

High Magnetic Field X-ray Magnetic Circular Dichroism in Valence Fluctuating Compounds

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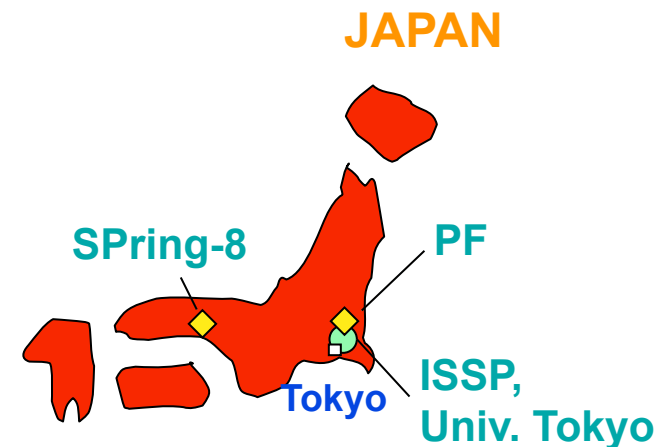
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→HUST, China)

Toshiya INAMI (JAEA / SPring-8)

Kenji OHWADA (JAEA / SPring-8)

Jim-Long HER (ISSP, Univ. Tokyo)



Outline

1. Introduction

Why X-ray experiments in high magnetic fields ?

2. A compact capacitor bank and a mini-magnet

High field X-ray diffraction and X-ray absorption spectroscopy

3. XMCD of valence fluctuating compounds at the SPring-8

$\text{EuNi}_2(\text{Si}_{1-x}\text{Ge}_x)_2$, EuNi_2P_2 , (YbInCu_4)

4. High-magnetic-field DXAFS system at the Photon Factory

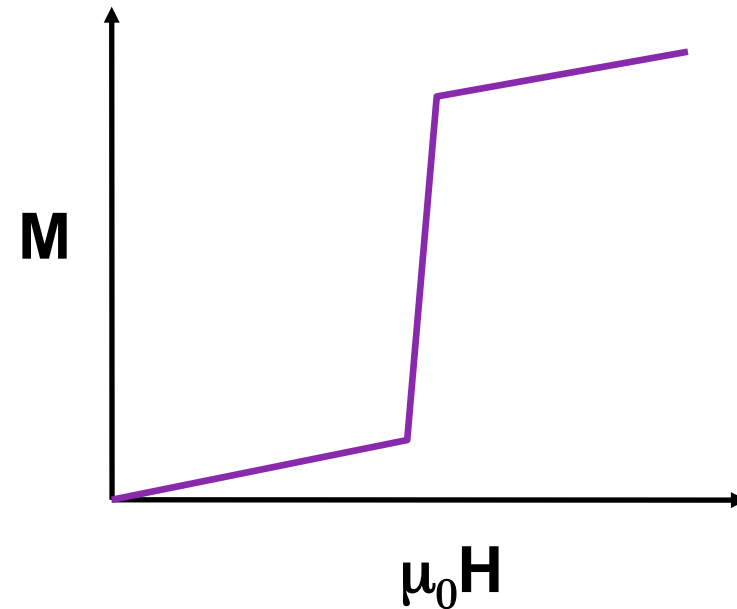
$\text{Pr}_{0.6}\text{Ca}_{0.4}\text{MnO}_3$

High magnetic fields

- Zeeman splitting
- Landau quantization
- Wave function shrinkage



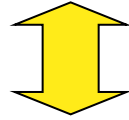
Phase transition
Quantum Phenomena



Synchrotron X-rays
Crystal Structure
Electronic States

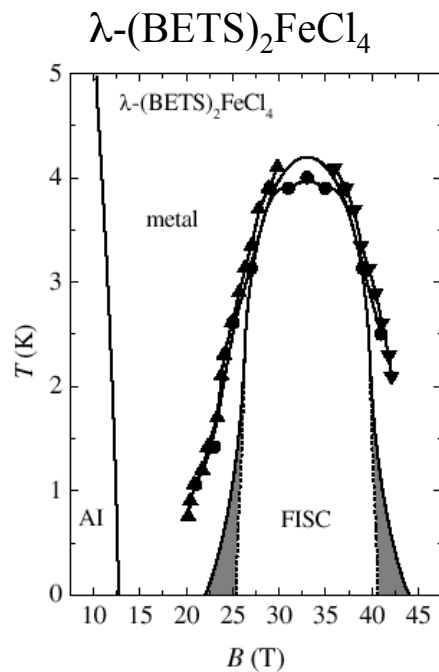
Diffraction
Spectroscopy

DC superconducting magnet ; $B_{\max} = 15 \text{ T}$

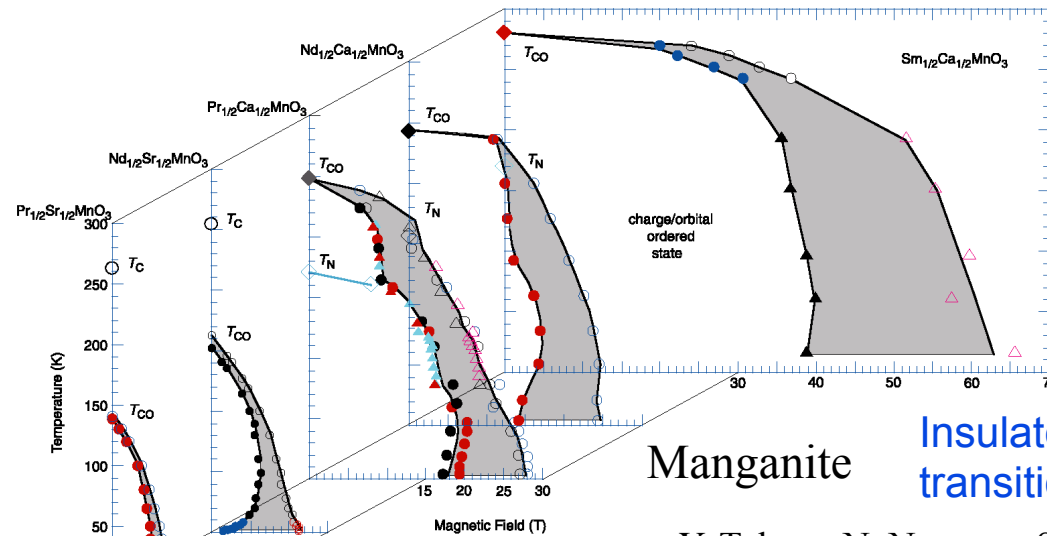


We have many interesting phenomena in **40 T** range.

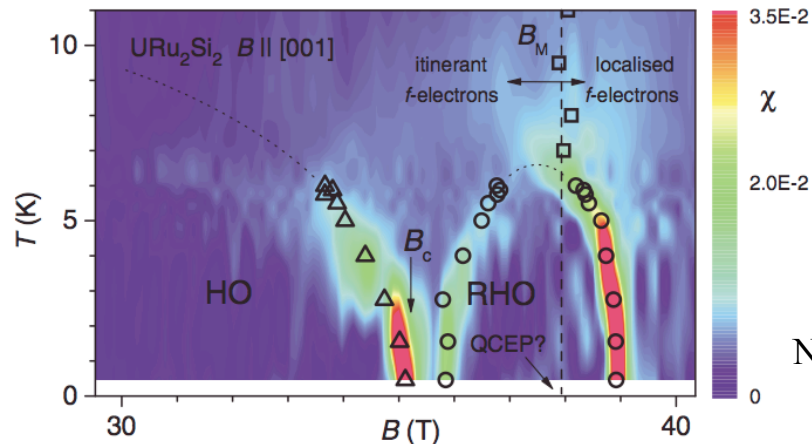
Field-induced superconductivity



Balicas *et al.*,
Phys. Rev. Lett. **87**
(2001) 067002



Y. Tokura, N. Nagaosa, *Science* (2000)

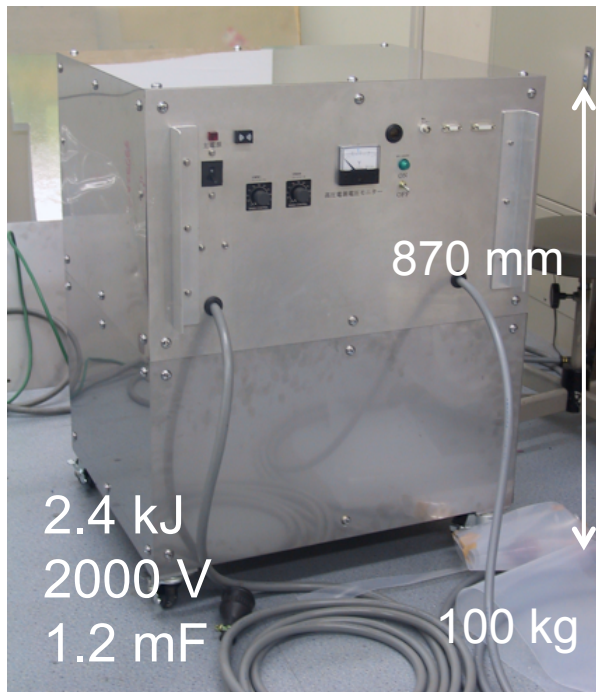


Quantum phase transition

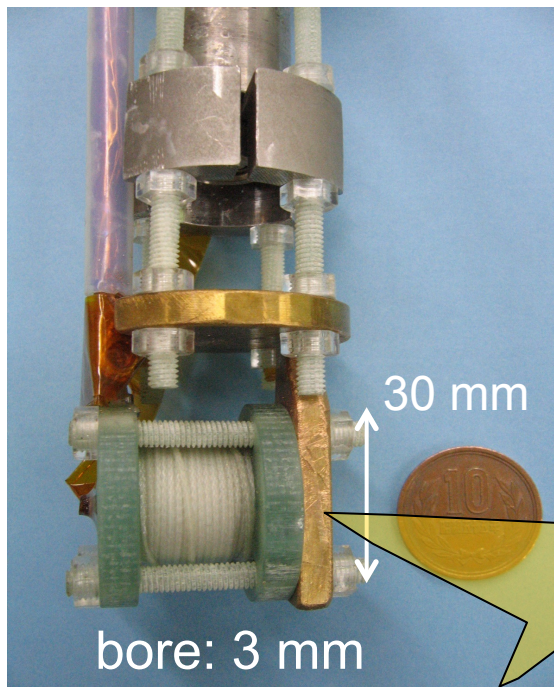
URu_2Si_2

N. Harrison *et al.*, *PRL* 90 (2003)

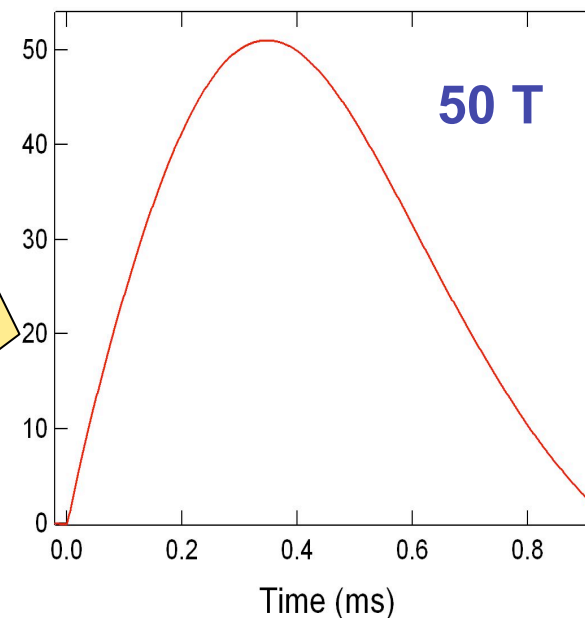
Miniature Pulsed Magnet and Portable Capacitor Bank



