Advances in X-ray magnetic diffraction by using optimum polarization

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Argonne National Laboratory

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Photons diffraction by electronic spins

The first X-ray magnetic diffraction data

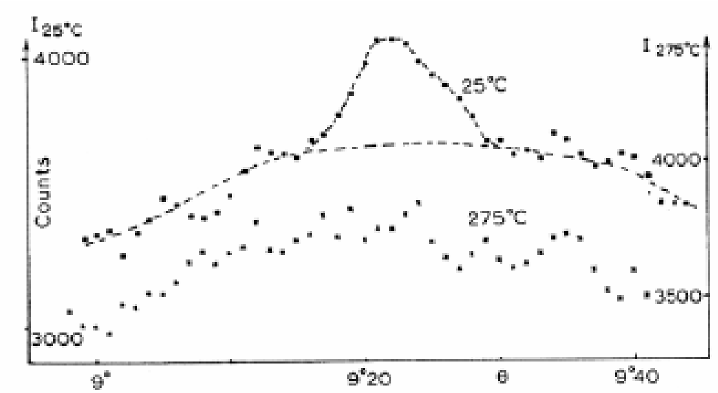


Fig. 1. Intensity $I_T(\theta)$ near the $(\frac{1}{2}\frac{1}{2}\frac{1}{2})$ position at t = 25° C and 275° C in counts/225 min. The hump which cover the interval could be due to some impurity.

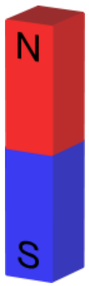
- Antiferromagnet NiO using a **tube source**.
- Count rate was approximately 0.03 cps.
- Scans required 3.5 hours per data point (3.5 days per plotted curve).
- It took **one week** to obtain these data.

F. de Bergevin and M. Brunel, Phys. Lett. **39**A, 141 (1972).

What can we do with ever brighter X-ray source?

LS separation

A magnetic moment consists of spin- and orbital angular momentum

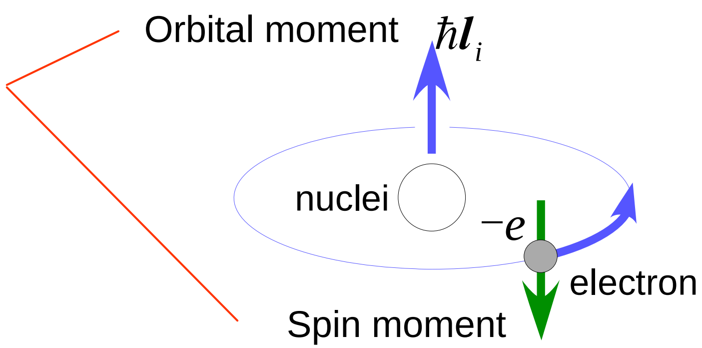


$$\boldsymbol{M} = -\mu_B \sum_i (\boldsymbol{l}_i + 2\boldsymbol{s}_i)$$

Relativistic effect

Spin-orbit coupling

- Magnetism
- Material design
- Spintronics
- etc.



Experimental separation of spin- and orbital-magnetization

Magnetic Circular dichroism	Non Resonant X-ray Magnetic Scattering
zero momentum transfer	Momentum transfer dependence
net moment	Spatial extent of respective moment densities

Outline

- Background
  - Photons scattering by electronic spins
  - Early studies of *LS* separation
  - Limits of conventional technique
- Experimental
  - New technique without using polarization analyzer
- Results
  - Significant improvement in precision
- Summary and Future plan

Collaborators

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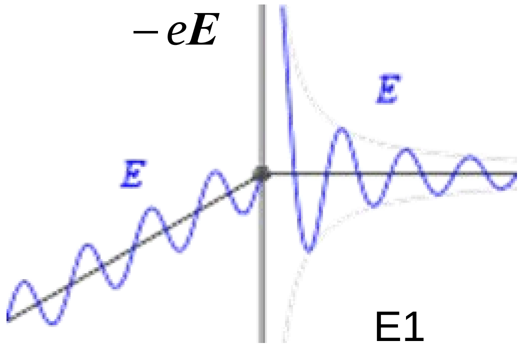
Photons scattering by electronic spins

Charge scattering
(Thomson scattering)



