Magnetism and X-ray dichroism

A. Rogalev
Introduction to X-ray Magnetic Circular Dichroism

Experimental aspects: ID12 beamline at the ESRF

Selected Results

- Single molecular magnets
- Orbital magnetic moment in actinides

Conclusions
Nobel Prize in Physics 1994: B. N. Brockhouse and C. G. Shull

Press release by the Royal Swedish Academy of Sciences:
“Neutrons are small magnets…… (that) can be used to study the relative orientations of the small atomic magnets. ….. the X-ray method has been powerless and in this field of application neutron diffraction has since assumed an entirely dominant position. It is hard to imagine modern research into magnetism without this aid.”
The Agilent Technologies Europhysics Prize for outstanding achievement in condensed-matter physics in 2000:
P. Carra, G. Schütz and G. van der Laan

“for their pioneering work in establishing the field of magnetic X-ray dichroism. ...it is possible to obtain information about the material that cannot be obtained with traditional measurements.”
Nowadays:
X-ray magnetic circular dichroism (XMCD) is considered to be one of the most important discoveries in the field of magnetism research in the last two decades. It is hard to imagine modern research into magnetism without the aid of X-ray spectroscopy.
“Magnetism, as you recall from physics class, is a powerful force that causes certain items to be attracted to refrigerators.”

- Dave Barry
APPLICATIONS OF PERMANENT MAGNETS

Rare Earth permanent magnets help make technologies more effective and more efficient.

- Magnetic resonance Imaging
- High-Efficiency Motors for Energy-Efficient Homes
- Computing and communication technologies
- Industry
- Clean Energy
- Aerospace
- 2008 Lexus RX Hybrid
  - 1118 magnets (inc. 2.9kg of Nd-Fe-B)
- Direct-drive wind turbines require ~ 600 kg of permanent magnet material to produce 1 megawatt of electric power
Dividing a magnet

~cm

Bar magnets

~10^{-8} \text{ cm}

Atomic magnets
Spin and orbital magnetic moments are coupled via spin-orbit interaction, which is the key ingredient in magneto-optics, magnetocrystalline anisotropy, magnetic chirality, etc.
The experimental technique capable to measure separately SPIN and ORBITAL moments of an atom is **X-ray Magnetic Circular Dichroism (XMCD)**.

Difference in absorption cross-section of circularly polarized X-rays for sample magnetization either parallel or antiparallel to the X-ray wavevector.
Photon could be
- absorbed (photoelectric effect)
- elastically scattered
- inelastically scattered

below 200 keV absorption dominates
XANES

X-ray Absorption Near Edge Structure

EXAFS

Extended X-ray Absorption Fine Structure
In the early days of XAFS, absorption edges taken with use of photographic plates, appeared as unexposed bands on the plate (developed in negative), or “white lines”
Photon could be
- absorbed (photoelectric effect)
- elastically scattered
- inelastically scattered

For magnetism research, the key word - POLARIZATION
**REMINDER: LIGHT AS A EM FIELD**

Wavelength, $\lambda$, the distance between 2 peaks

Electric field, perpendicular to magnetic field

Source

Magnetic field, perpendicular to electric field

Radiant energy propagation direction

\[ A = A_0 e^{i(\omega t - k \cdot r)} \]

\[ E = -iE_0 e^{i(\omega t - k \cdot r)} \quad E_0 = \omega A_0 \]

\[ B = -iB_0 e^{i(\omega t - k \cdot r)} \quad B_0 = k \times A_0 \]

with the wave vector $k$ such that

\[ k^2 = \frac{\omega^2}{c^2} \quad k = \frac{2\pi}{\lambda} \, u = \frac{\omega}{c} \, u \]

When $k$ along $z$: $A_0 = \begin{pmatrix} A_{0x} e^{i\varphi_{0x}} \\ A_{0y} e^{i\varphi_{0y}} \\ 0 \end{pmatrix}$:

- $\varphi_{0x} = \varphi_{0y}$
  - linearly polarized light
- $A_{0x} = A_{0y}$
  - circularly polarized light

$\varphi_{0x} - \varphi_{0y} = \pm 90^\circ$
Polarization vector \( \epsilon = \frac{E}{E_0} \)

\[ \mathbf{k} \parallel z \]