Small Energy



Small Space



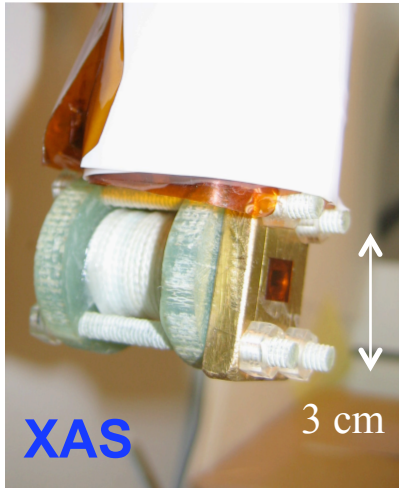
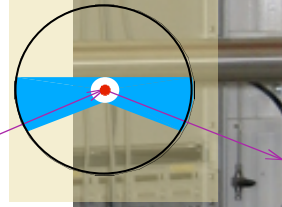
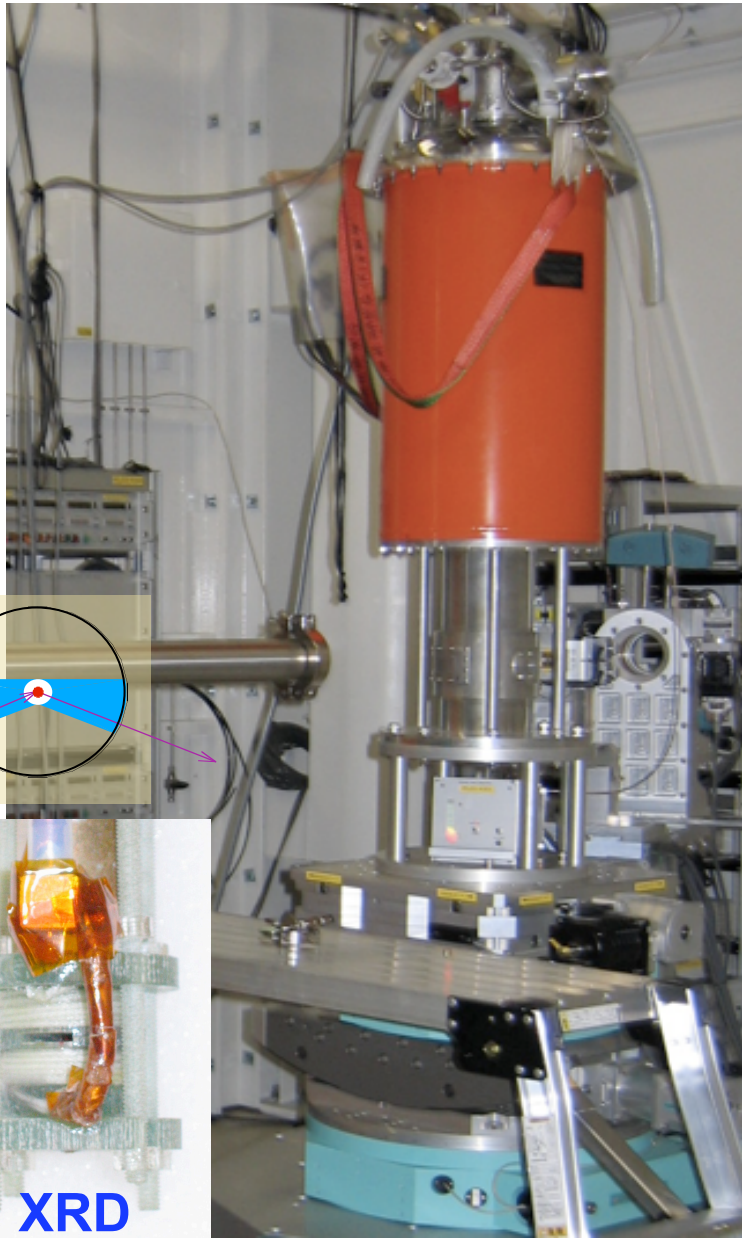
High Magnetic Field

Y. H. Matsuda et al., *Physica B* **346-347** (2004) 519-523.

Y. H. Matsuda et al., *Nuclear Instruments and Methods in Physics Research A* **528** (2004) 632-635.

High Magnetic Field Experiments

at **SPring-8**



◆ Low Temperature

Closed cycle He-gas refrigerator ($T > 10$ K)

He flow cryostat (Orange Cryo.) ($T > 2$ K)

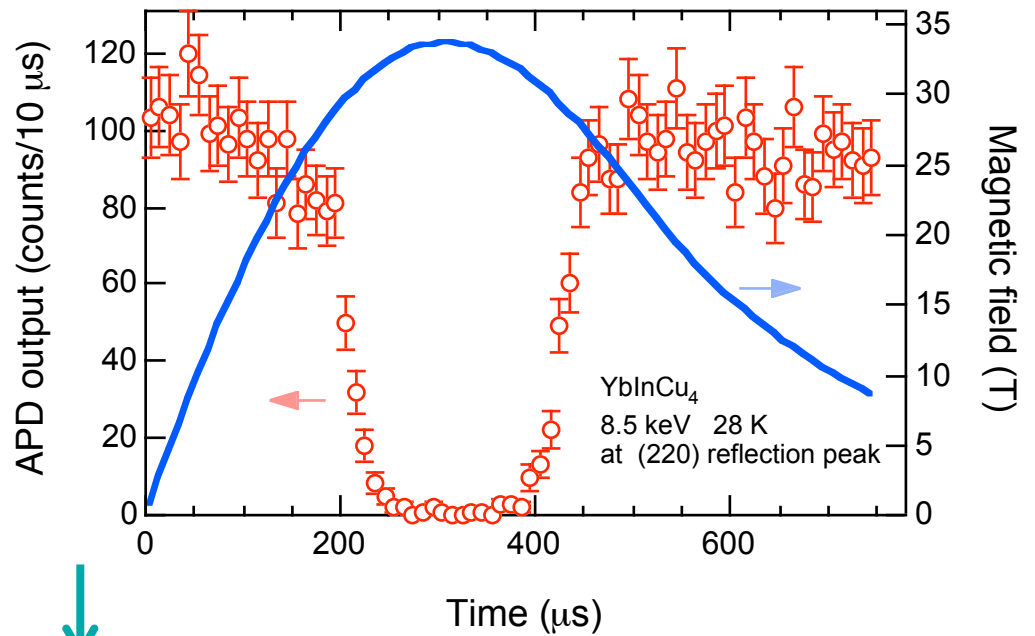
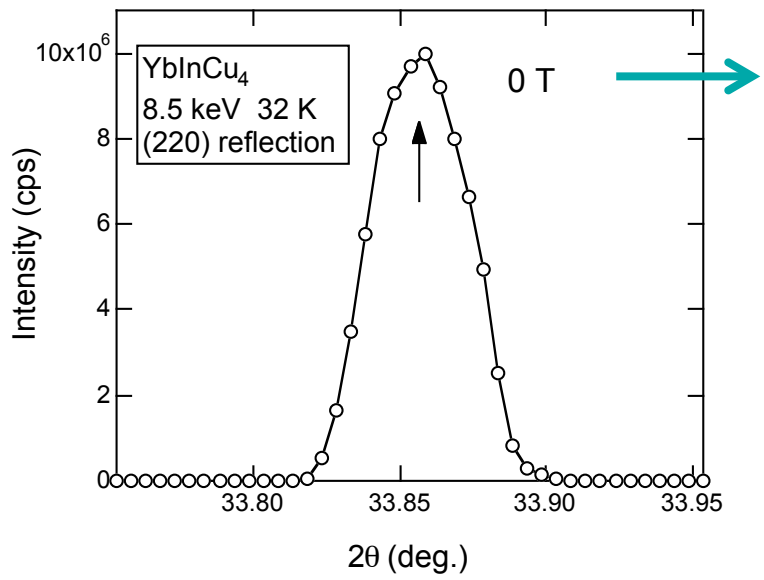
◆ Detection

APD + MCS

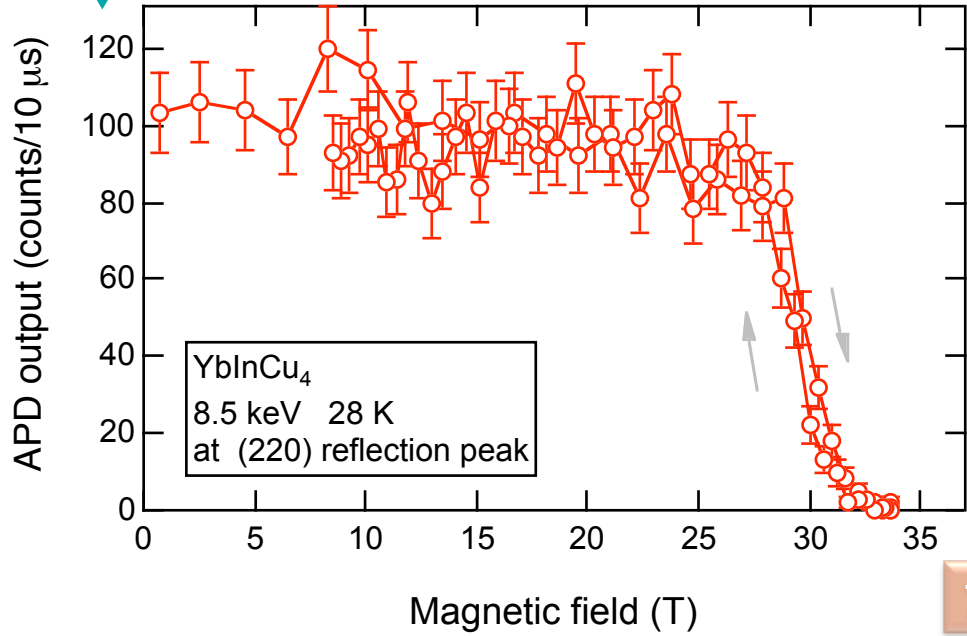
PIN + Oscilloscope

2D detector (Hamamatsu C7942)