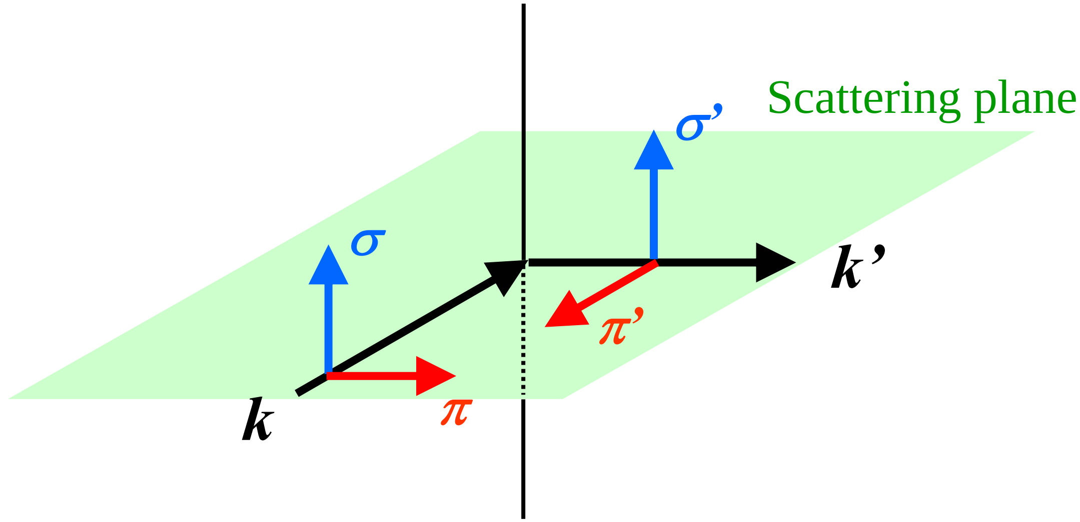
Charge amplitude ~ N_{total}

$$\begin{pmatrix} E'_\sigma \\ E'_\pi \end{pmatrix} = \begin{pmatrix} C_{\sigma\sigma} & 0 \\ 0 & C_{\pi\pi} \end{pmatrix} \begin{pmatrix} E_\sigma \\ E_\pi \end{pmatrix}$$

Not induce orthogonal polarization

Zero off-diagonal elements

Definition of polarization

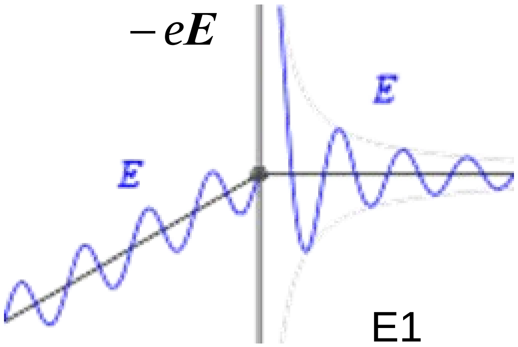


σ polarization is normal to the scattering plane.

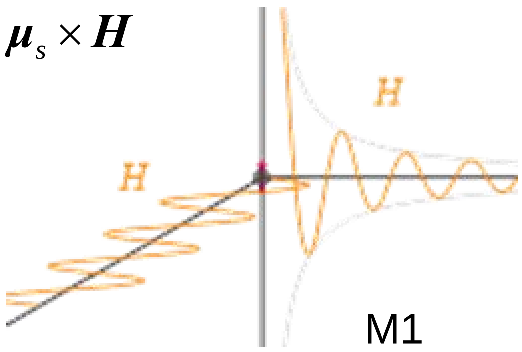
π polarization is parallel to the scattering plane.

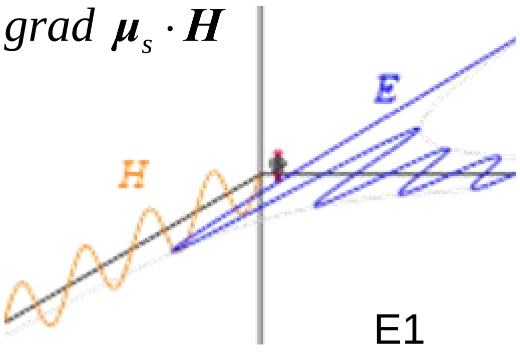
Photons scattering by electronic spins

Charge scattering
(Thomson scattering)