Note: the phase conventions are highly variable.
The first serious approach to the problem of absorption of circularly polarized X-rays

Calculation of the $M_{23}$ magneto-optical absorption spectrum of ferromagnetic nickel

J. L. Erskine*
Department of Physics, University of Illinois, Urbana, Illinois 61801

E. A. Stern†
Department of Physics, University of Washington, Seattle, Washington 98195
(Received 28 April 1975)

The $M_{23}$ magneto-optical absorption spectrum of ferromagnetic nickel is calculated using an approach similar to the component state-density method that has been successfully used in obtaining valence-band emission and absorption x-ray spectra of metals. The $M_{23}$ magneto-optical effects result predominantly from spin-orbit splitting of the $3p$ core state in conjunction with the final $d$-state spin polarization. The calculated spectrum exhibits features that are directly related to electronic structure parameters including the $3p$ core spin-orbit splitting, and the unfilled $d$-band spin polarization. Temperature variations in the magneto-optical structure can be used to determine separately the exchange-splitting variation and spin-wave excitation contributions to the decrease in the magnetization. Experimental verification of these predictions should provide insight into the applicability of the Stoner model to ferromagnetic nickel and may be helpful in resolving some of the apparently conflicting results of other experimental probes of the spin polarization near the Fermi level in nickel.

Two-step model
Absorption of a right circularly polarized photon electric dipolar transitions $p \rightarrow d$ ($\Delta m_1 = +1; \Delta m_S = 0$)

Excited photoelectrons are spin polarized
Exchange splitting of the valence band is driving the second step
First experimental evidence

**XMCD is a new approach to study ferromagnetic system**

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**First experimental observations**


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**Absorption of Circularly Polarized X Rays in Iron**

G. Schütz, W. Wagner, W. Wilhelm, and P. Kienle
Physik Department, Technische Universität München, D-8046 Garching, West Germany

R. Zeller
Institut für Festkörperforschung der Kernforschungsanlage Jülich, D-5175 Jülich, West Germany

and

R. Frahm and G. Materlik
Hamburger Synchrotronstrahlungslabor am Deutschen Elektronen-Synchrotron DESY, D-2000 Hamburg 52, West Germany

(Received 22 September 1986)

The transmission of synchrotron radiation through magnetized iron at energies above the K-absorption edge shows relative differences for right and left circular polarization of several times 10^{-4}. The observed spin dependence of the near-edge photoabsorption is proportional to the difference of the spin densities of the unoccupied bands. In the extended absorption region up to 200 eV above the Fermi level a small spin-dependent absorption is observed and thus is expected to give information on the magnetic neighborhood of the absorbing atom.

PACS numbers: 75.50.Bh, 75.10.Lp, 75.25.+z, 78.70.Dm

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**Rapid Communications**

**Soft-x-ray magnetic circular dichroism at the L_{2,3} edges of nickel**

C. T. Chen, F. Sette, Y. Ma, and S. Modesti
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
(Received 2 March 1990)

Magnetic circular dichroism (MCD) has been observed at the L_{2,3} absorption edges of ferromagnetic nickel by use of circularly-polarized soft-x-ray synchrotron radiation. The MCD intensity ratio between the L_{3} and the L_{2} edges is found to differ appreciably from that predicted by a simple exchange-split-valence-band model. Fine MCD features, imperceptible in the absorption spectra, are also observed and a tentative interpretation is given. This work, demonstrating the feasibility of MCD measurements in the soft-x-ray region, provides a new approach to study 3d and 4f ferromagnetic systems with their respective dipole-permitted 2p\rightarrow 3d and 3d\rightarrow 4f transitions.
Sum rules relate experimental XMCD spectra to the spin and orbital moments:

**Orbital sum rule**

\[
\int (\mu^+ - \mu^-) = \frac{2l(l+1)}{l(l+1)+2-c(c+1)} \times C \times \langle L_z \rangle
\]

**Spin sum rule**

\[
\int (\mu^+ - \mu^-) = \frac{c+1}{c} \int (\mu^+ - \mu^-) = C \times [A\langle S_z \rangle + B\langle T_z \rangle]
\]

