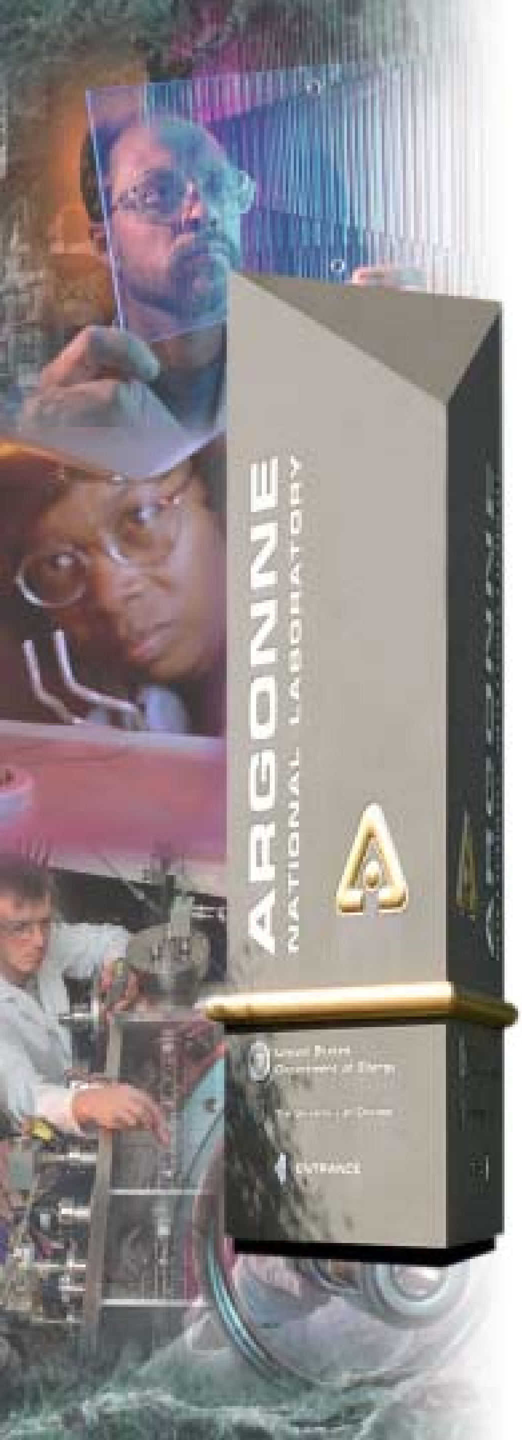
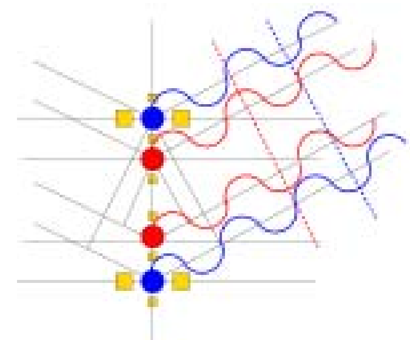


Beyond element-specific magnetism: Magnetic spectroscopy in the diffraction channel

Daniel Haskel
Advanced Photon Source

Nanomagnetism using x-ray techniques
Lake Geneva, WI, August 2004



APS

Andy Cady
Zahir Islam
Jonathan Lang
George Srajer
Julie Cross
Boris Deriy
Kurt Goetze
Ruben Khachatryan

Samples

Paul Canfield, Ames Lab
Joaquin Garcia, U. Zaragoza
Bao-min Ma, Magnequench Inc.

Theory

Michel van Veenendal
Alexei Ankudinov

* LDRD funding

Argonne National Laboratory

Office of Science
U.S. Department of Energy

A U.S. Department of Energy
Office of Science Laboratory
Operated by The University of Chicago

Outline

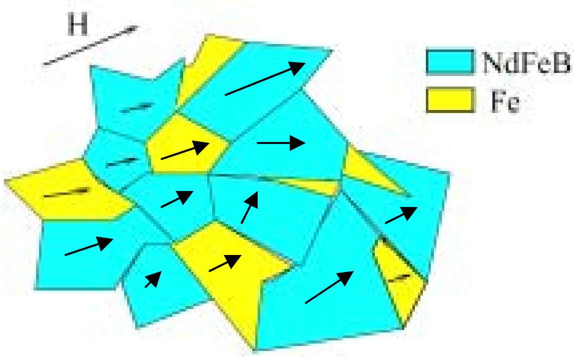
- The Need: to go beyond element specific magnetism.
- The Tool: Resonant diffraction of CP x-rays.
- Example: Site-specific magnetism in $Nd_2Fe_{14}B$.
- Magnetic spectroscopy in the diffraction channel.
- Outlook and Summary.

The Need: to go beyond element specific magnetism

Element-specific XMCD lacks phase, site selectivity.

Phase-Specific

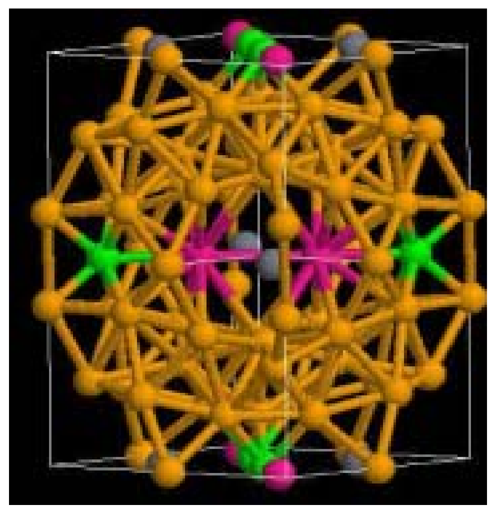
Same element in coexisting phases



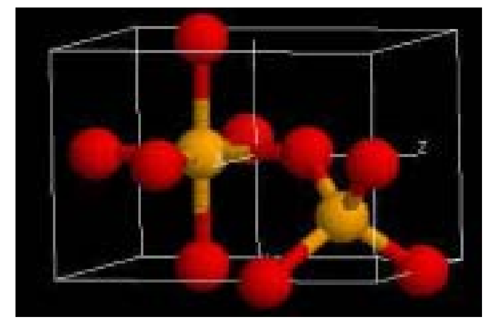
Nanocomposites: NdFeB/Fe
Impurity phases: $Sm_2Co_7/SmCo_5$
Intrinsic (CMR, stripes)