Magnetic scattering





$$\text{Charge amplitude} \sim N_{total}$$

$$\text{Magnetic amplitude} \sim \frac{\hbar\omega}{mc^2} \cdot N_{mag} \sim \frac{\text{Charge amplitude}}{1000}$$

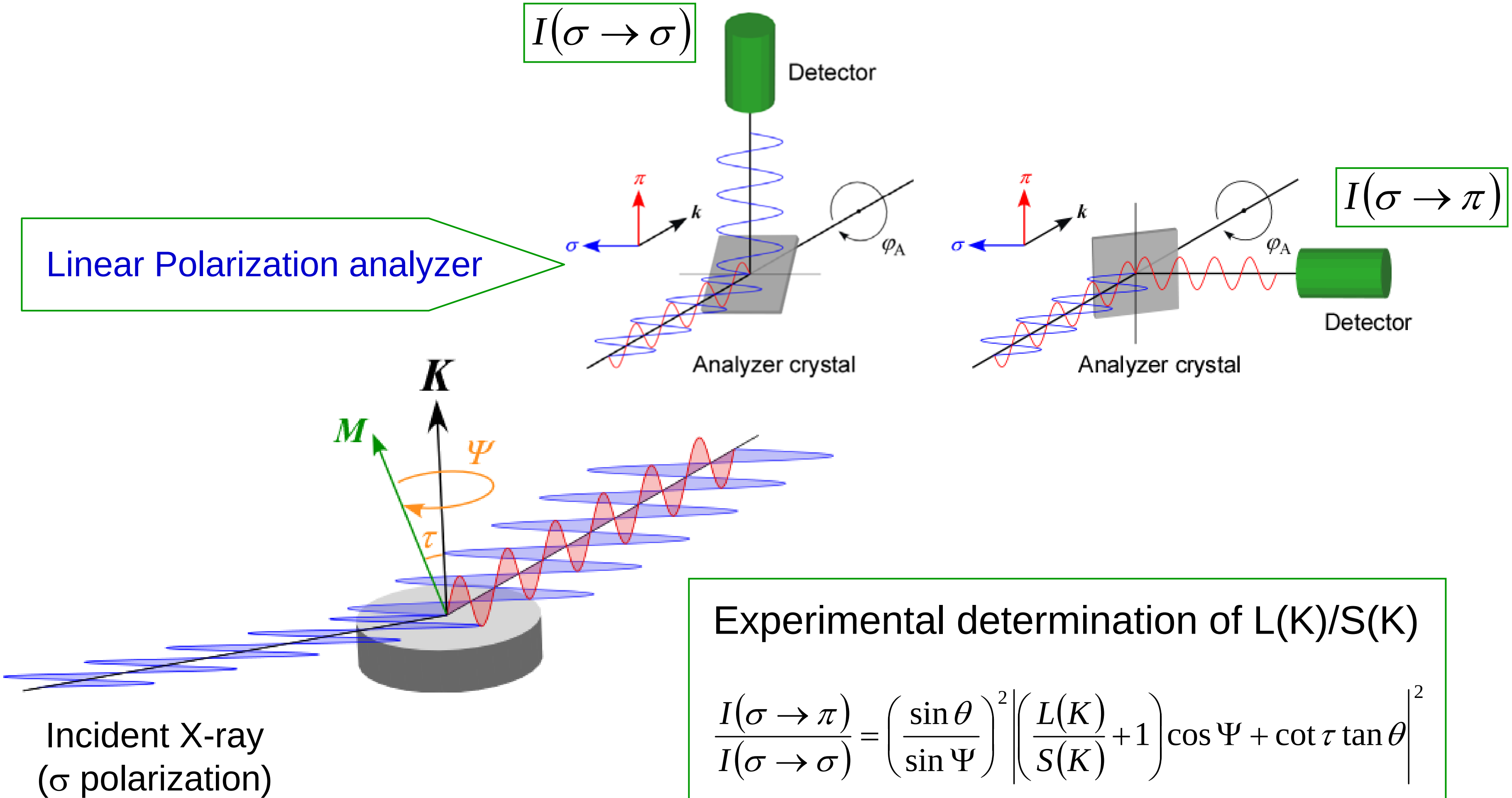
$$\begin{pmatrix} E'_\sigma \\ E'_\pi \end{pmatrix} = \begin{pmatrix} C_{\sigma\sigma} & 0 \\ 0 & C_{\pi\pi} \end{pmatrix} \begin{pmatrix} E_\sigma \\ E_\pi \end{pmatrix}$$

$$\begin{pmatrix} E'_\sigma \\ E'_\pi \end{pmatrix} = -i\frac{\hbar\omega}{mc^2} \begin{pmatrix} M_{\sigma\sigma} & M_{\sigma\pi} \\ M_{\pi\sigma} & M_{\pi\pi} \end{pmatrix} \begin{pmatrix} E_\sigma \\ E_\pi \end{pmatrix}$$