**Equation for X-ray absorption cross section per hole**

\[
T = \sum_i (s_i - 3r_i (r_i \cdot s_i)/r_i^2)
\]

\[
A = \frac{l(l+1)-2-c(c+1)}{3c}
\]

\[
B = \frac{l(l+1)[l(l+1)+2c(c+1)+4]-3c(c-1)^2(c+2)^2}{6c \cdot l(l+1)}
\]

**References**

- X-Ray Circular Dichroism and a Probe of Orbital Magnetization
  - B. T. Thole, A. Rogalev, P. Carra, T. Sette, and G. van der Laan
  - European Synchrotron Radiation Facility, BP 220, F-38043 Grenoble CEDEX, France
  - Received 2 December 1991

- X-Ray Circular Dichroism and Local Magnetic Fields
  - Paolo Carra, B. T. Thole, Massimo Altarelli, and Xindong Wang
  - European Synchrotron Radiation Facility, BP 220, F-38043 Grenoble CEDEX, France
  - Department of Chemical Physics, Materials Science Center, University of Groningen, Nijenborgh 16, 9747 AG Groningen, The Netherlands
  - Ames Laboratory and Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011
  - Received 3 July 1992
integrated whitelines intensity is measure for number of holes in the valence band => valence state

APPLICATION OF THE XMCD SUM RULES FOR THE L₂,₃ EDGES

integrated spectra related to spin and orbital magnetic moments in the ground state


\[
\begin{align*}
\langle L_z \rangle &= - \frac{4}{3} \cdot C \cdot (A + B) \\
\langle S_z \rangle - 7\langle T_z \rangle &= -2C \cdot (A - 2B) \\
\frac{\langle L_z \rangle}{\langle S_z \rangle - 7\langle T_z \rangle} &= \frac{2}{3} \cdot \frac{(A + B)}{(A - 2B)}
\end{align*}
\]

in the case of L₃,₂ absorption edges

\[
C = \frac{(n_{h}^{Pt} - n_{h}^{Au})}{A}
\]

A ~ integrated intensity of transitions into unoccupied d band

\[
n_{h}^{d} = \text{number of holes in d band}
\]
ELEMENT SELECTIVITY OF XMCD

**Ni$_2$/Pt$_2$ multilayer**


\[ T \approx 10K \]
\[ H = \pm 5 \text{ T} \]

**RESULTS**

- **Ni magnetic moments:**
  \[ \mu_{S}^{3d} = 0.35 \mu_B/\text{atom} \]
  \[ \mu_{L}^{3d} = 0.038 \mu_B/\text{atom} \]

- **Pt induced magnetic moments:**
  \[ \mu_{S}^{5d} = 0.14 \mu_B/\text{atom} \]
  \[ \mu_{L}^{5d} = 0.03 \mu_B/\text{atom} \]
<S_z> = 0.0353(5)μ_B  <L_z> = 0.0054(5)μ_B (per Au atom)
To compare with 4.15μ_B per Mn atom
SENSITIVITY OF XMCD: A SINGLE SURFACE-ADSORBED ATOM

Ho atoms on a two-monolayer-thick MgO film deposited on Ag(100)

Ho = 0.01 ML

T = 6.5 K


ESRF ID32

STM Image
**Induced Magnetism on Gold Atoms**


Magnetic moment of Au at Au/Co interfaces: A direct experimental determination


European Synchrotron Radiation Facility (ESRF), Route des Particules 22, 38043 Grenoble, France

Department of Physics, Aristotle University of Thessaloniki, 54124 Thessaloniki, Greece

Materials Science Department, University of Patras, 26504 Patras, Greece

Institut de Physique et Chimie des Matériaux de Strasbourg (IPCMS), 2, rue du Loess, 67037 Strasbourg, France

Max Planck Institute for the Physics of Complex Systems, D-01187 Dresden, Germany

Forschungszentrum Karlsruhe, Institute for Nanotechnology, PO. Box 3640, 76321 Karlsruhe, Germany

Received 17 January 2004; revised manuscript received 23 April 2004; published 18 June 2004

![Graph showing magnetic moment of Au at Au/Co interfaces](image)