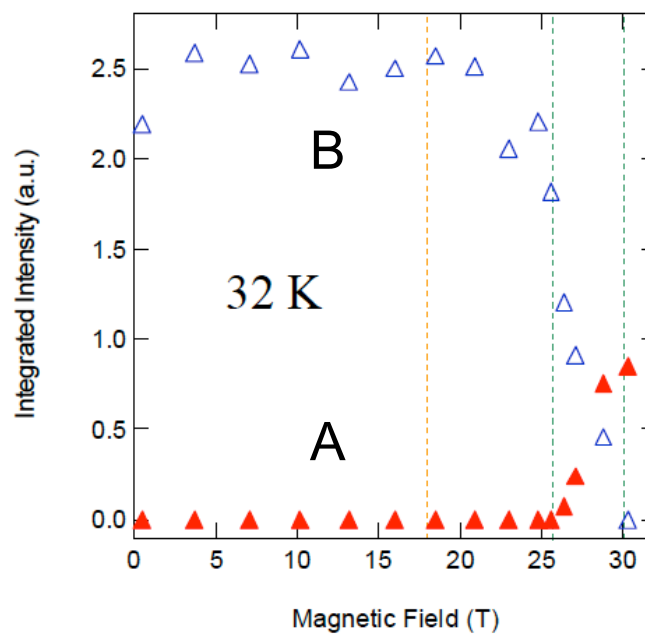
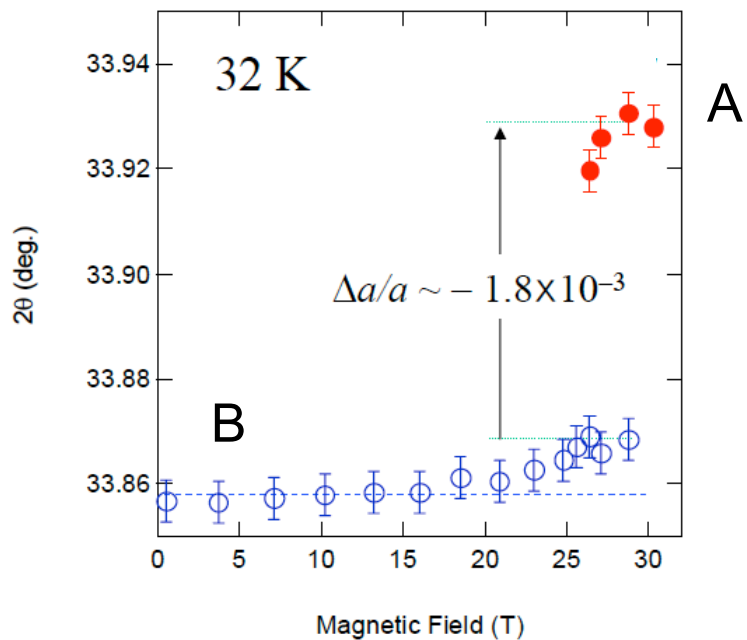
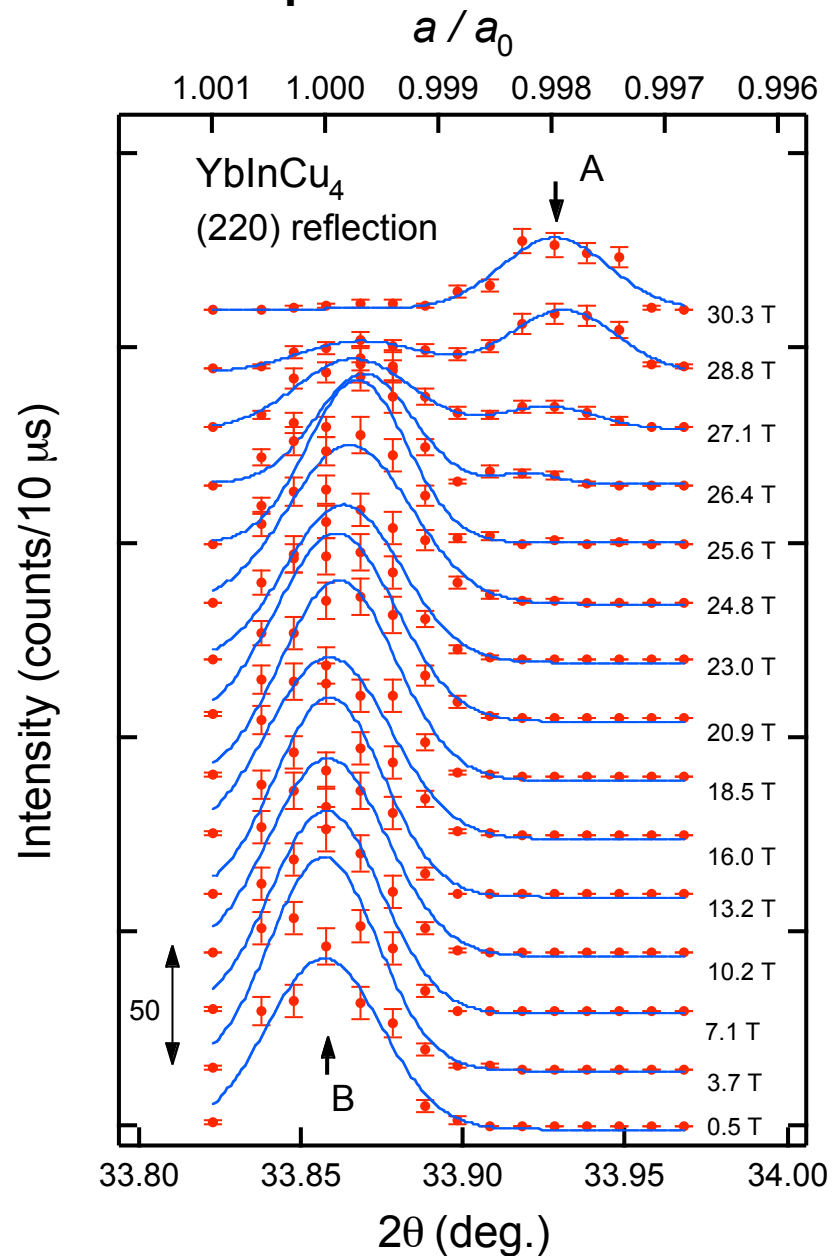
X-ray diffraction measurement



Field variation of the Bragg reflection intensity can be measured by only one shot.

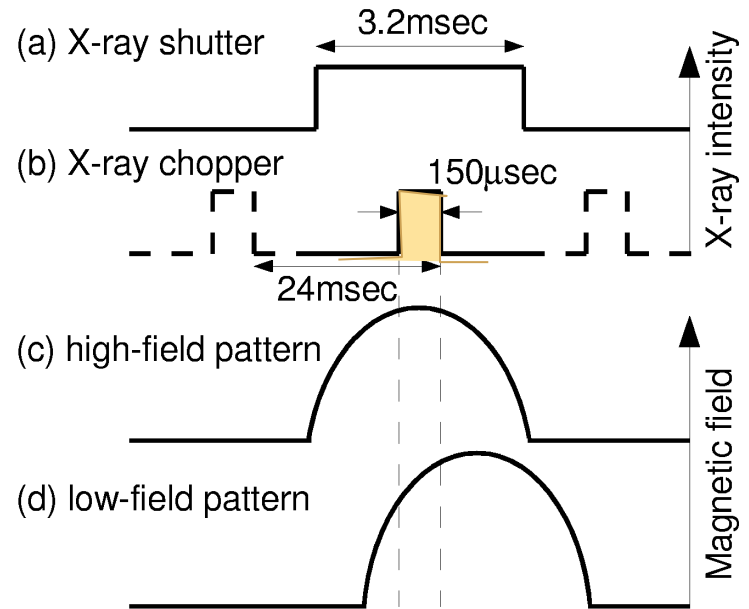
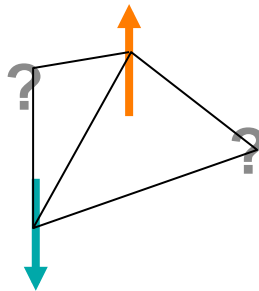
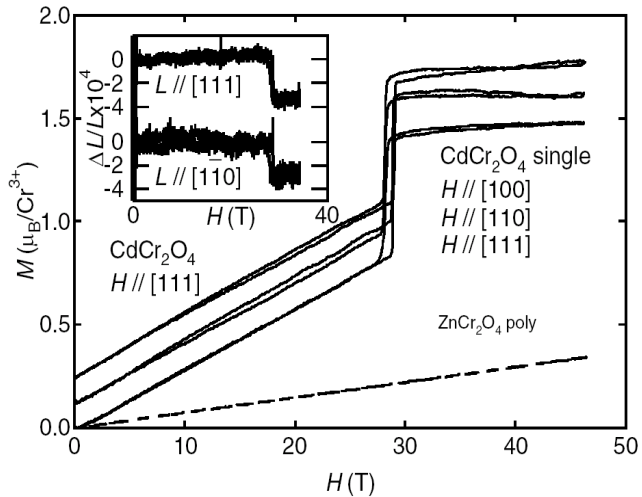