Site-specific

Same element in inequivalent crystal sites



Fe Nd B



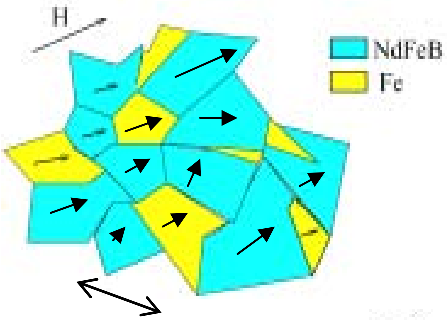
Fe_3O_4
$R_2Fe_{14}B$
$Gd_5Si_2Ge_2$

The Tool: resonant diffraction of CP x-rays

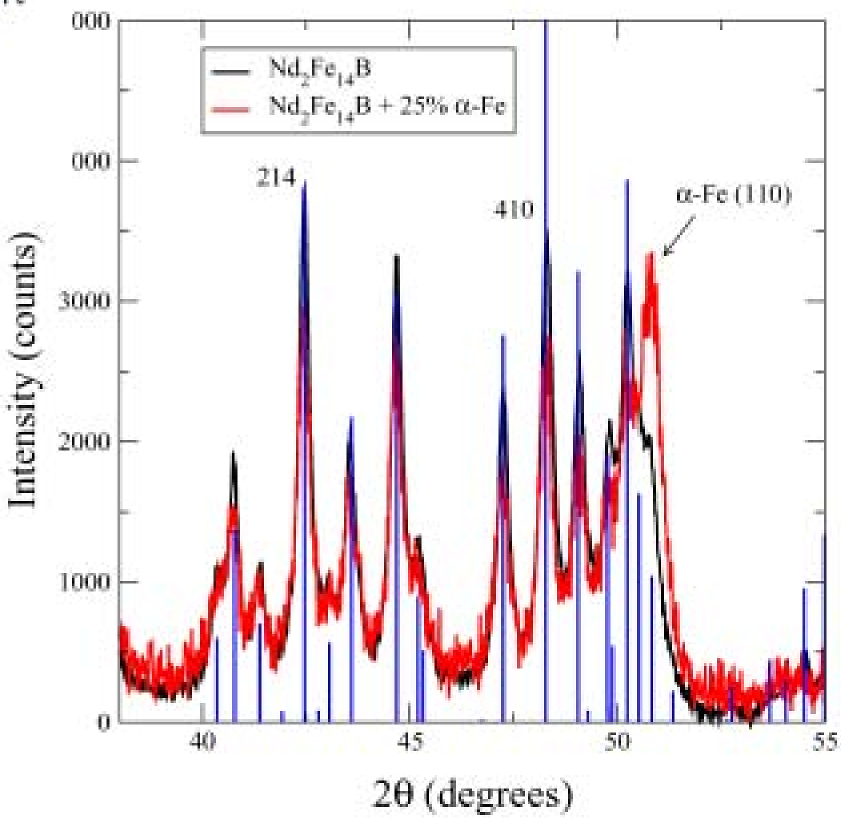
Diffraction = phase and site selectivity

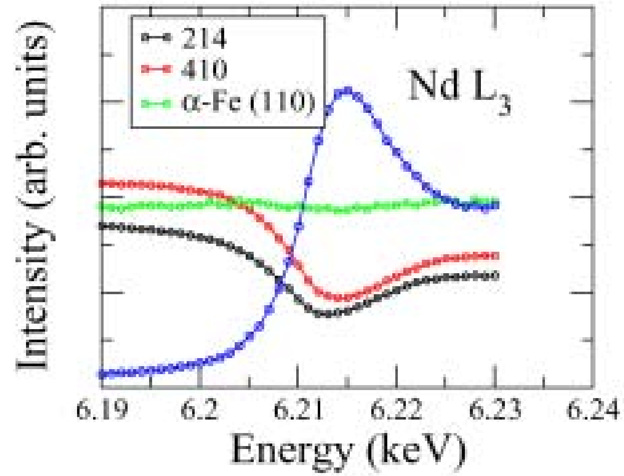
Atomic resonance = element specificity

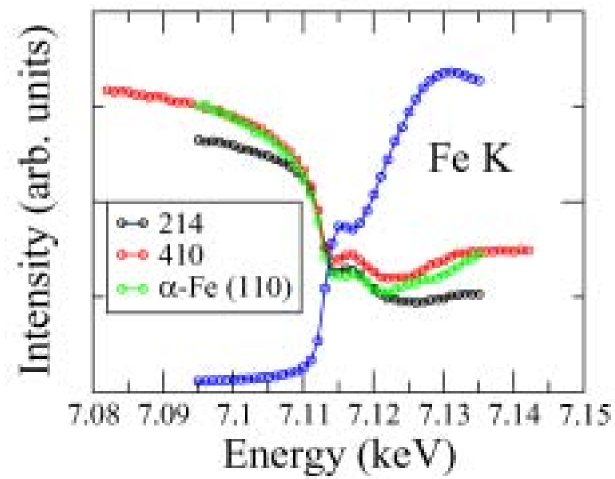
CP x-rays = coupling to sample magnetization