**Physical Review B** 77, 234414 (2008)

Au and Fe magnetic moments in disordered Au-Fe alloys

F. Wilhelm, P. Pourpoint, V. Kapaklis, J. P. Kaplan, N. Jansen, A. Rogalev, A. N. Yarotski, and C. Politis

European Synchrotron Radiation Facility (ESRF), Route des Particules 22, 38043 Grenoble, France

School of Engineering, Engineering Science Department, University of Patras, 26504 Patras, Greece

Institut de Physique et Chimie des Matériaux de Strasbourg (IPCMS), 2, rue du Loess, 67037 Strasbourg, France

Max Planck Institute for the Physics of Complex Systems, D-01187 Dresden, Germany

Revised manuscript received 27 April 2008; published 9 June 2008

![Graph showing magnetic moments of Au and Fe in disordered Au-Fe alloys](image)


Strong paramagnetism of Gold Nanoparticles Deposited on a Sulfopropyl Acrylamide S Layer


Instituto de Ciencia de Materiales de Aragón (ICMA), CSIC–Universidad de Zaragoza, E-50009 Zaragoza, Spain

Departamento de Física de la Materia Condensada, Universidad de Zaragoza, E-50009 Zaragoza, Spain

Instituto de Nanociencia de Aragón (INMA), Instituto de Nanociencia de Aragón (INMA), Zaragoza, Spain

Institute of Research and Development High Magnetic Field Laboratory, Institute for High Magnetic Fields, Dresden, Germany

Department of Microscopy, University of Graz, E-8010 Graz, Austria

European Synchrotron Radiation Facility (ESRF), BP 220, F-38043 Grenoble, France

Received 28 June 2012; published 16 December 2012

![Graph showing magnetic moment of Au nanoparticles](image)

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**Graphical Data**

- **Au**
  - $\mu_{tot, Au} \approx 0.33 \mu_B/\text{atom}$
  - $\mu_{tot, Au} \approx 0.197 \mu_B/\text{atom}$
  - $\mu_{tot, Au} \approx 0.099 \mu_B/\text{atom}$

- **Au$_3$Fe$_{97}$ Au$_{50}$Fe$_{50}$ Au$_{75}$Fe$_{25}$**
  - $\mu_{tot} \approx 0.051 \mu_B/\text{atom}$

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**Equation**

$$\mu_{tot} \approx \frac{0.031 \mu_B}{\text{atom}}$$
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Conclusions
Quantity to measure: $\Delta \mu = \mu^+ - \mu^-$

$\mu^+, \mu^- \Rightarrow$ Absorption cross-sections for CP X-rays with

(+) helicity \textit{parallel} to the sample magnetization

(-) helicity \textit{antiparallel} to the sample magnetization

- Source of monochromatic circularly polarized X-rays
- Magnetic field to magnetize a sample
- Highly performing X-ray detectors

The best possible at the 3\textsuperscript{rd} generation synchrotron radiation facilities
Full control of polarization: flipping time ~ 5 seconds


B\textsubscript{x} = B\textsubscript{z}

Flux: \(10^{14}\) ph/s/0.1%bw

E\textsubscript{1}\textsuperscript{min}=1.7 keV, E\textsubscript{1}\textsuperscript{max}=6.2 keV

31 periods

\(\lambda\textsubscript{u} = 52\) mm

thg = 6.0 mm

(B\textsubscript{z}=0.493 T)

bhg = 6.0 mm

(B\textsubscript{x}=0.349 T)
High Pressure XMCD 6T; >2.7K

XMCD on nanostructures 6T; >1.5 K

High field XMCD 17 T; > 2K

XDMR
Low field XMCD 0.9 T; >15K

XNCD/XNLD

X-ray Magnetic Scattering
0<Θ_B<18°
0.6 T; >20K

Spot size 1µm(V) x 20µm(H) with Be refractive lenses
over whole spectral range 2-15 keV
### Periodic Table

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<th>IA</th>
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<td>(227)</td>
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#### X-ray Absorption Edges

- **K-edge**
- **L-edges**
- **M-edges**

- 2.05 keV - 15 keV

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**2.05 keV - 15 keV**

<table>
<thead>
<tr>
<th>Element</th>
<th>K-edge Energy</th>
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<td>K</td>
<td>2.05 keV</td>
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<tr>
<td>L edges</td>
<td>2.05 keV - 15 keV</td>
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<tr>
<td>M edges</td>
<td>2.05 keV - 15 keV</td>
</tr>
</tbody>
</table>