YbInCu₄



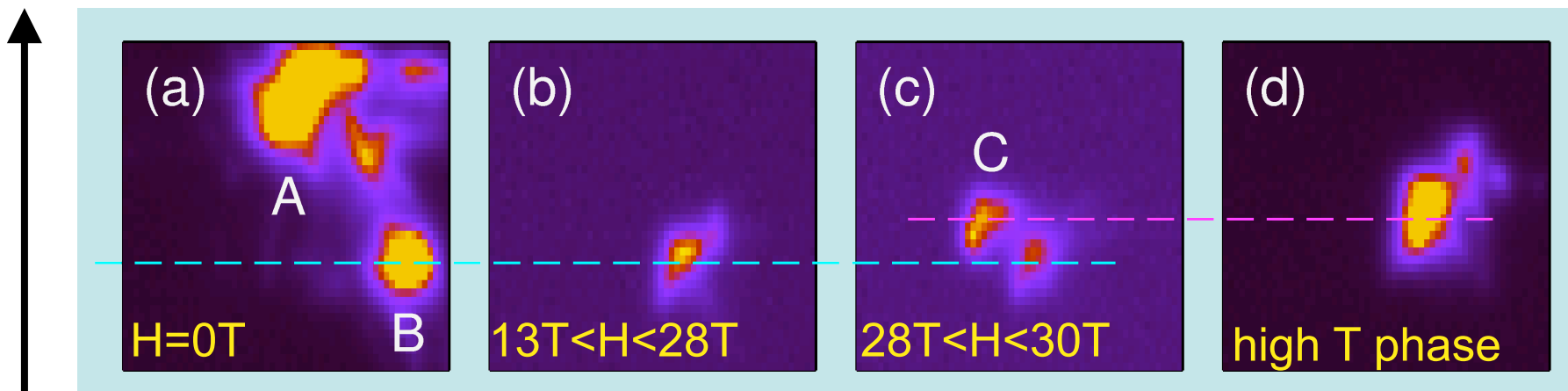
2D-detector

CdCr₂O₄ $T_N=8K$ $\Theta_{CW}=-70K$



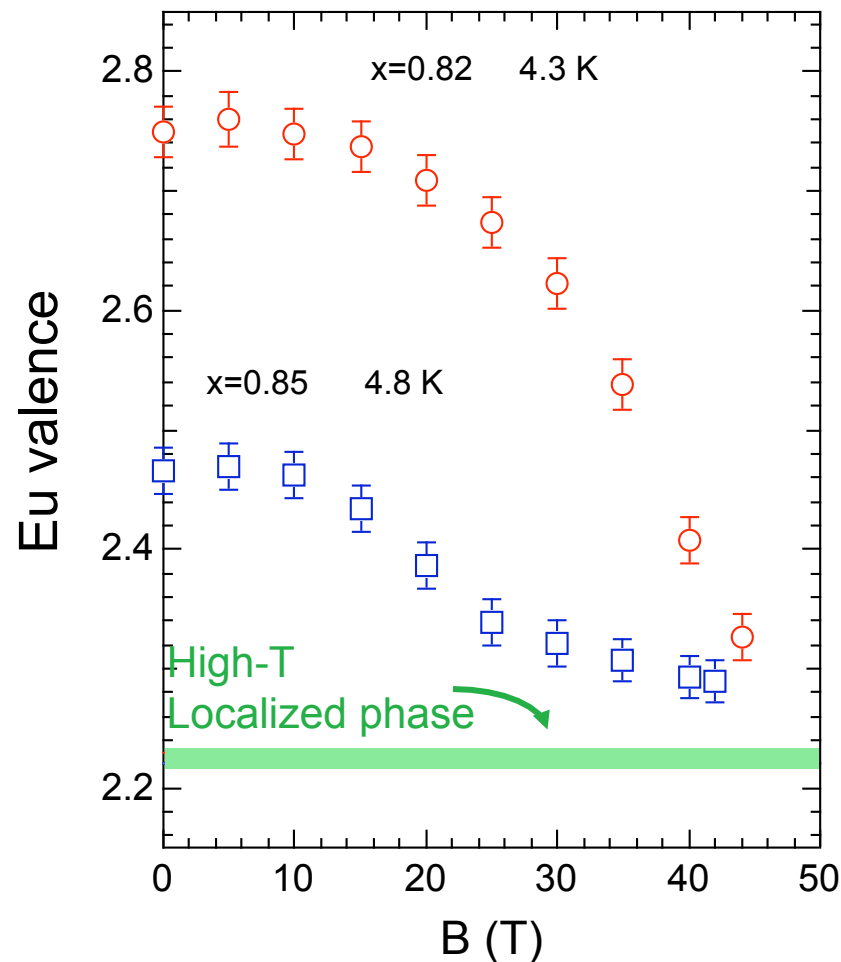
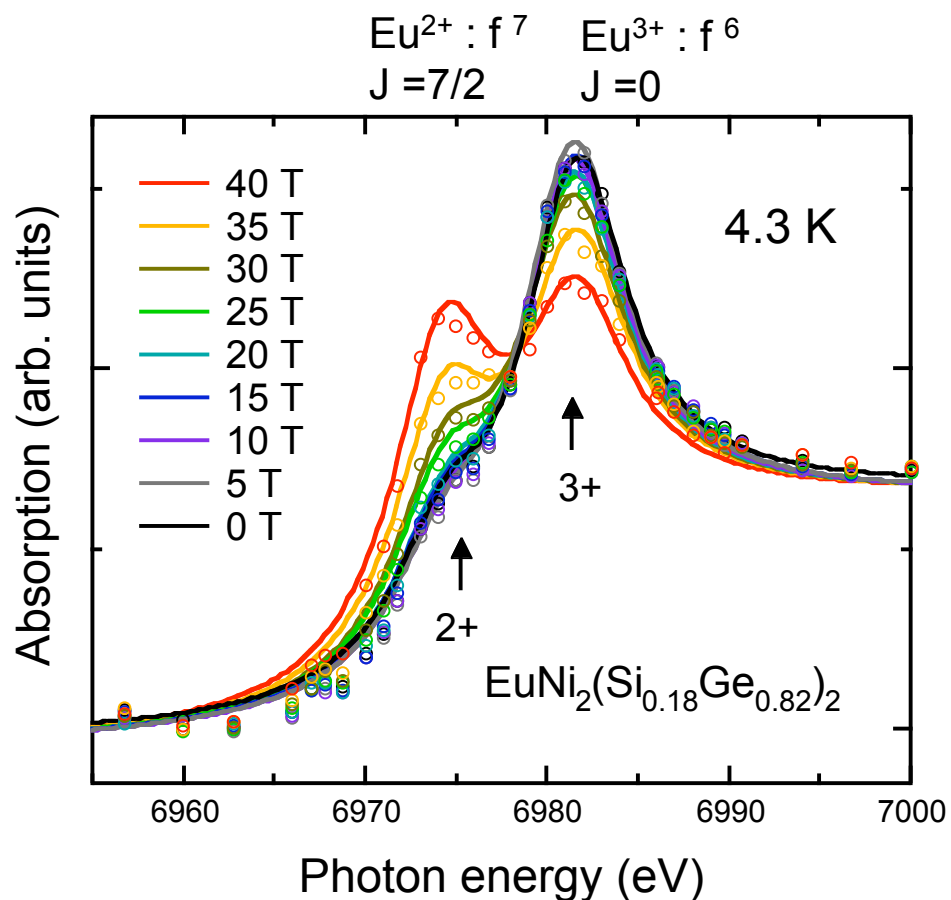
H. Ueda et al., PRL 94 (2005) 047202

2θ 440 reflection ($T=5K$)



X-ray Absorption Spectroscopy

$\text{EuNi}_2(\text{Si}_{1-x}\text{Ge}_x)_2$



Y. H. Matsuda et al., *J. Phys. Soc. Jpn.* **77** (2008) 054713 1-7.

Y. H. Matsuda et al., *J. Phys.: Conference Series* **51** (2006) 490-493

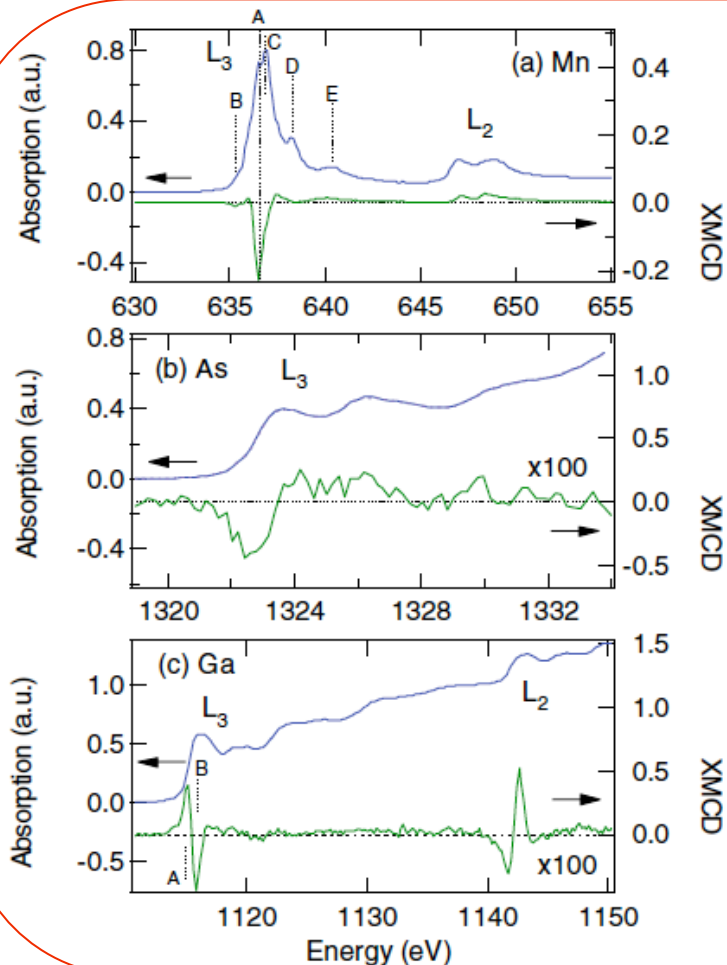
XMCD

$$\Delta\mu = \mu_+ - \mu_-$$

Element and shell selective
microscopic probe for magnetism



Powerful means for
investigation of
electronic states



GaMnAs

D. J. Keavney et al.,
PRL91, (2003)
187203

Most studies were
for **ferromagnetic**
materials

using **High B**

Field induced phenomena
in **antiferromagnetic** and
paramagnetic materials

Valence Fluctuation

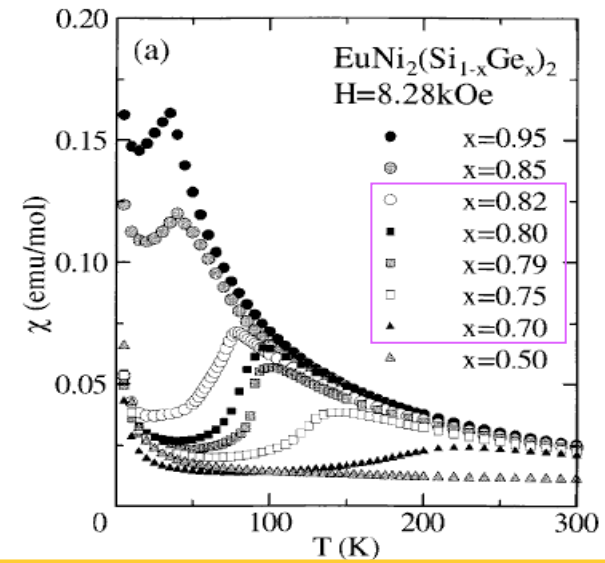
Rare-earth
Intermetallic Compound



Ce (f^1, f^0), Sm (f^6, f^5),
Eu (f^7, f^6), Yb (f^{14}, f^{13})