Not induce orthogonal polarization
Zero off-diagonal elements

Induce orthogonal polarization
Nonzero off-diagonal elements

Conventional technique for *LS* separation



Experimental determination of L(K)/S(K)

$$\frac{I(\sigma \to \pi)}{I(\sigma \to \sigma)} = \left(\frac{\sin\theta}{\sin\Psi}\right)^2 \left| \left(\frac{L(K)}{S(K)} + 1\right) \cos\Psi + \cot\tau \tan\theta \right|^2$$

Early studies of *LS* separation

First recognition of the ability to determine orbital moment in magnetic materials

F. de Bergevin and M. Brunel, Acta Cryst. A **37**, 314 (1981).

M. Blume, J. Appl. Phys. **57**, 3615 (1981).

Holmium (4*f*)

D. Gibbs *et al*., PRL **61**, 1241 (1988). First demonstration

D. Gibbs *et al*., PRB **43**, 5663 (1991).

Uranium arsenide (5*f*)

D. B. McWhan *et al*., PRB **42**, 6007 (1990).

S. Langridge *et al*., PRB **55**, 6392 (1997).

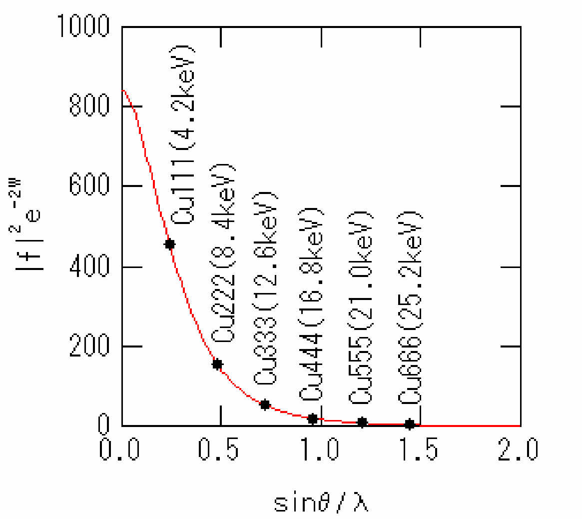
Nickel monoxide (3*d*)

V. Fernandez *et al*., PRB **57**, 6392 (1998).

Conducted by using Polarization analyzer

Drawbacks of polarization analyzer

A polarization analyzer exploits Bragg reflection at the Brewster angle to remove π polarization component for the analyzer crystal.



Expected diffraction intensity of Cu[111] crystal, where form factor and Debye-Waller factor are taken into account.

- **Considerable loss of signal intensity**

  Reflectivity decreases rapidly as X-ray energy increases.

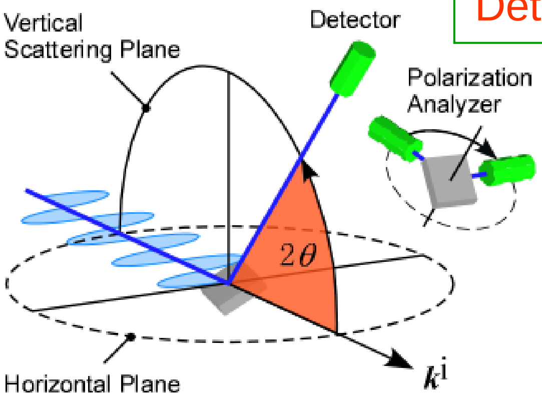
- **Strong restriction on available X-ray energy**

  Interatomic Spacing is discrete and uncontrollable.

- **Complicated procedure for obtaining correct intensity**

  Diffraction intensities should be integrated by rocking both sample and analyzer crystals.