25-40 nm





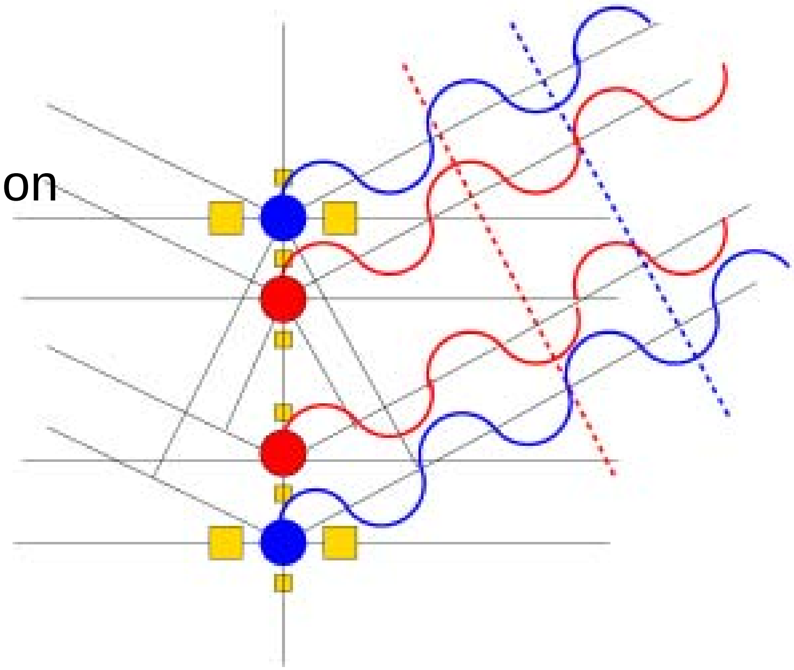


The Tool: resonant diffraction of CP x-rays

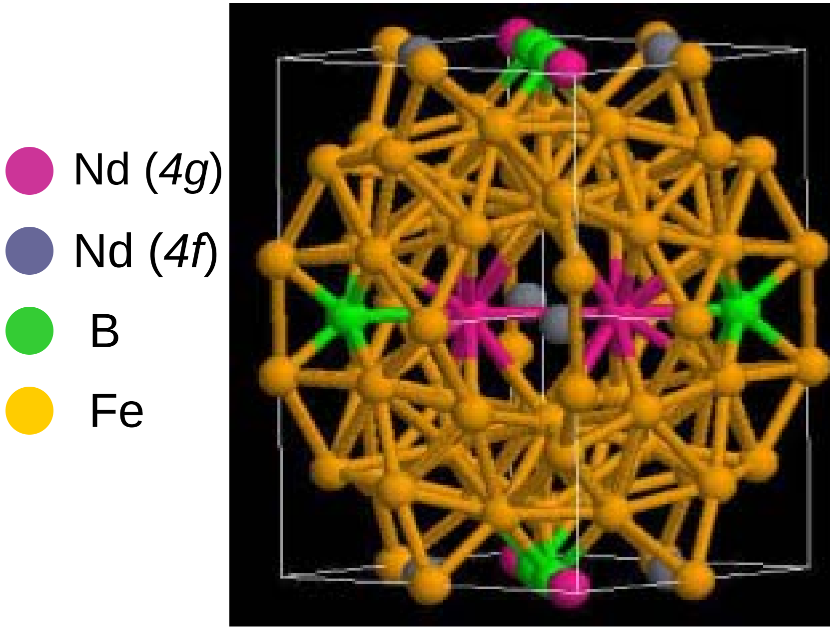
Diffraction = phase and site selectivity

Atomic resonance = element specificity

CP x-rays = coupling to sample magnetization



$Nd_2Fe_{14}B$



$$(4g)\ (x,\bar{x},0);(\bar{x},x,0);(\tfrac{1}{2}+x,\tfrac{1}{2}+x,\tfrac{1}{2});(\tfrac{1}{2}-x,\tfrac{1}{2}-x,\tfrac{1}{2})$$

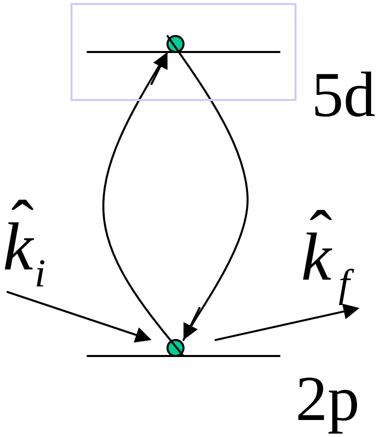
$$(4f)\ (x,x,0);(\bar{x},\bar{x},0);(\tfrac{1}{2}+x,\tfrac{1}{2}-x,\tfrac{1}{2});(\tfrac{1}{2}-x,\tfrac{1}{2}+x,\tfrac{1}{2})$$

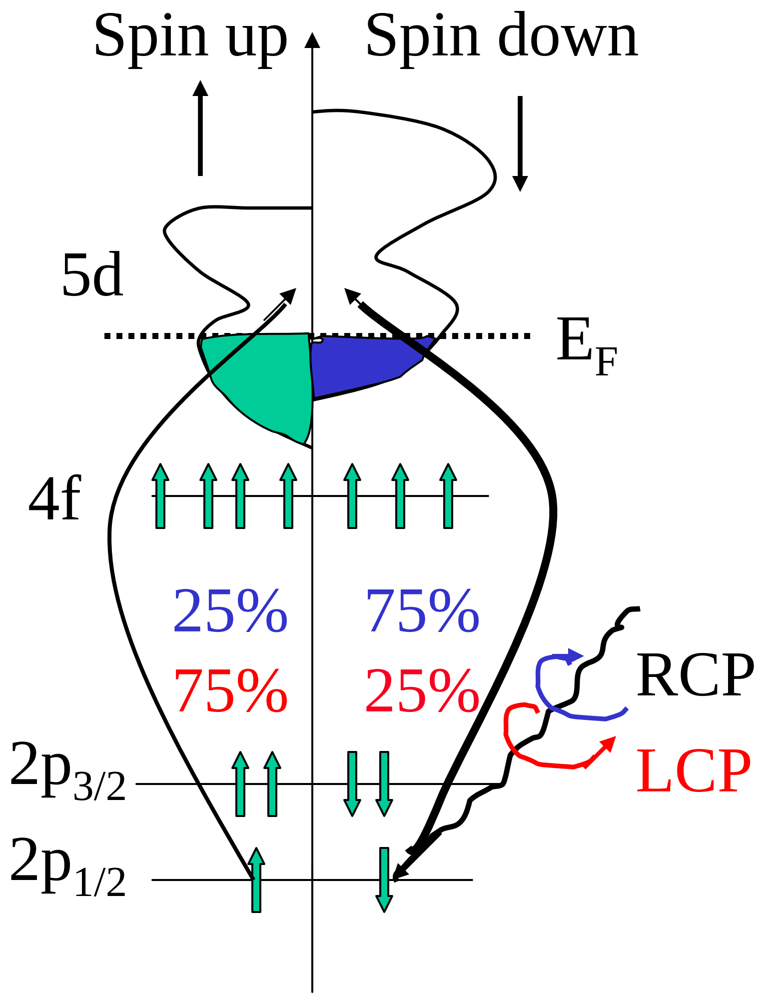
Resonant absorption and scattering of CP x-rays