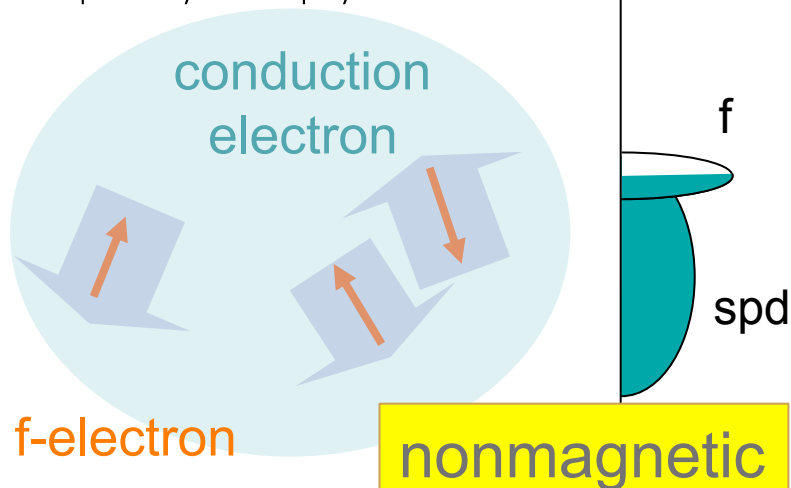
Eu²⁺ $J=7/2$
Eu³⁺ $J=0$

H. Wada et al.,
J. Phys.: Condens.
Matter. **9** (1997)
7913

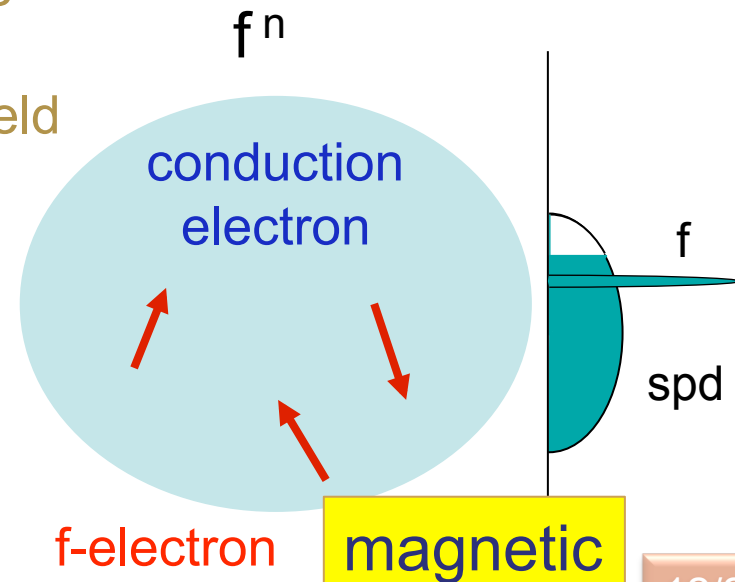


Low T Valence fluctuating state

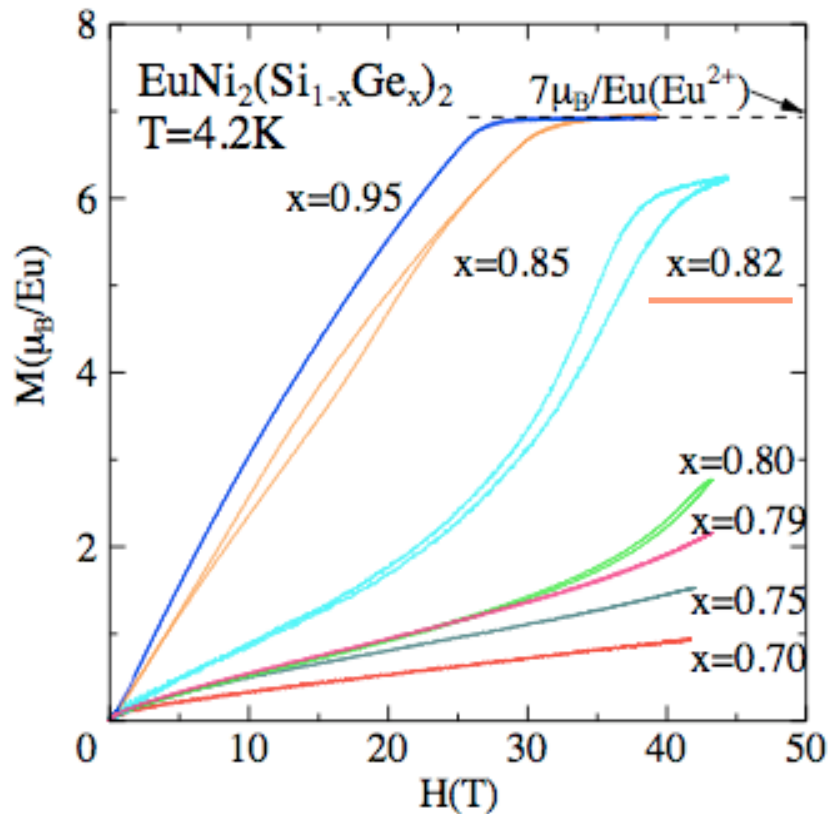
$$\alpha |n-1\rangle + \beta |n\rangle$$



Temperature
Pressure
Magnetic Field

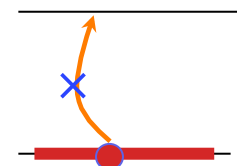


Field-induced valence transition studied by XMCD



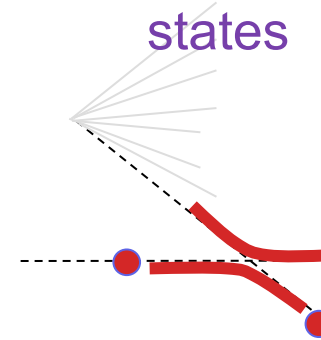
no thermal excitation

Low *T*



control of electronic states

High *B*

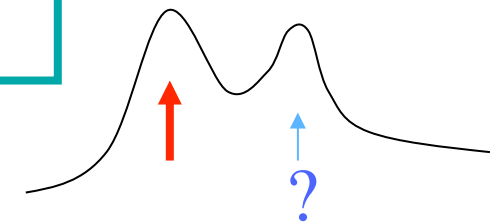


J = 7/2

J = 0

valence selectivity using x-rays

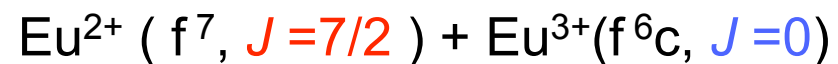
$$\alpha|f^7\rangle + \beta|f^6c\rangle$$



Magnetic polarization

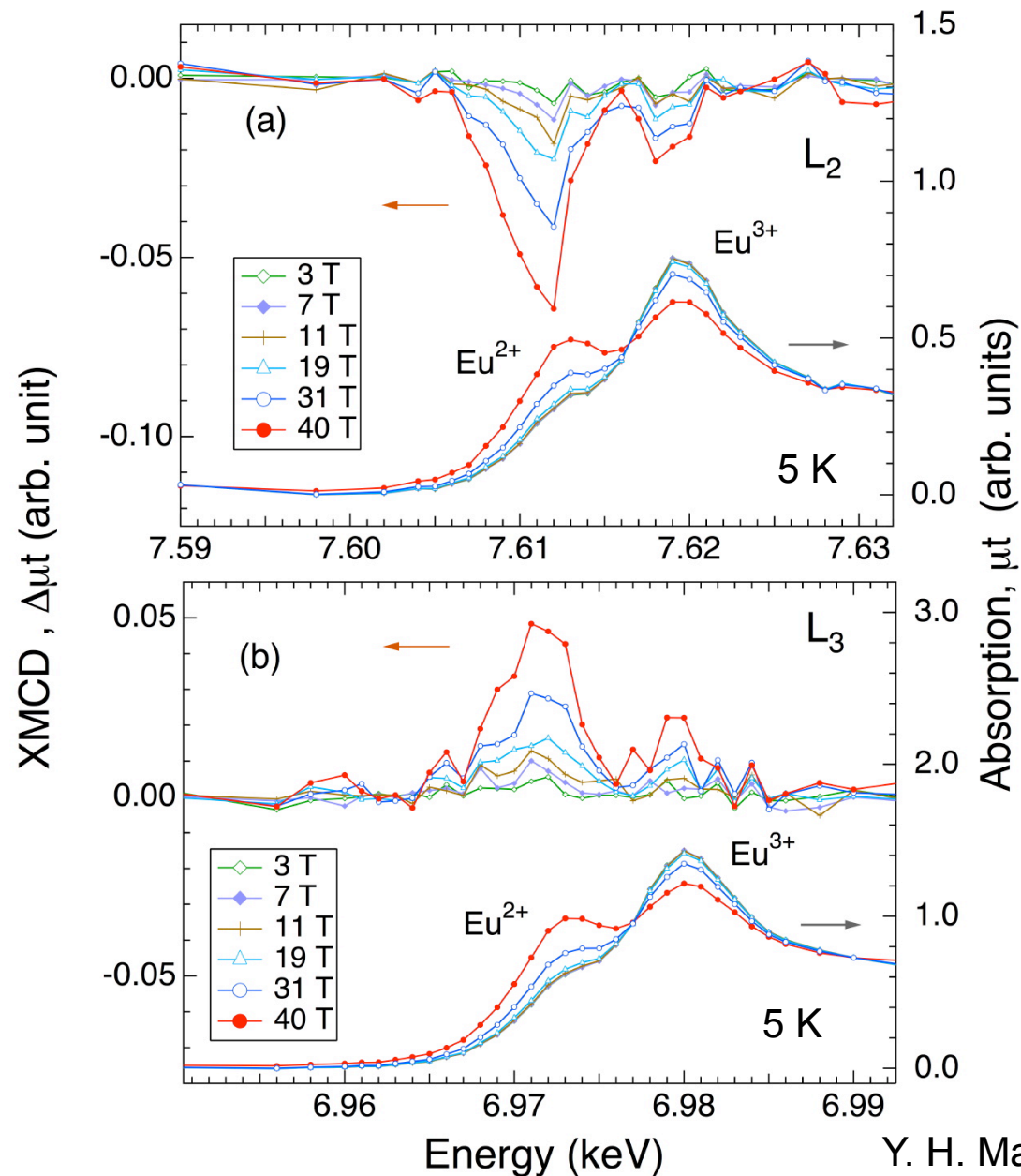


Hybridization



A. Mitsuda, thesis (1999) Kyoto Univ.
H. Wada, A. Nakamura, A. Mitsuda et al.,
J.Phys.:Condens. Matter 9 (1997) 7913.

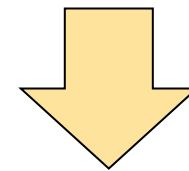
XMCD of $\text{EuNi}_2(\text{Si}_{0.18}\text{Ge}_{0.82})_2$



Double peak structure

Eu^{3+} (f^6 , $J=0$)

Eu^{2+} (f^7 , $J=7/2$)



L-edge $2p \rightarrow 5d$

5d electrons of Eu
are magnetically
polarized

Magnetic Field Dependence of the XMCD intensity

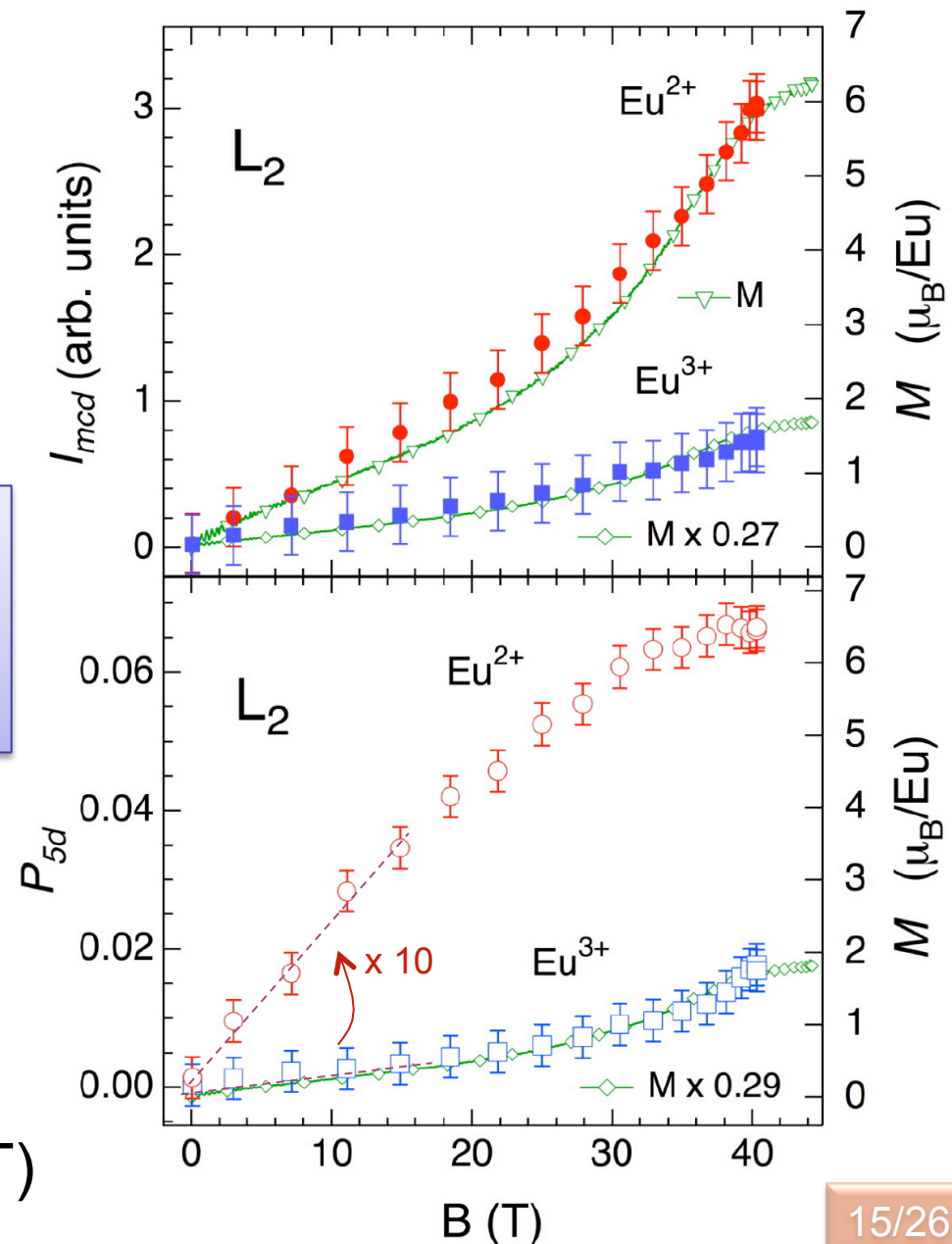
Integrated XMCD peaks I_{mcd} scale together with the magnetization curve.