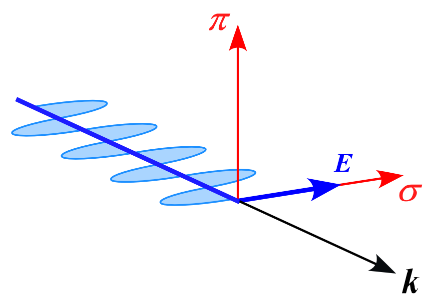
Unconventional method of magnetic diffraction



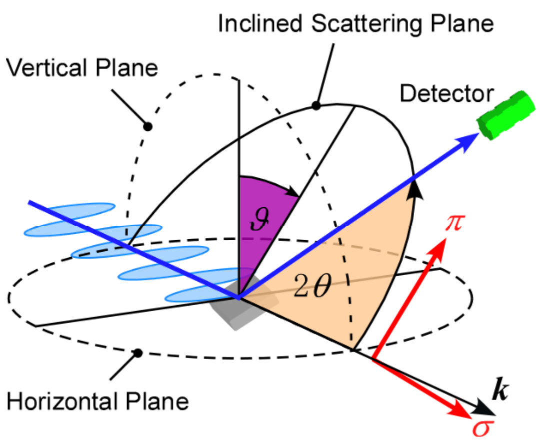
Conventional polarization analysis

Detach the polarization analyzer to remove the cause of inefficiency

Incident X-ray : linear σ polarization



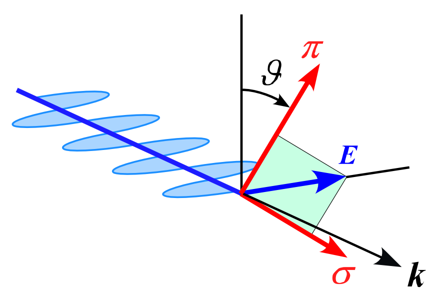
$$\begin{pmatrix} E_\sigma \\ E_\pi \end{pmatrix} = \begin{pmatrix} E \\ 0 \end{pmatrix}$$



Variable scattering plane method

Incline the scattering plane to change the incident polarization

Incident X-ray : linear variable polarization



$$\begin{pmatrix} E_\sigma \\ E_\pi \end{pmatrix} = \begin{pmatrix} E\cos\vartheta \\ E\sin\vartheta \end{pmatrix}$$

Polarization profile specifies scattering origin

Diffraction intensity depends on ϑ.

Charge scattering

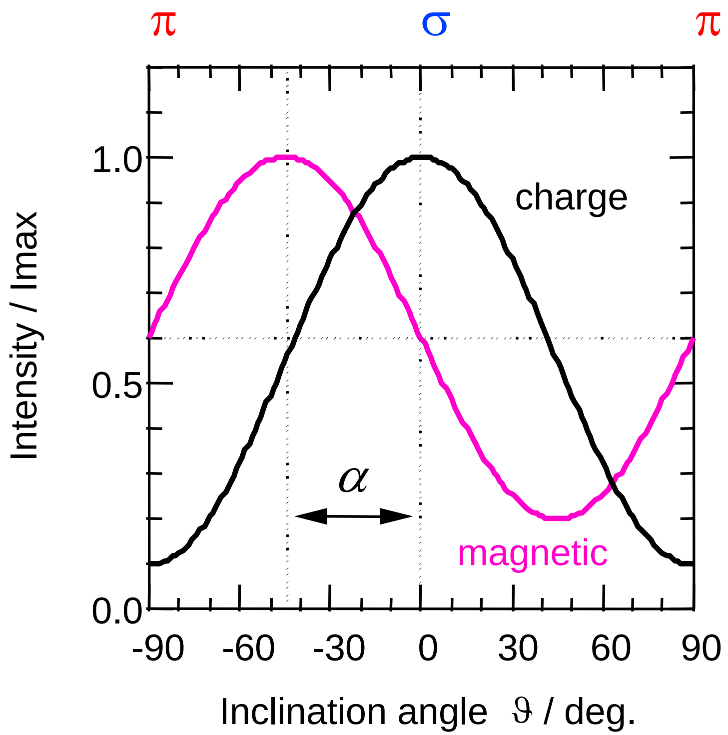
$$\begin{pmatrix} E'_\sigma \\ E'_\pi \end{pmatrix} = \begin{pmatrix} C_{\sigma\sigma} & 0 \\ 0 & C_{\pi\pi} \end{pmatrix} \begin{pmatrix} E\cos\vartheta \\ E\sin\vartheta \end{pmatrix}$$

$$I(\vartheta) \propto \mathrm{tr}\left[\langle M_c^\dagger\rangle\langle M_c\rangle\right] + \mathrm{tr}\left[\langle M_c^\dagger\rangle\langle M_c\rangle\sigma_3\right]\underline{\cos 2\vartheta}$$