Scattering

(XRES)

$$\Delta I_s = I^L - I^R$$

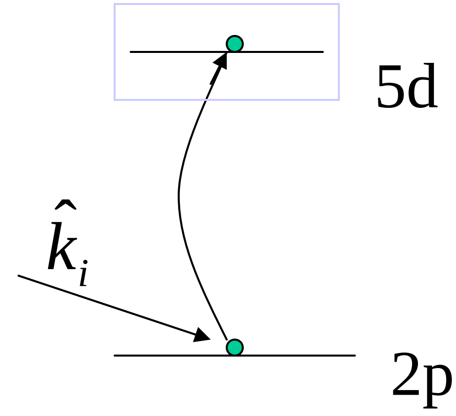




Absorption

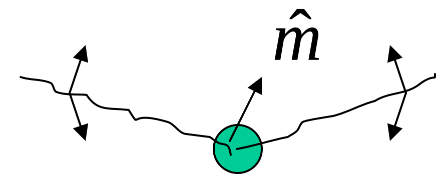
(XMCD)

$$\mu_m = \mu^L - \mu^R$$



Scat. Amp. (E1) for magnetic ion near resonance (Hannon *et al.*, 1988)

$$f = (f_0 + f_e)(\varepsilon'^* \cdot \varepsilon) + i f_m (\varepsilon'^* \times \varepsilon) \cdot \hat{m} + \cancel{f^{mag}}$$



Charge	Resonant charge	Resonant Magnetic	Non-resonant magnetic
1	0.2-0.5	0.01-0.5	0.001

$I \propto f^2 =$ charge + magnetic + <u>charge ×magnetic</u>

Needs to get rid of strong charge scattering (ferro-ferrimagnets)

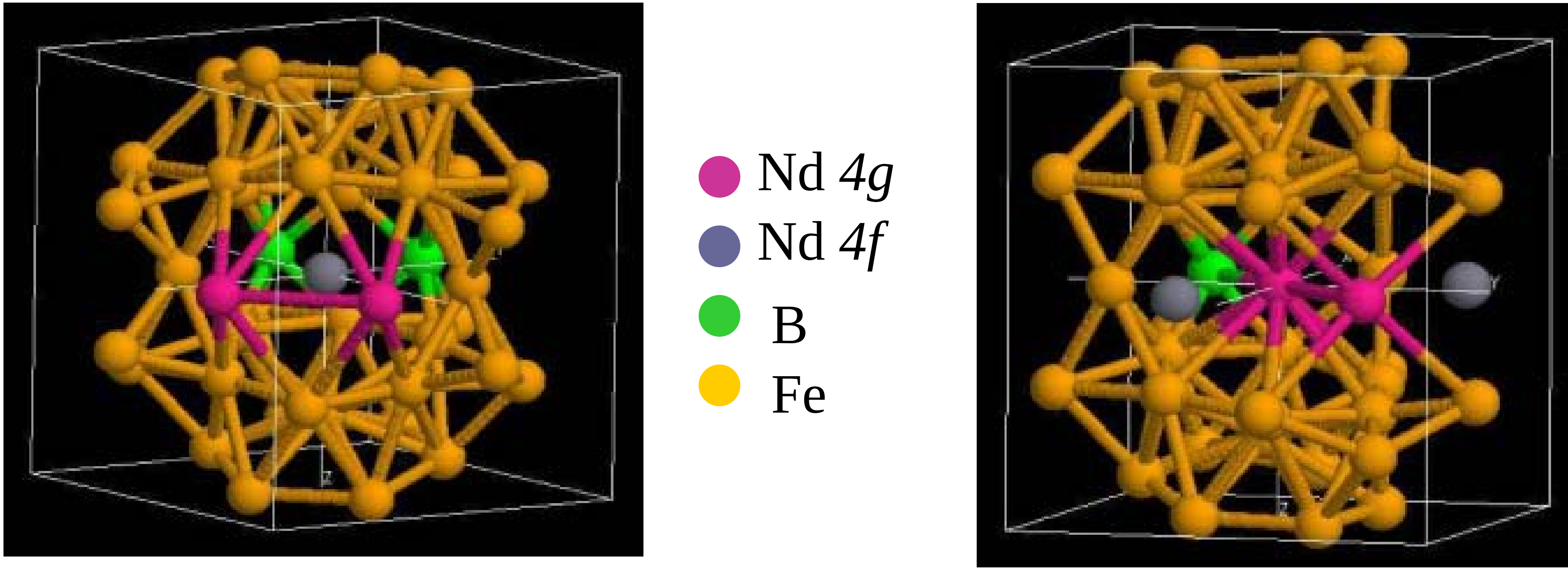
$$\Delta I_s = I^L - I^R \propto \{ i[(\varepsilon'^* \cdot \varepsilon)^* \underline{(\varepsilon'^* \times \varepsilon)} \cdot \hat{m}](f_0 + \underline{f_e})^* \cdot \underline{f_m} \} + c.c.$$

$\hat{m} \| $ scat. plane

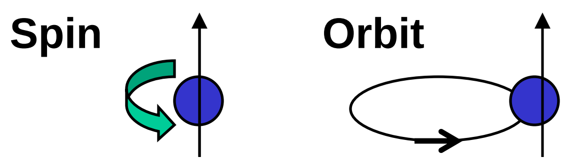
Strong E-dependence

$$\Sigma I_s = I^L + I^R = A(f_0 + f_e)^2 + \cancel{B f_m^2}$$

Example: Site-specific magnetism in $Nd_2Fe_{14}B$



- Fe: ~ 31 μ_B/f.u.
  Nd: ~ 6 μ_B/f.u.

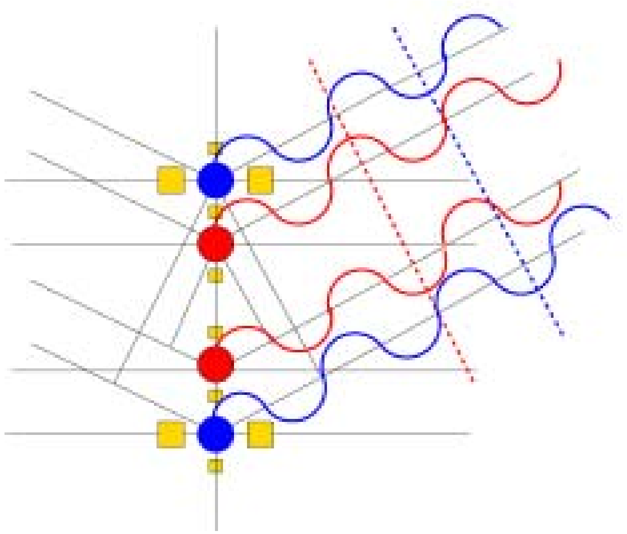


- Easy axis: [001] at RT, tilts towards [110] at low T
- Magnetic "hardness" due to large orbital moment of Nd ions.