Degree of the magnetic polarization

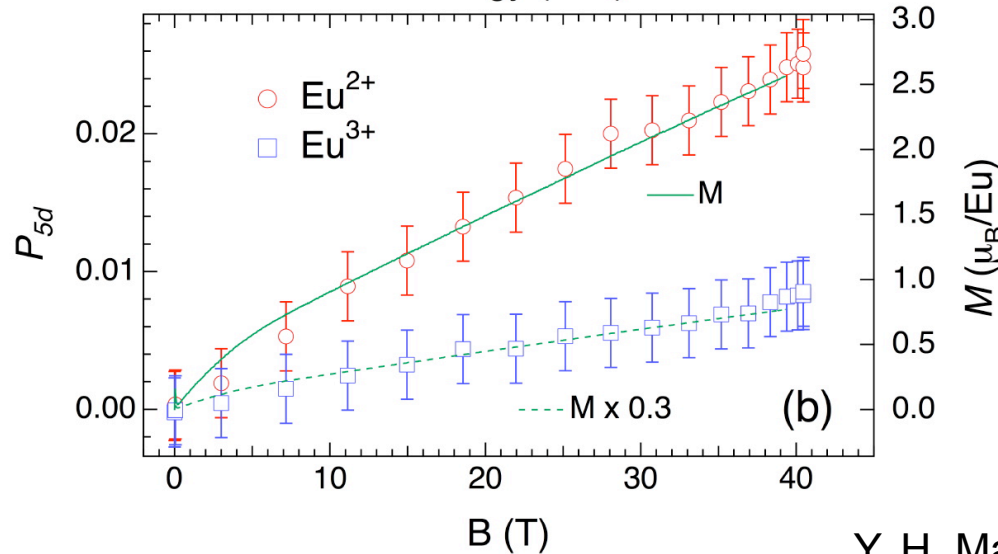
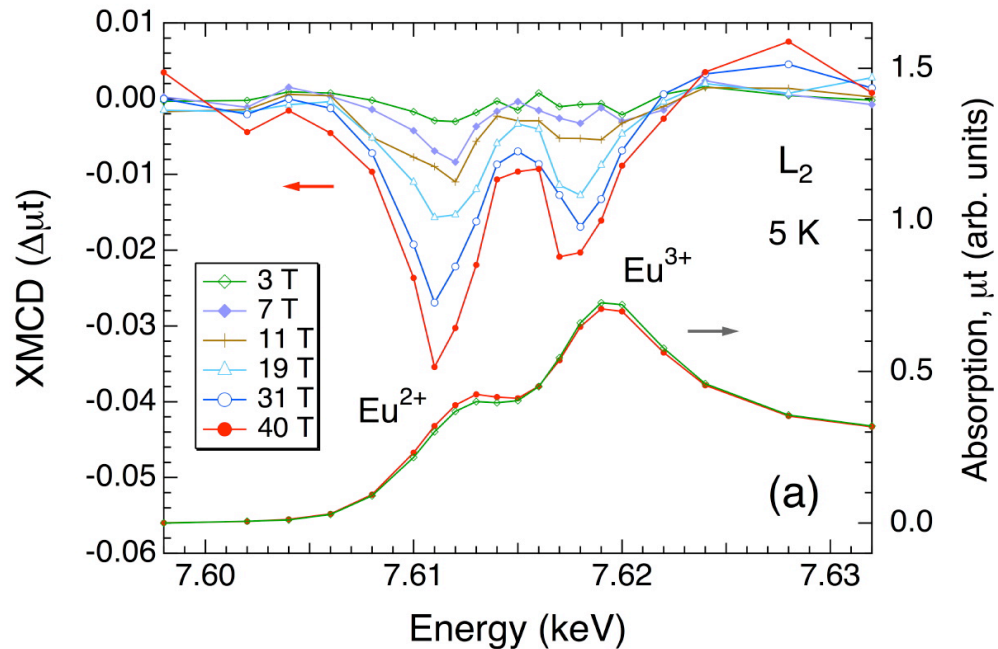
$$P_{5d} = \frac{\int \Delta\mu_t dE}{\int \mu_t dE}$$

Magnetic field dependence of P_{5d} is qualitatively different between Eu^{3+} and Eu^{2+} states.

$$P_{5d}(3+)/P_{5d}(2+) \sim 0.1 \quad (B < 20 \text{ T})$$



XMCD of EuNi_2P_2



◆ Hybridization is larger than $\text{EuNi}_2(\text{Si}_{0.18}\text{Ge}_{0.12})_2$.

◆ Valence is insensitive to magnetic field.

◆ $\text{Eu}^{3+}(J=0)$ XMCD is more significant than $\text{EuNi}_2(\text{Si}_{0.18}\text{Ge}_{0.12})_2$.

$$P_{5d}(3+)/P_{5d}(2+) \sim 0.3$$

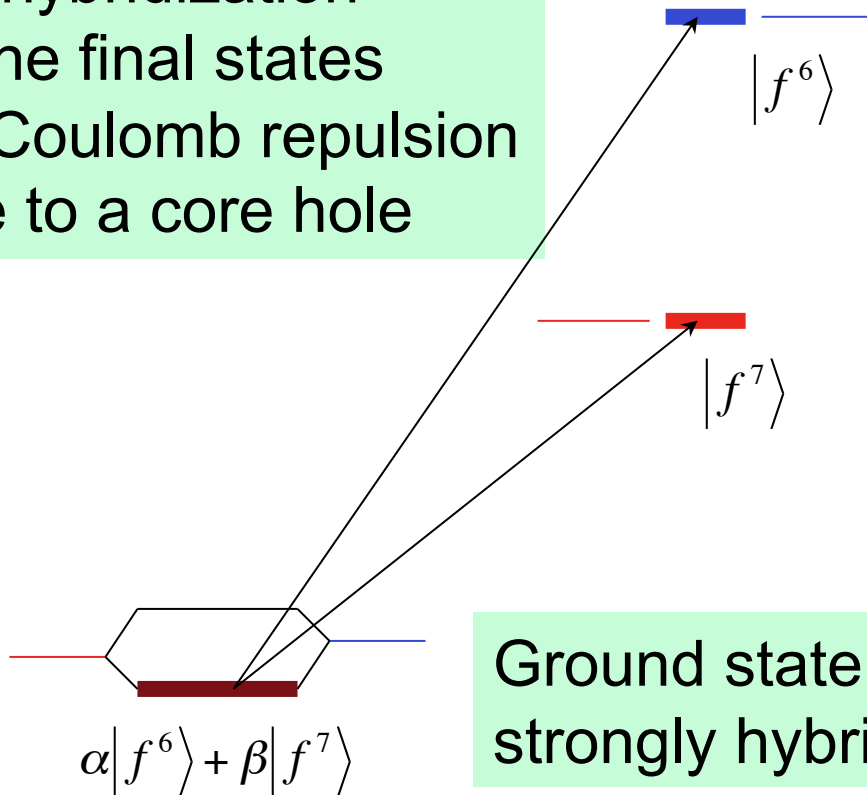
Discussion

1. XMCD of Eu^{3+} (f^6 , $J=0$)

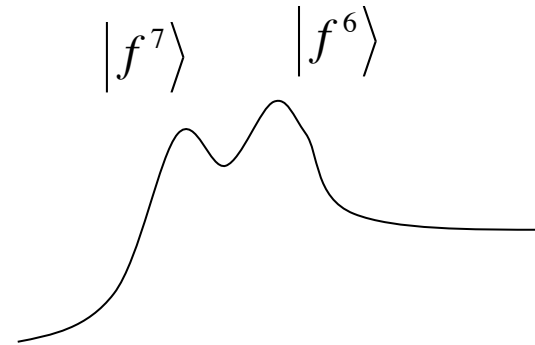
① Interference Effect



No hybridization of the final states by Coulomb repulsion due to a core hole



Ground state is strongly hybridized

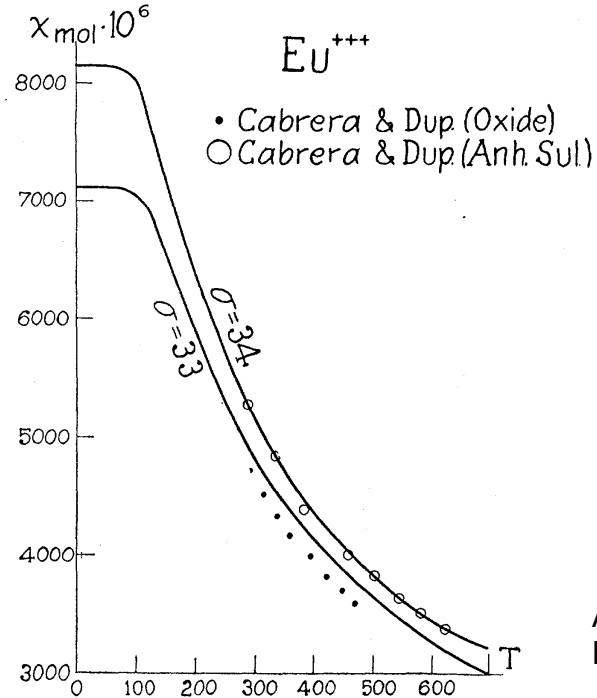
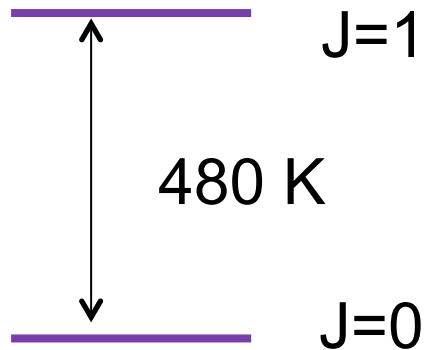


each absorption peak is due to the pure state

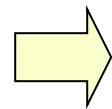
No Eu^{2+} component in the XMCD peak defined as Eu^{3+} XMCD