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**References:**

- A. Rogalev | X-rays and Magnetism | ESRF | 12 November 2015
H < ± 17 Tesla, T > 2.0 K

Typical sample size
0.65mm X 0.80mmx0.12mm

Aldred et al., PRB10, 1011(1974)
Wilhelm et al., PRB 88, 024424 (2013)
X-ray Magnetic Circular Dichroism is a unique tool to study microscopic magnetic properties

- Element-specific and orbital-selective magnetometry tool
- Sensitive to the electronic structure (valence state, symmetry,…)
- Possibility to extract Spin and Orbital magnetic moments of absorbing atoms only
- Small size samples (focusing the x-ray beam)
- Single crystals, polycrystalline and amorphous materials, thin films, nanoparticles, monolayers, ad-atoms, …
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MAGNETIC STRUCTURES

**classical**
- permanent magnets

**nanoscopic**
- micro-particles
- nano-particles
- clusters

**quantum**
- Paramagnets and Single-molecule magnets

\[ \tau(T) = \tau_0 \exp\{\Delta_{eff} / (k_B T)\} \]

\[ \hat{H} = D \hat{S}_Z^2 \]

**Mn\textsubscript{12}ac**

Low temperature!
\[ \lambda = \frac{1}{2} m_e Z^4 \alpha^4 c^2 \frac{1}{n^3 l(l + \frac{1}{2})(l + 1)} \]
Sr$_2$IrO$_4$

Electronegativity
Mass and size
Redox-innocence

….

Ir$_6^{8-}$

$J_{eff} = 1/2$ Mott-Insulating State in Rh and Ir Fluorides

Turan Birol and Kristjan Haule

Department of Physics and Astronomy, Rutgers University, Piscataway, New Jersey 08854, USA

(Received 17 August 2014; published 5 March 2015)

Kim et al. Science, 2009, 323, 132
Machida et al. Nature 2010, 463, 210

Chun et al. Nat. Phys. 2015, 11, 462
Chen et al. Nat. Commun. 2015, 6, 6593
Kim et al. Nat. Phys. DOI: 10.1038/NPHYS3503
Zhao et al. Nat. Phys. DOI: 10.1038/nphys3517
GETTING SOME $[\text{MF}_6]^\text{x-}$

$[\text{ReCl}_6]^2-$ $\xrightarrow{\text{NH}_4\text{HF}_2(l)}$ $[\text{ReF}_6]^2-$

SOME INDICATIONS OF ORBITAL MAGNETISM

\[ \chi T = \frac{g^2}{8} S(S+1) \]
XMCD OF $[\text{IrX}_6]^2-$ COMPLEXES

Iridates from the molecular side

Kasper S. Pedersen$^{1,2,3,4}$, Jesper Bendix$^5$, Alain Tressaud$^{3,4}$, Etienne Durand$^{3,4}$, Hagni Weihe$^5$, Zaher Salman$^6$, Thorbjørn J. Morsing$^5$, Daniel N. Woodruff$^7$, Yanhua Lan$^8$, Wolfgang Wernsdorfer$^5$, Corine Mathonière$^{3,4}$, Stengios Piligkos$^5$, Sophia I. Klokishner$^9$, Serghei Ostrovsky$^9$, Katharina Ollefs$^{10,11}$, Fabrice Wilhelm$^{10}$, Andrei Rogalev$^{10}$ & Rodolphe Cléroux$^{1,2}$

\[
\langle S_z \rangle \propto L_3^{\text{XMCD}} + L_2^{\text{XMCD}}
\]

\[
\langle S_{\text{eff}} \rangle \propto L_3^{\text{XMCD}} - 2 \times L_2^{\text{XMCD}}
\]

$\begin{array}{ccc}
[\text{IrF}_6]^2- & [\text{IrCl}_6]^2- & \text{Sr}_2\text{IrO}_4 \\
\langle S_z \rangle & 0.13 & 0.15 & 0.15 \\
\langle L_z \rangle & 0.77 & 0.65 & 0.63 \\
\end{array}$
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Laves phase, C-15 structure, fcc unit cell