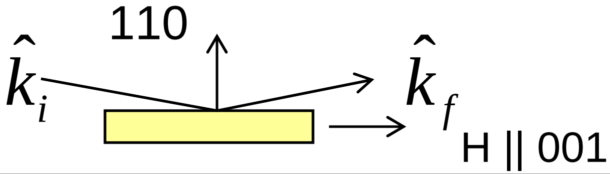
Site-specific Nd structure factors in $Nd_2Fe_{14}B$

$$F_{hkl} = f_{Nd}(Q,E)\sum_{i=1}^{4} e^{i\vec{Q}\cdot\vec{r}_i}$$

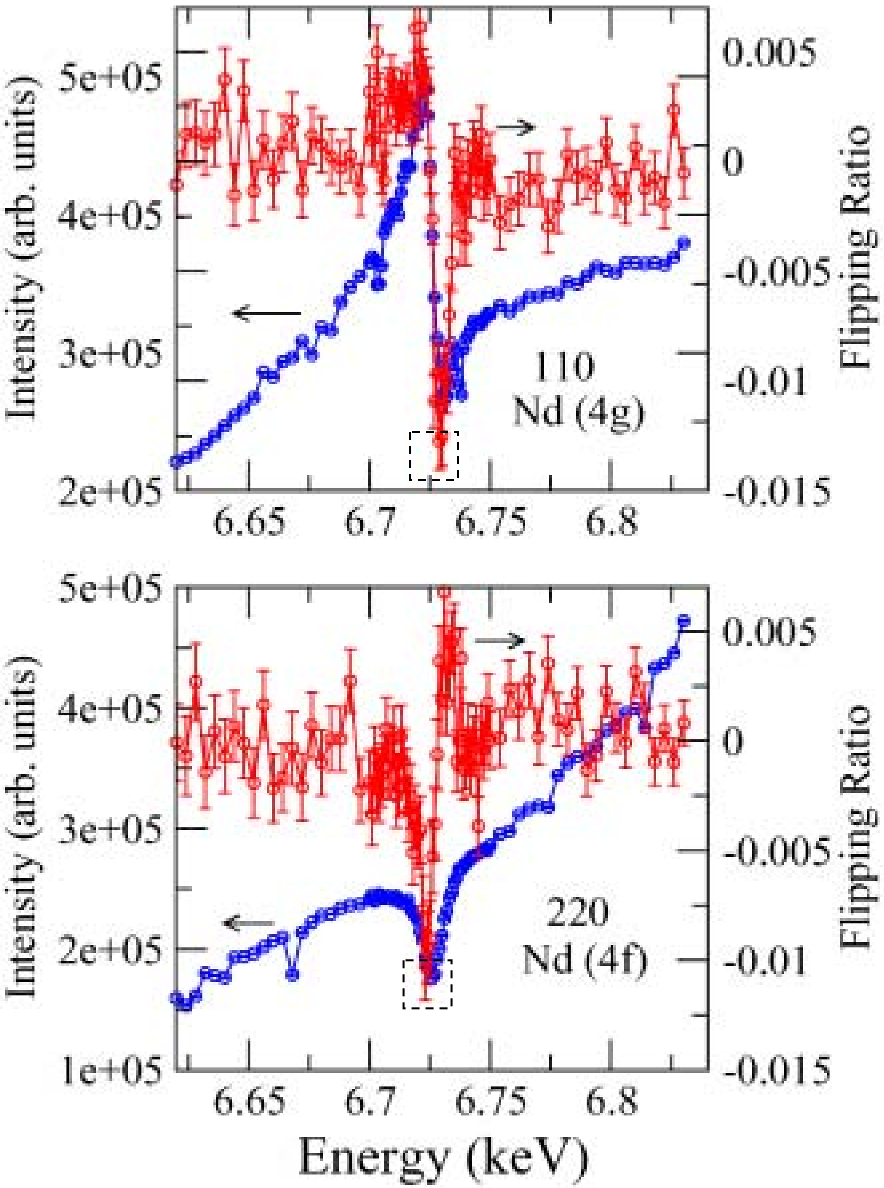
$$\vec{Q} = 2\pi(\frac{h}{a},\frac{k}{b},\frac{l}{c}), \vec{r}_i = (x_i a + y_i b + z_i c)$$

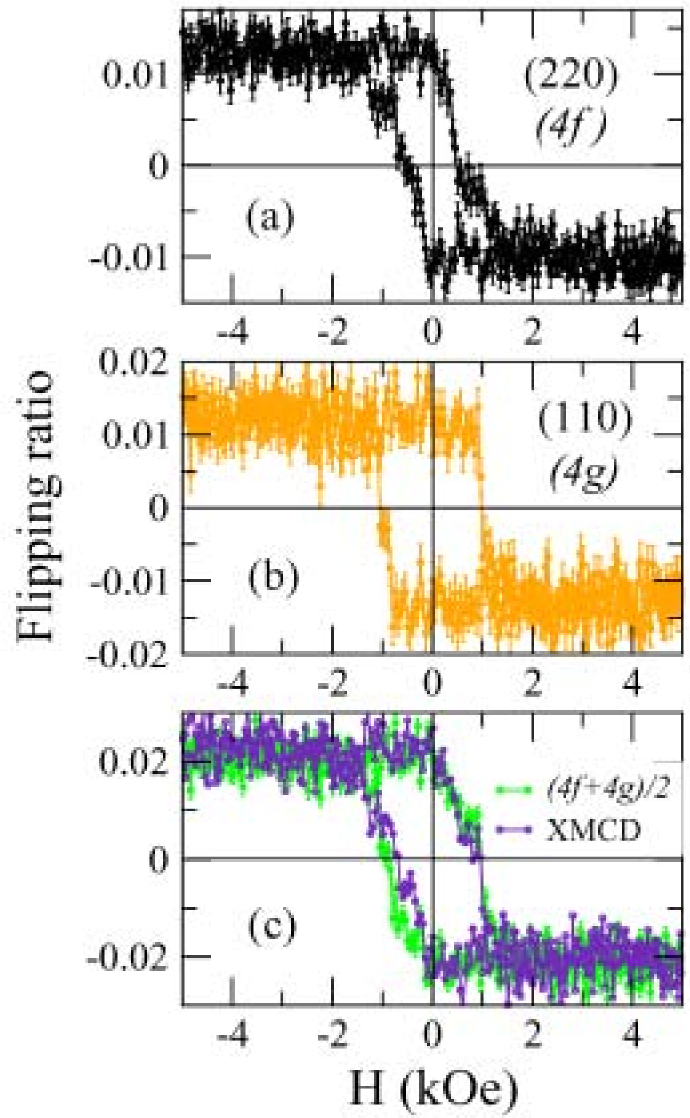


Wyckoff Site	(110)	(220)	(440)
4f	2.2+0.5i	140+35i	86+29i
4g	69+15i	5.2+1.3i	92+31i