② Van Vleck term

Eu³⁺



0.3 μ_B/Eu³⁺
at 20 T



	EuNi ₂ (Si _{0.18} Ge _{0.82}) ₂	EuNi ₂ P ₂
$M(\text{Eu}^{3+})/M(\text{Eu}^{2+})$	0.06	0.09

Polarization of
Eu 5d electron

$P_{5d}(\text{Eu}^{3+})/P_{5d}(\text{Eu}^{2+})$	0.12	0.30
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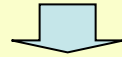
except for the
Van Vleck term

0.06

0.21

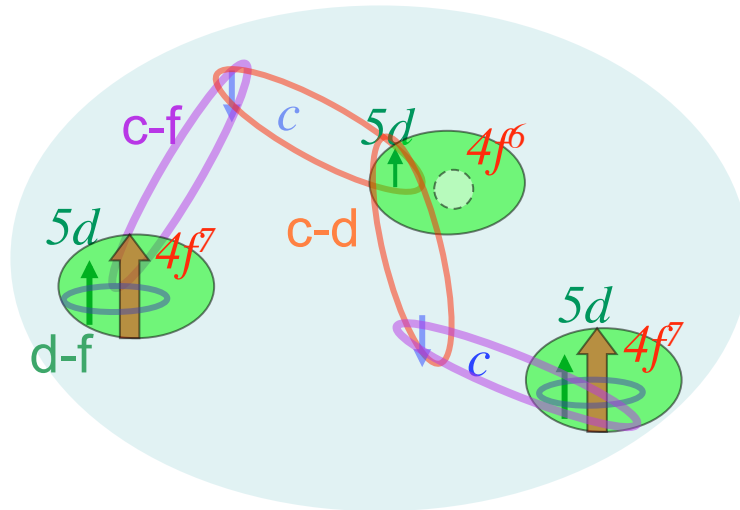
③ c-f hybridization effect

c-f hybridization → conduction electrons polarized by Eu^{2+} ($J=7/2$)

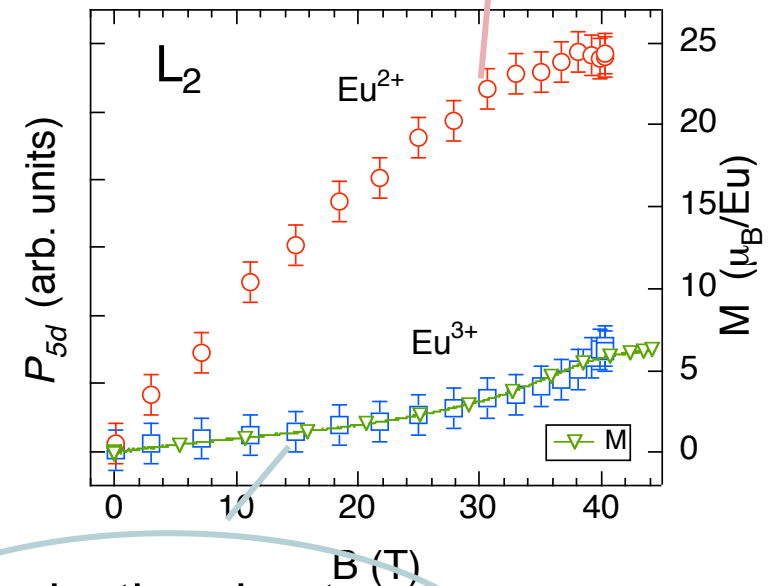


5d electrons of Eu^{3+} ($J=0$) are polarized by the conduction electrons

Polarization due to a local Eu^{2+} ($J=7/2$) state.



Eu^{3+} MCD reflects the strength of the c-f hybridization



Polarization due to valence fluctuating Eu ions at neighboring sites.

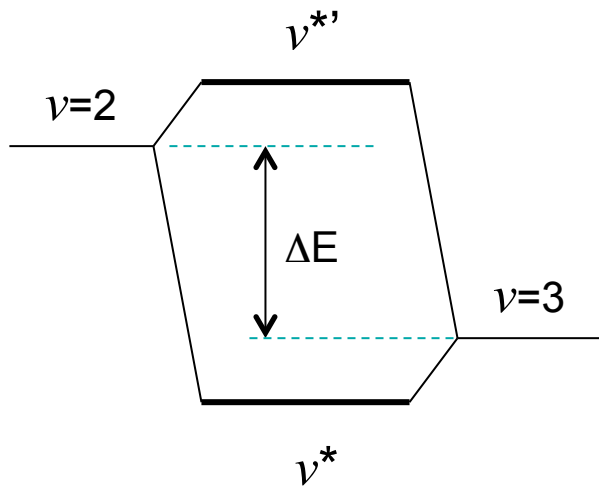
Material dependence

XMCD



	$\text{EuNi}_2(\text{Si}_{0.18}\text{Ge}_{0.82})_2$	EuNi_2P_2
$P_{5d}(\text{Eu}^{3+})/P_{5d}(\text{Eu}^{2+})$	0.06~0.12	0.21~0.30

× 3.5~2.5



	$\text{EuNi}_2(\text{Si}_{0.18}\text{Ge}_{0.82})_2$	EuNi_2P_2
Low T, zero field		
Eu valence ν^*	2.8	2.6
V	$0.7\Delta E$	$2.3\Delta E$

× 3.3

$$V = -\langle \nu=2 | \mathcal{H} | \nu=3 \rangle$$

Hybridization parameter

P_{5d} of Eu^{3+}

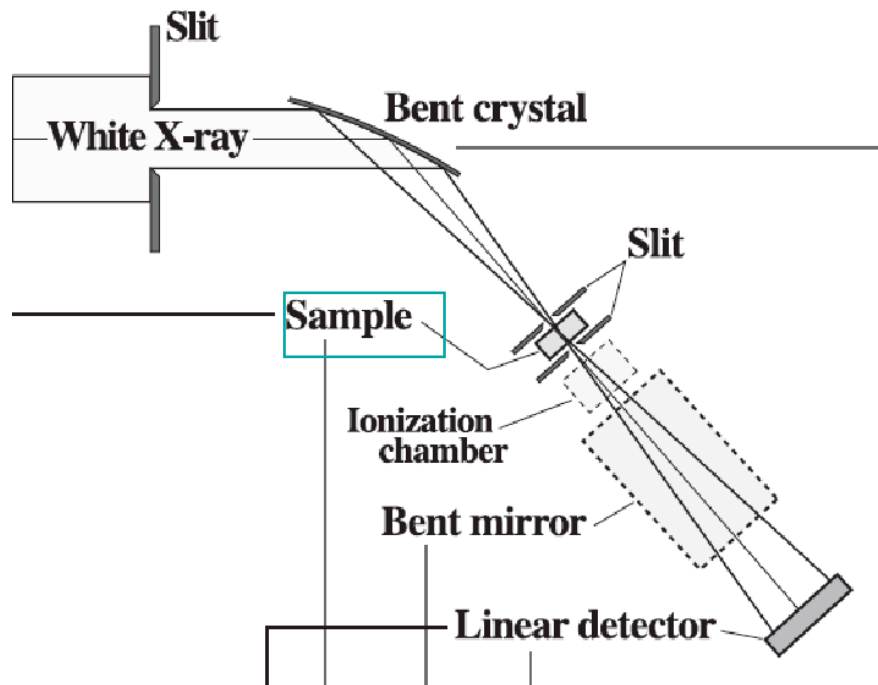


strength of the hybridization

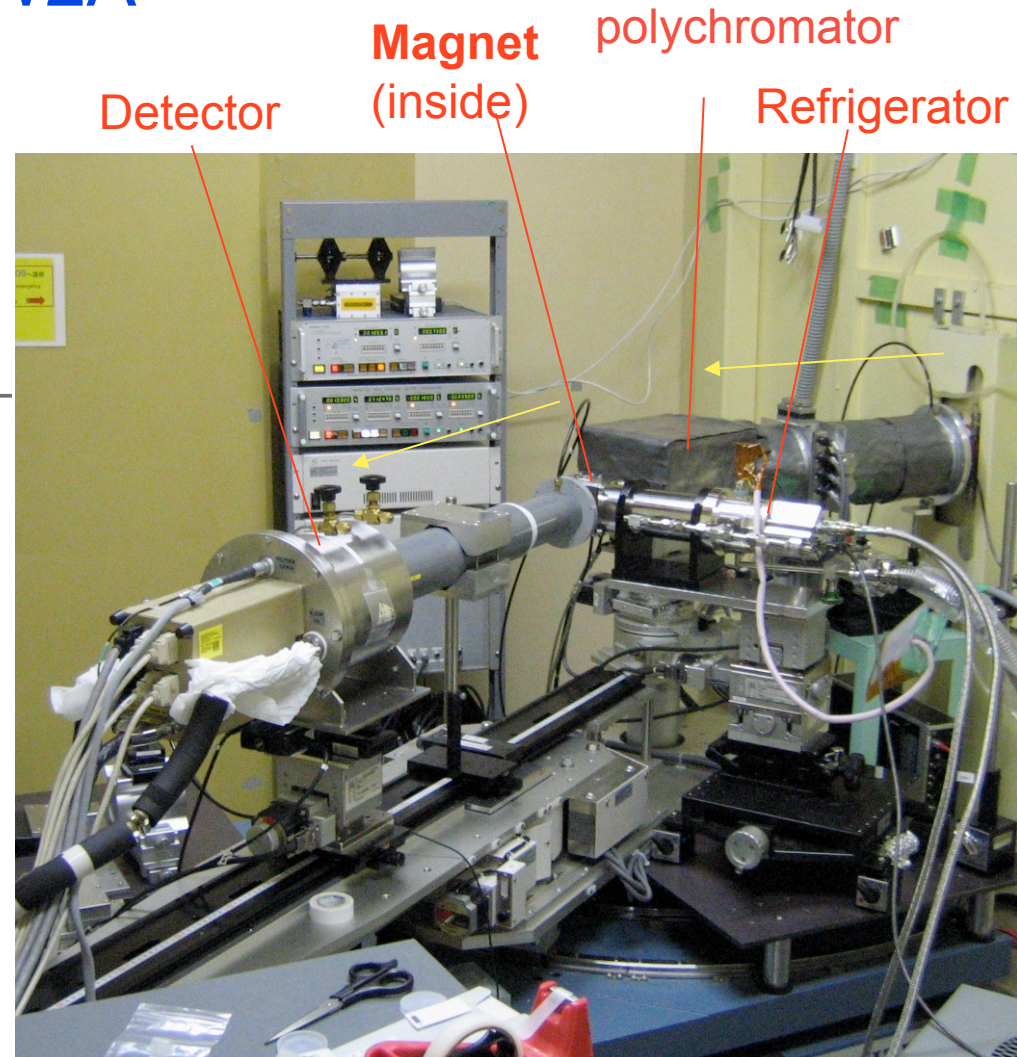
DXAFS : PF-AR NW2A (KEK Tsukuba)