Magnetic scattering

$$\begin{pmatrix} E'_\sigma \\ E'_\pi \end{pmatrix} = -i\frac{\hbar\omega}{mc^2} \begin{pmatrix} M_{\sigma\sigma} & M_{\sigma\pi} \\ M_{\pi\sigma} & M_{\pi\pi} \end{pmatrix} \begin{pmatrix} E\cos\vartheta \\ E\sin\vartheta \end{pmatrix}$$

$$I(\vartheta) \propto \mathrm{tr}\left[\langle M_m^\dagger\rangle\langle M_m\rangle\right] + \mathrm{tr}\left[\langle M_m^\dagger\rangle\langle M_m\rangle\sigma_3\right]\underline{\cos 2\vartheta} + \mathrm{tr}\left[\langle M_m^\dagger\rangle\langle M_m\rangle\sigma_1\right]\underline{\sin 2\vartheta}$$

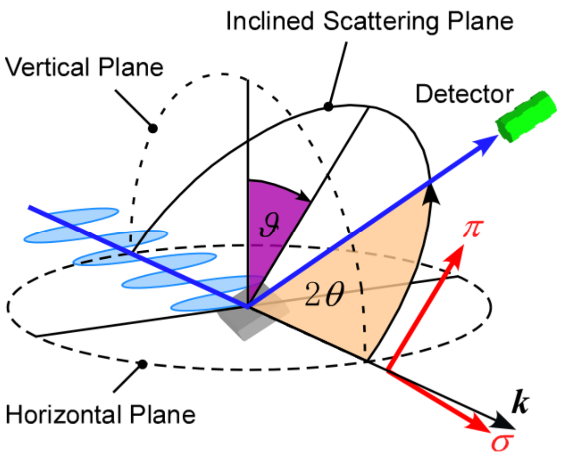


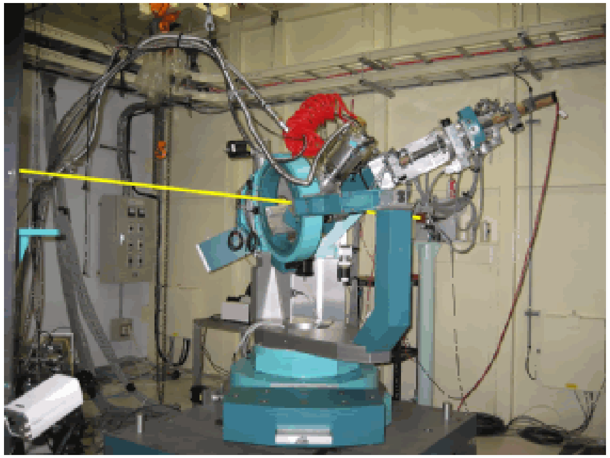
Polarization profiles of diffraction intensities.

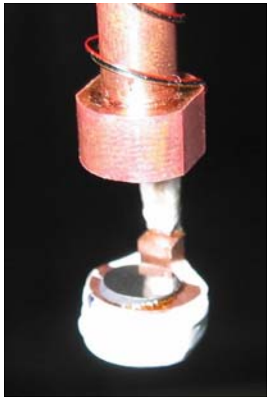
Experimental details

Experimental facility

BL46XU/SPring-8
6-circle diffractometer
NaI Scintillation detector
He refrigerator
X-ray energy:16 keV







Holmium

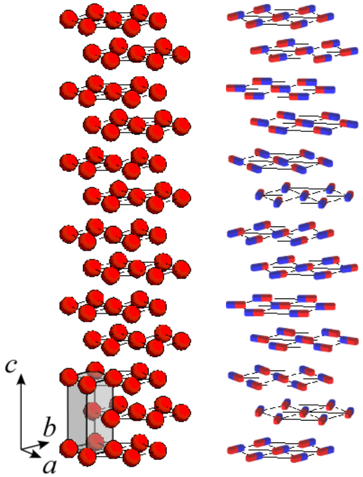
Sample shape: disk (5mm in diameter and 1mm thick)