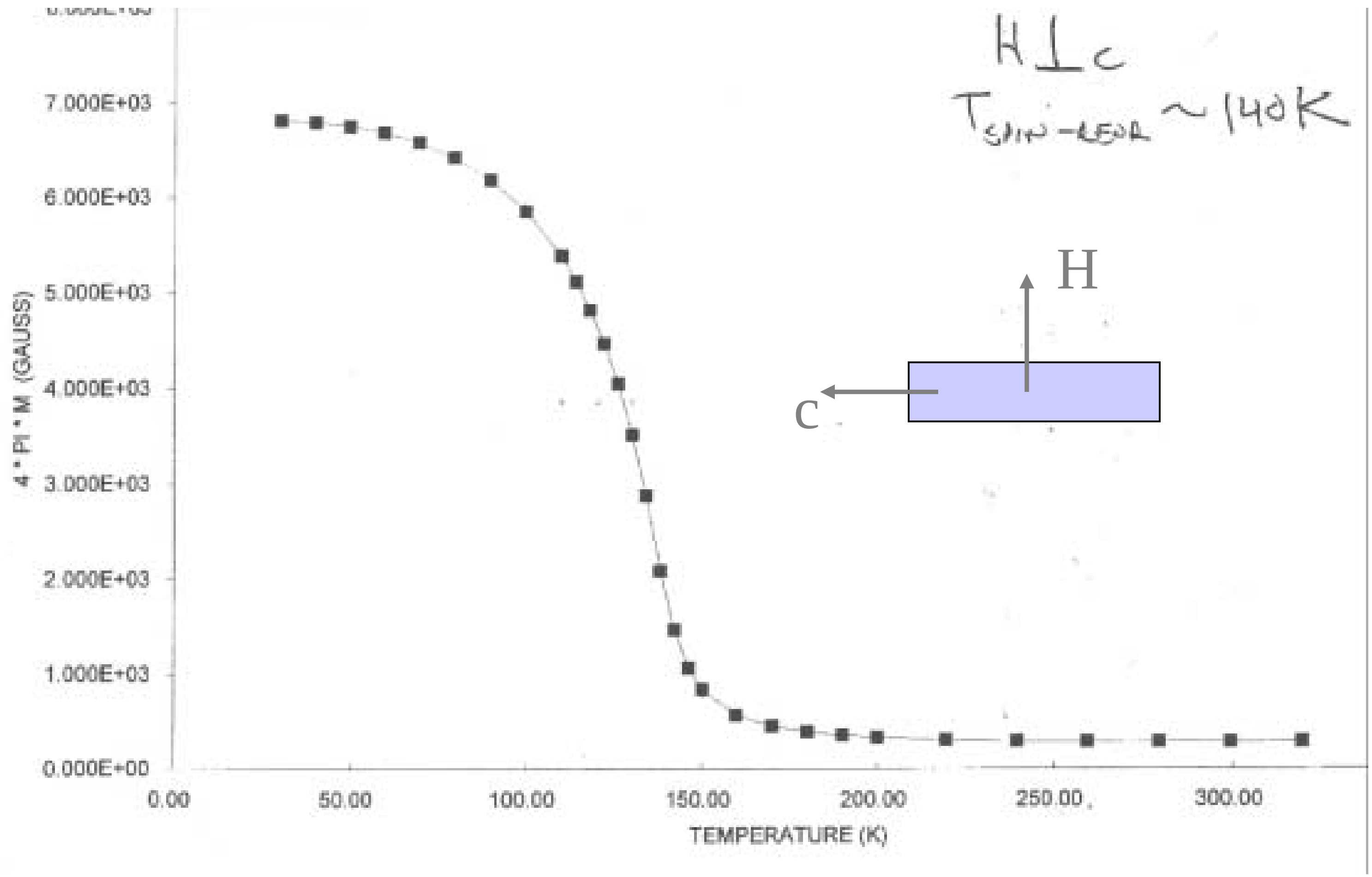


$$I = | f_e(\varepsilon'^* \cdot \varepsilon) + i f_m (\varepsilon'^* \times \varepsilon) \cdot \hat{m} |^2$$