$T_c$'s between 160 K (UFe$_2$) and ~700 K (AmFe$_2$)

Easy magnetization direction: $<111>$ (U,Np) or $<100>$ (Pu,Am)
dipolar transitions
\( \Delta l = \pm 1; \Delta s = 0; \Delta j = 0; \pm 1 \)

\[
\Delta \ell = \pm 1; \Delta s = 0; \Delta j = 0; \pm 1
\]

\[
dl = \pm 1, \quad \Delta s = 0, \quad \Delta j = 0, \pm 1
\]

\[
\hbar \omega = \pm 1; \quad \Delta s = 0; \quad \Delta j = 0; \pm 1
\]
Intensity of the $M_5$ XANES spectra decreases from U to Cm.

Intensity of the $M_4$ XANES spectra decreases from U to Pu but increases for Cm.

**SPIN-ORBIT SUM RULE:**

\[
< l.s > = -\frac{3}{4}n_h(2I_{M5} - 3I_{M4})/(I_{M5} + I_{M4}) + \Delta
\]

\[
= \frac{3}{2} n_{7/2} - 2 n_{5/2}
\]

\[\Delta = -0.014, -0.010, -0.005, 0.000, +0.005, +0.015\]

for $n_e^{5f} =$ 2, 3, 4, 5, 6, 7

APPLICATION OF THE SPIN-ORBIT SUM RULE

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<tr>
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<th>$n_e^{5f}$</th>
<th>$\frac{2}{3}\langle l.s. \rangle$</th>
<th>$n_{5/2}$</th>
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XANES spectra were measured on

- UO$_2$ for ionic state of U close to $5f^2$ configuration
- U/Fe multilayers, UFe$_2$ for $5f^3$ configuration
- NpFe$_2$ for $5f^4$ configuration
- PuFe$_2$ for $5f^5$ configuration
- AmFe$_2$ for $5f^6$ configuration
- Cm metal for $5f^7$ configuration

Uncertainty of a few %
APPLICATION OF THE SPIN-ORBIT SUM RULE

Experiment: EELS and XAS (K.T. Moore and G. van der Laan)

5f states are well described with intermediate coupling scheme
X-RAY ABSORPTION SPECTROSCOPY: SPIN-ORBIT SUM RULE

dipolar transitions
\( \Delta l = \pm 1; \Delta s = 0; \Delta j = 0; \pm 1 \)

\[ \hbar \omega \]

5f spin-orbit splitting

\( \Delta l = \pm 1; \Delta s = 0; \Delta j = 0; \pm 1 \)

US
single crystal

Normalized absorption (a.u.)

0.8 eV

\[ M_5 (3d_{5/2} \rightarrow 5f_{7/2}) \]

\[ M_4 (3d_{3/2} \rightarrow 5f_{5/2}) \]
- **XMCD spectral shape at the $M_5$-edge** has an asymmetric S shape for light actinides (U-Am) but becomes symmetric for Curium metal.

- **XMCD spectral shape at the $M_4$-edge** has slight asymmetry on high energy side and negative for light actinides but changes the sign for Curium metal.
ferromagnet with $T_c = 160$ K

$\mu_{Fe} = 0.58 \mu_B$ and $\mu_U \sim 0 \mu_B$

<table>
<thead>
<tr>
<th>XMCD</th>
<th>$\mu_L (\mu_B)$</th>
<th>$\mu_S (\mu_B)$</th>
<th>$-\mu_L / \mu_S$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Neutron</td>
<td>$0.21 \pm 0.02$</td>
<td>$-0.20 \pm 0.02$</td>
<td>$0.97 \pm 0.05$</td>
</tr>
<tr>
<td>Theory</td>
<td>$0.23 \pm 0.01$</td>
<td>$-0.22 \pm 0.02$</td>
<td>$1.05 \pm 0.05$</td>
</tr>
</tbody>
</table>

$UFe_2$

$U-M_{IV,V}$ edges

$T = 20$ K

$H = 2$ T
<T_z> is a measure of a spin moment anisotropy induced either by a charge quadrupole moment or by the spin-orbit interaction.

There are no any direct measurements of this term (so far !!!)