Surface: (120) plane of hexagonal lattice

Magnetic structure: helix (20K~132K)

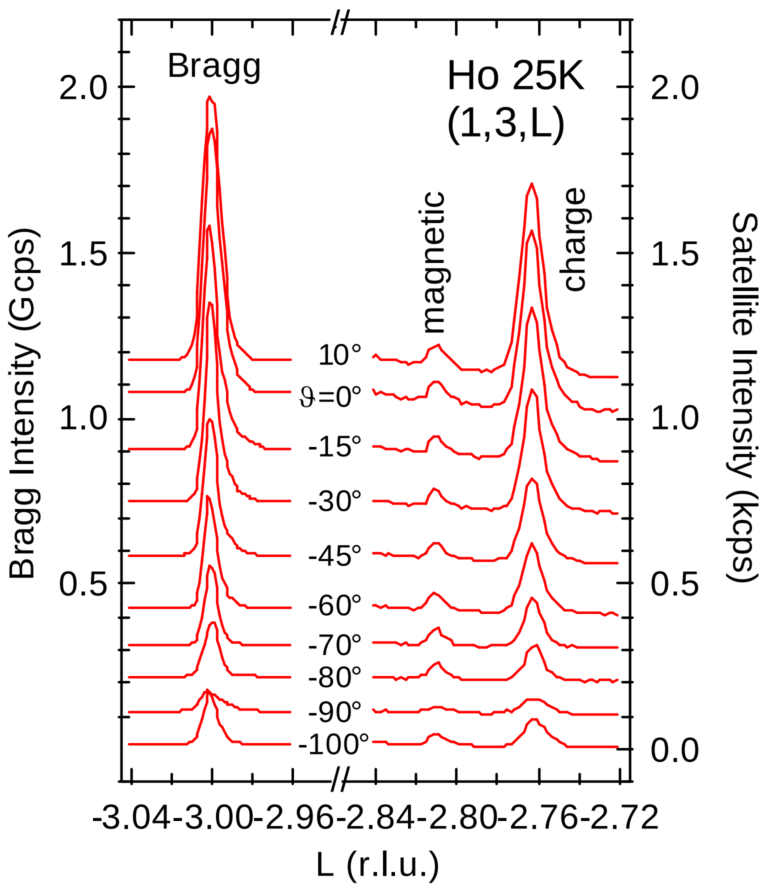
Modulation wave vector: 0.194c* (25K)