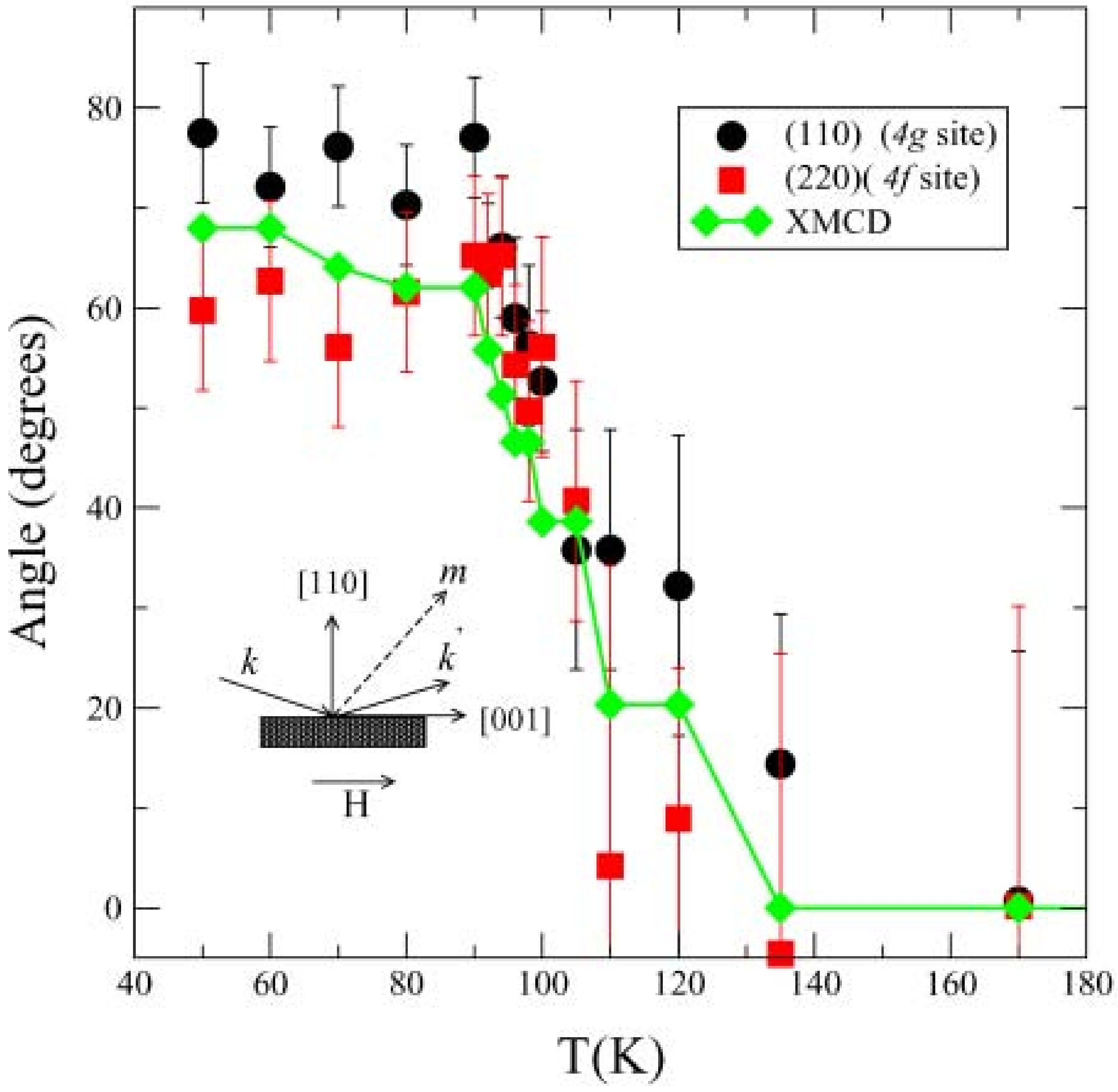


Spin reorientation transition in $Nd_2Fe_{14}B$



Paul Canfield, Ames Lab

Site-specific spin reorientation transition



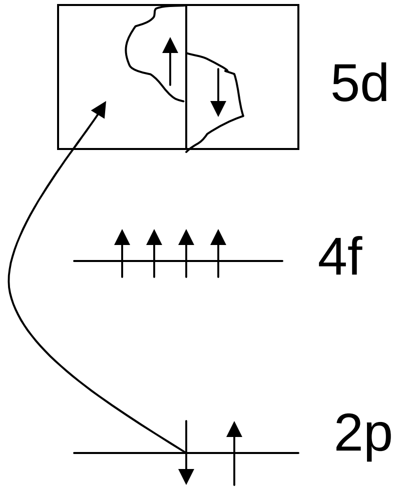
Magnetic spectroscopy in the diffraction channel

- Spin-dependent DOS (E_F).
- Size of moment (orbital, spin).
- Orientation of moment, magnetization reversal.

Easy interpretation in absorption channel (XMCD)
[optical theorem $\mu_m \propto f''_m(q=0)$].

$$\mu_m \propto N(\uparrow) - N(\downarrow)$$

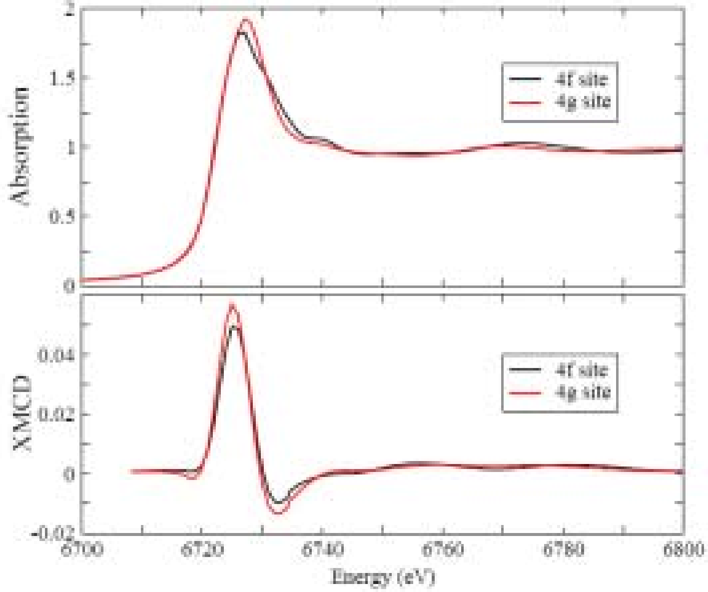
$$\mu_m \propto M \cos\theta$$

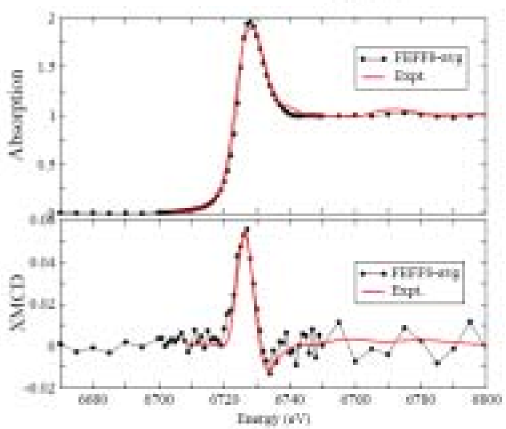


Resonant diffraction can yield same information, but harder to retrieve.
(Intensity depends on real and imaginary parts of f_e, f_m; phase problem).

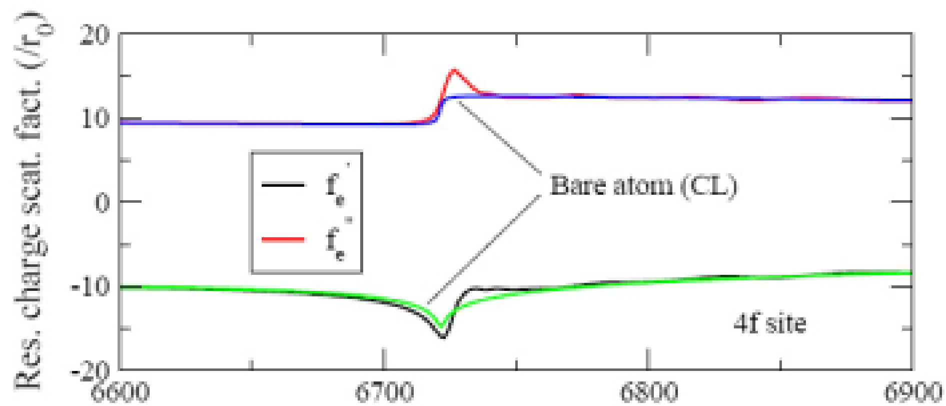
Magnetic spectroscopy in the diffraction channel