One can estimate <T_z> via combination of XMCD with Neutron scattering, magnetic Compton scattering or SQUID measurements.

Sum rules analysis:

\[ \langle L_z \rangle = -0.44(5) \]
\[ \langle S_z \rangle + 3\langle T_z \rangle = -0.135(15) \]
\[ \langle J_z \rangle = \langle L_z \rangle + \langle S_z \rangle = 0 \]

\[ \mu_L = -\langle L_z \rangle = +0.44 \mu_B \]
\[ \mu_S = -2\langle S_z \rangle = -0.88 \mu_B \]
\[ 3\langle T_z \rangle = -0.57 \]

Calculated: (with \( H_{\text{int}} = 180 \) T)
\[ \mu_L = -\langle L_z \rangle = +0.47 \mu_B \]
\[ \mu_S = -2\langle S_z \rangle = -0.94 \mu_B \]
\[ 3\langle T_z \rangle = -0.51 \]
Both are ferromagnets

- UIrAl ($\mu_{\text{TOT}} = 0.98 \mu_B$) $T_C=64K$
- UPtAl ($\mu_{\text{TOT}} = 1.38 \mu_B$) $T_C=43K$

Isotropic spectra are similar at M5-edge

M4-edge XANES shows that there are more 5f\(_{5/2}\) holes in UIrAl

Different expectation value of the 5f spin-orbit interaction per hole

U valence state in UIrAl seems to be U\(^{4+}\) whereas in UPtAl it is U\(^{3+}\)

Exp. error bars \(\sim\)%

<table>
<thead>
<tr>
<th></th>
<th>B</th>
<th>(2&lt;s&gt;/3.n_h^{5f}) - (\Delta)</th>
<th>(n_e^{5f})</th>
<th>(n_{5/2})</th>
<th>(n_{7/2})</th>
</tr>
</thead>
<tbody>
<tr>
<td>UIrAl</td>
<td>0.654</td>
<td>-0.135</td>
<td>2 (U(^{4+}))</td>
<td>1.62</td>
<td>0.38</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>3 (U(^{3+}))</td>
<td>1.96</td>
<td>1.04</td>
</tr>
<tr>
<td>UPtAl</td>
<td>0.692</td>
<td>-0.230</td>
<td>2 (U(^{4+}))</td>
<td>2.11</td>
<td>-0.11</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>3 (U(^{3+}))</td>
<td>2.42</td>
<td>0.58</td>
</tr>
</tbody>
</table>

for \(n_e^{5f} = 2\)
\(\Delta = -0.014\)

for \(n_e^{5f} = 3\)
\(\Delta = -0.010\)
Strong XMCD at the M4-edge

s-like shape XMCD at the M5-edge

Element specific magnetization curves recorded at U similar to the macroscopic one
XMCD at the Ir L$_{2,3}$-edges in UIrAl crystal

- Strong XMCD at the L$_3$-edge
- Small XMCD at the L$_2$-edge
- Large Ir 5d orbital moment aligned parallel to the spin

\[
\begin{array}{|c|c|c|c|}
\hline
\mu_{L}^{Ir}(5d) & \mu_{S}^{Ir}(5d) & \mu_{tot}^{Ir}(5d) & \mu_{L}^{Ir}(5d)/\mu_{S}^{Ir}(5d) \\
(\mu_{B}/\text{atom}) & (\mu_{B}/\text{atom}) & (\mu_{B}/\text{atom}) & \\
\hline
0.028 & 0.048 & 0.076 & 0.60 \\
\hline
\end{array}
\]
• \( M^U(5f) = 0.92 \mu_B / U \) atom for \( n_f=2 \) (\( U^{4+} \))

• \( M^U(5f) = 0.62 \mu_B / U \) atom for \( n_f=3 \) (\( U^{3+} \))

• \( M^{Ir}(5d) = 0.076 \mu_B / Ir \) atom (sum over two Ir sites)

\[
M_{total} = M^U + M^{Ir} = 0.996 \mu_B
\]

Al and U(6d) contributions are neglected

VSM Data: \( M_{total} = 0.98 \mu_B \) at 6 Tesla and 4.2K
XMCD is very powerful spectroscopy tool to unravel the microscopic origin of magnetism.
Thank you for your patience and your attention!