnetic material, and an important complement to neutron scattering.

- Magnetic effects in resonant scattering and absorption can be very large. They make possible the use of x-rays for magnetic structure analysis and for studies of surface phenomena.

- Absorption and polarization measurements provide information about excited electronic states and magnetic structure. Magnetic circular dichroism (MCD), Faraday rotation, magnetic extended fine structure (MEXAFS), and linear dichroism all give such information.