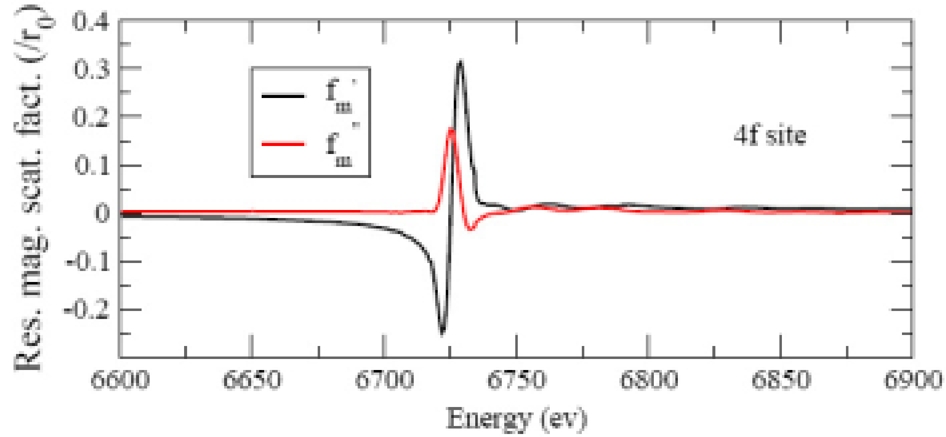
Ab-initio (FEFF8) calculations of μ_e, μ_m for each site separately.





$$\mu(E) \rightarrow f''(E) \rightarrow f'(E)$$





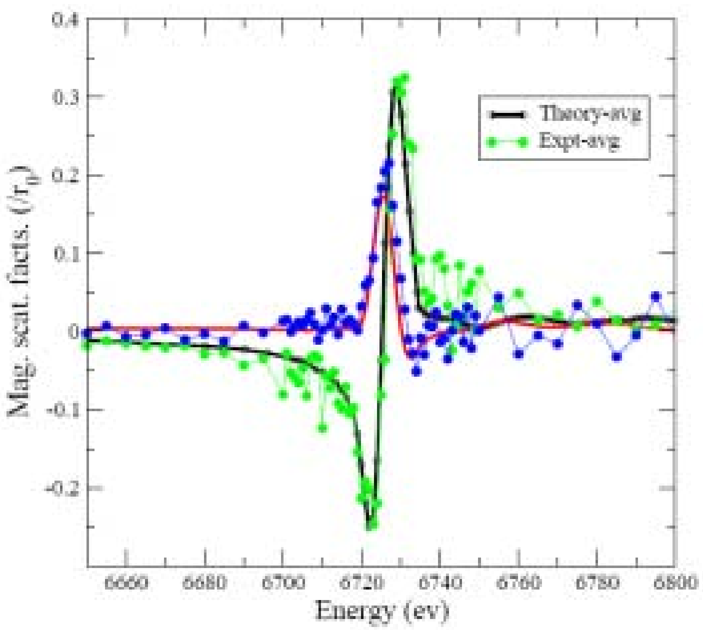
Magnetic spectroscopy in the diffraction channel

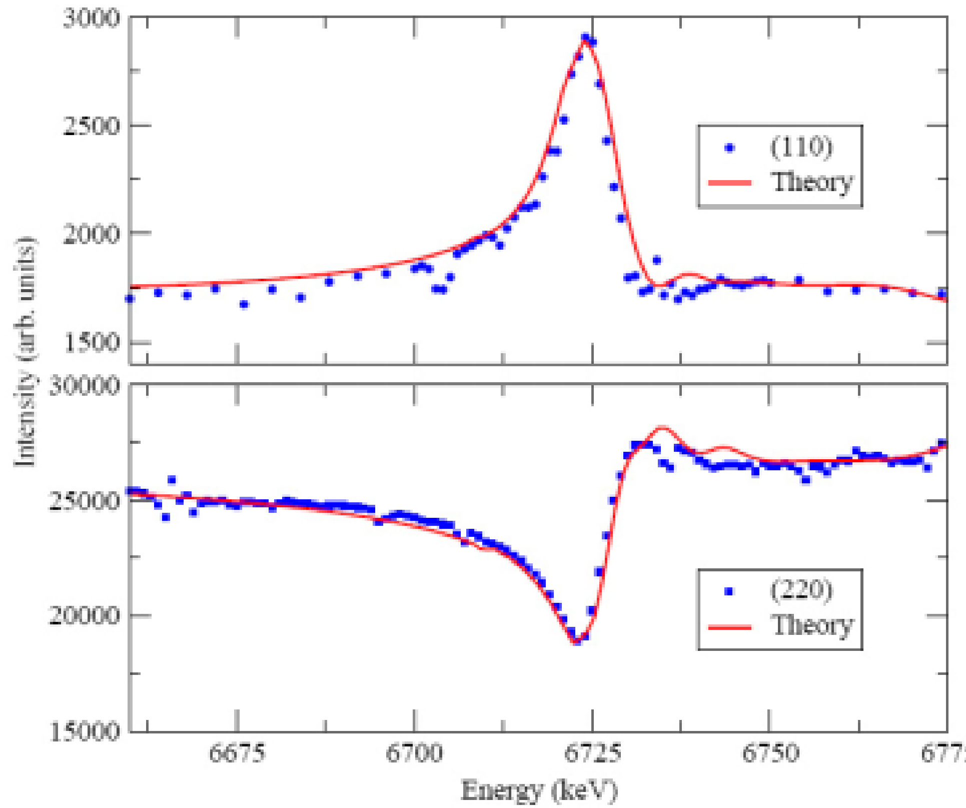
$$F_q = \sum_i (A f_{e,i} + iB f_{m,i}) e^{i\vec{q}\cdot\vec{r}_i}$$

Structure factor calcs with ab-initio res. scat. factors as initial guess; refine.

$$\Delta I_s = 2i \sum_{i,j} (-AB^* f_{m,i}^{\ *} f_{e,j} + A^* B f_{e,i}^{\ *} f_{m,j}) e^{i\vec{q}\cdot(\vec{r}_i - \vec{r}_j)}$$

$$\Sigma I_s = 2 \sum_{i,j} A^2 f_{e,i}^{\ *} f_{e,j} e^{i\vec{q}\cdot(\vec{r}_i - \vec{r}_j)}$$





Outlook and summary

- Diffraction (**Q**) + resonance (E) + CP x-rays (**M**) allows for phase and site specific studies of magnetism in artificially or naturally inhomogeneous systems.

- Combination with ab-initio calculations should yield phase/site specific spectroscopic information, such as SDOS(E_F) and magnetic moments.