Ho^{3+} ground state: S=2, L=6

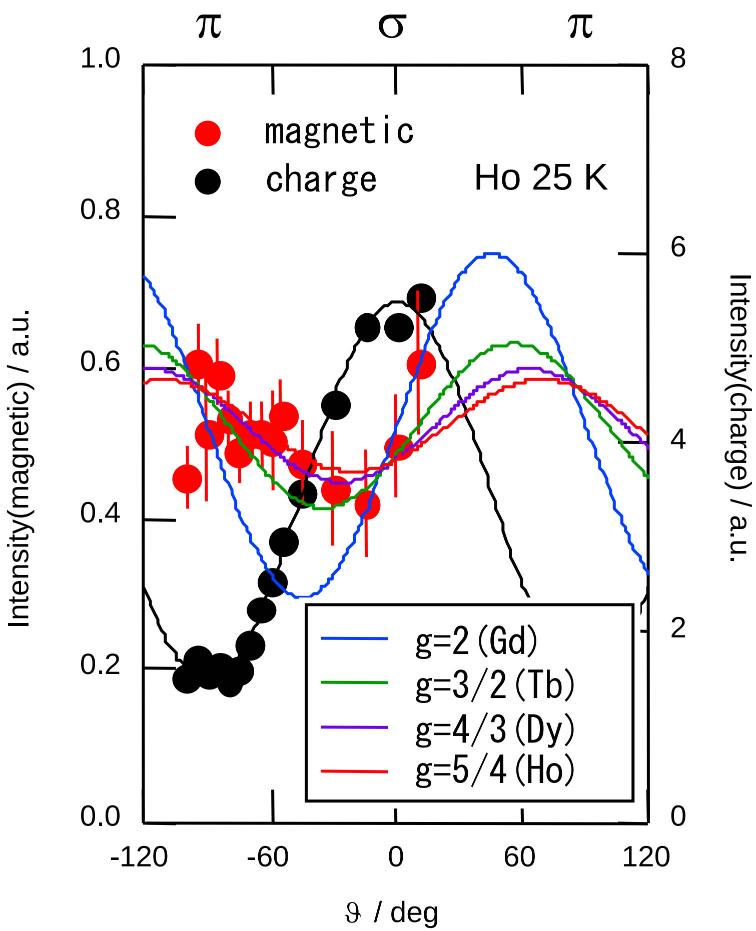


Crystal and magnetic structure of holmium

Successful but qualitative results



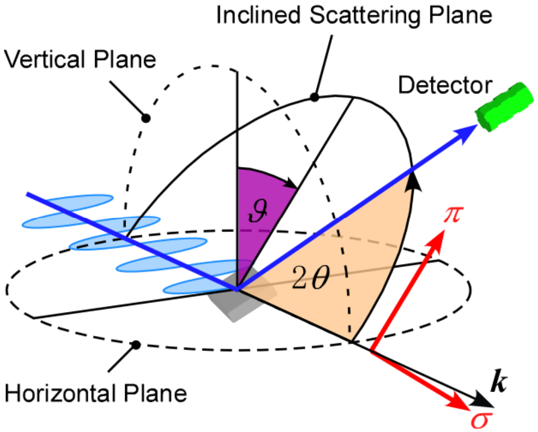
Diffraction profiles measured by using various polarization



Polarization profiles of diffraction intensities

Quantitative evaluation of L/S requires further improvement.

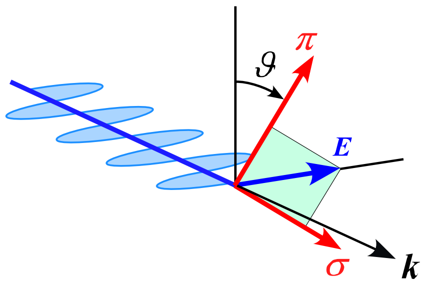
Reduction of diffractometer motion to the minimum



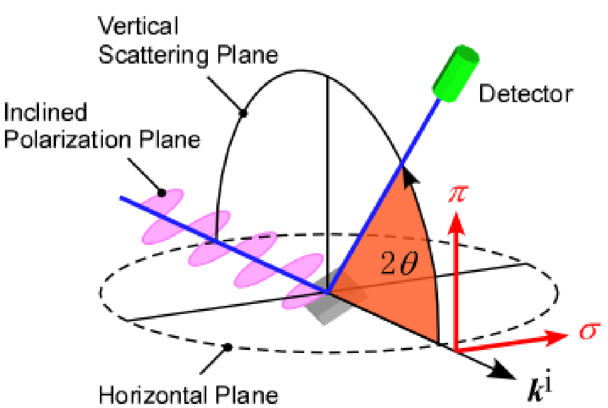
Variable scattering plane method

Incline the scattering plane to change the incident polarization

Incident X-ray : linear variable polarization



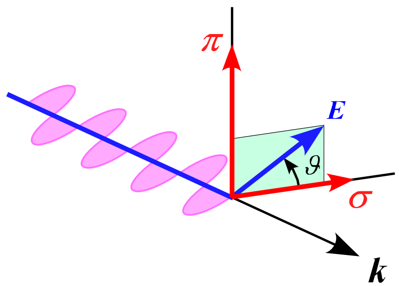
$$\begin{pmatrix} E_\sigma \\ E_\pi \end{pmatrix} = \begin{pmatrix} E\cos\vartheta \\ E\sin\vartheta \end{pmatrix}$$



Polarization rotation method

Rotate the incident polarization plane by X-ray phase retarder

Incident X-ray : linear variable polarization



$$\begin{pmatrix} E_\sigma \\ E_\pi \end{pmatrix} = \begin{pmatrix} E\cos\vartheta \\ E\sin\vartheta \end{pmatrix}$$

Summary and future plan

■ New techniques for *LS* separation have been developed to avoid the use of polarization analyzer .

■ *L*/*S* in holmium was qualitatively estimated by rotating the scattering plane.

■ Elemental resolution of *L*/*S* was achieved by rotating the polarization plane.

■ Polarization sweep technique will be integrated into spatial and temporal resolved measurements.