Photon Factory AR NW2A

DXAFS spectrometer



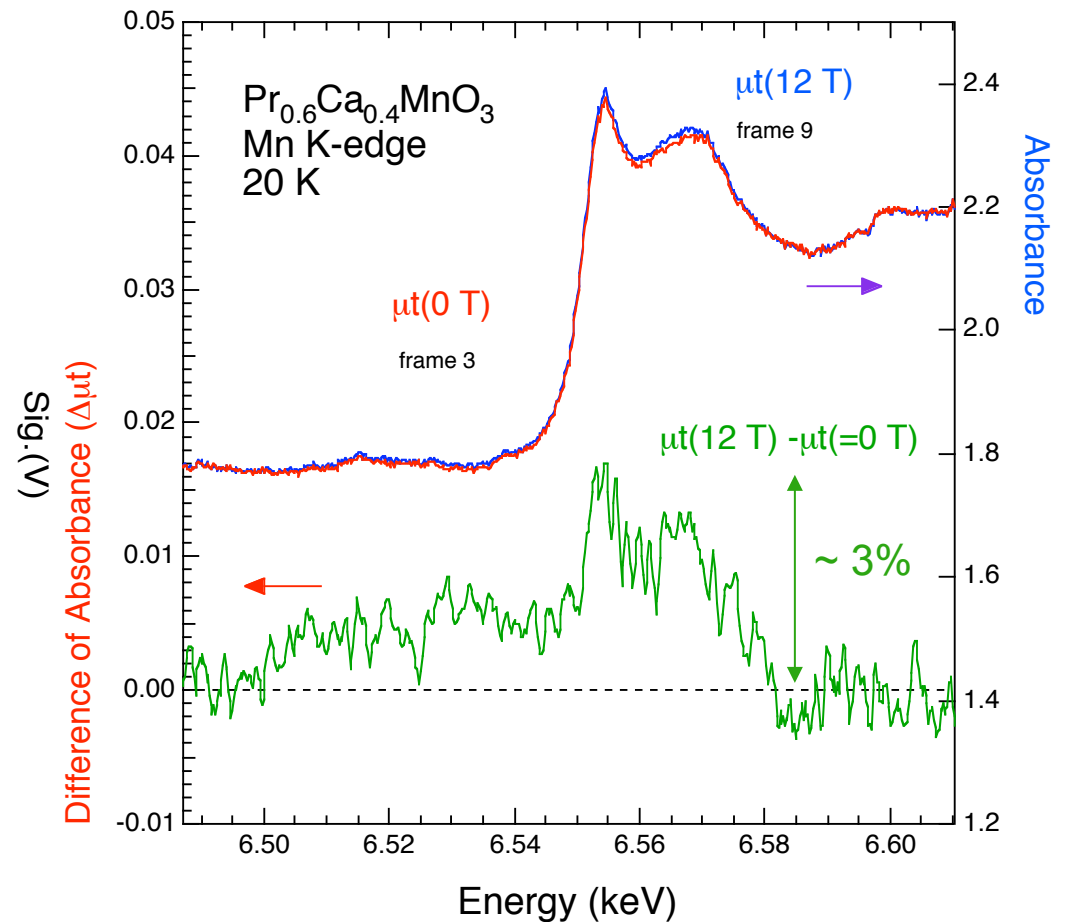
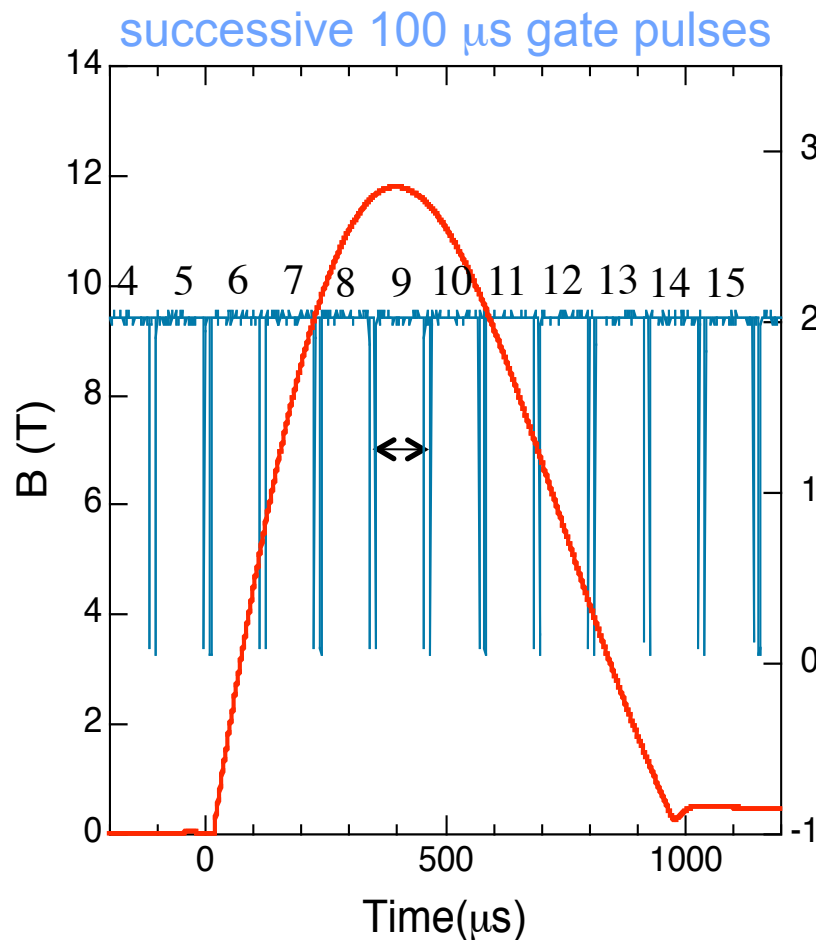
Y. Inada, M. Nomura
Photon Factory News
vol. 23 No.1(2005)

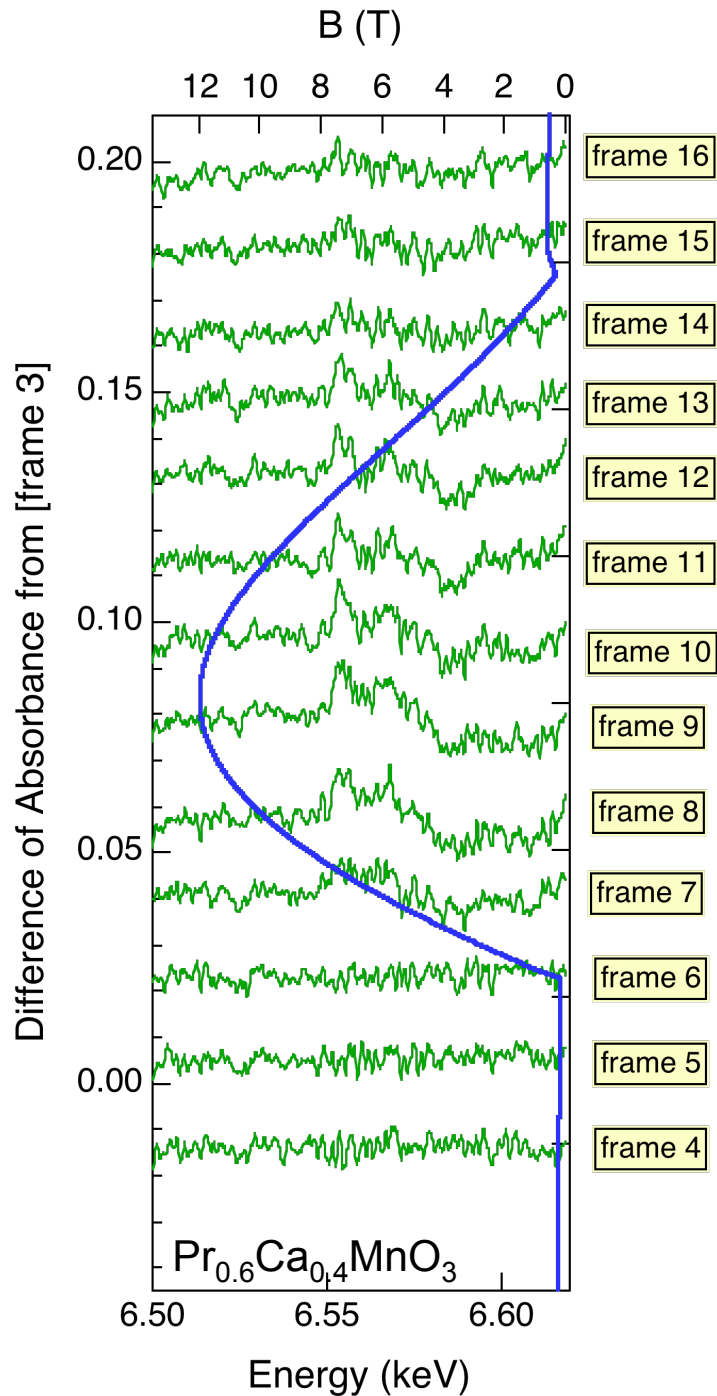


High-Magnetic-Field DXAFS Study

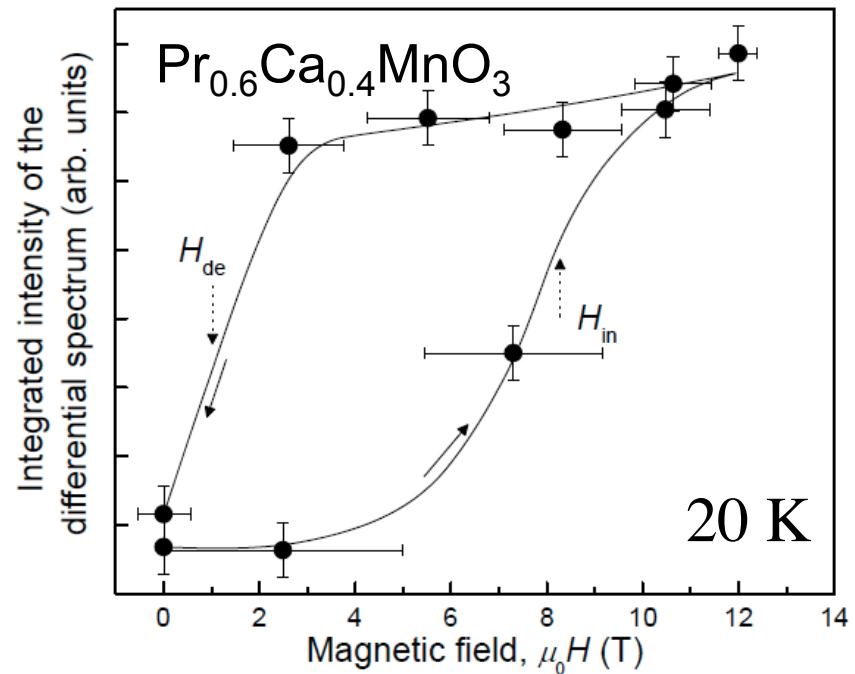
Gate $\Delta t = 100 \mu\text{s}$

$\text{Pr}_{0.6}\text{Ca}_{0.4}\text{MnO}_3$ Field-induced Insulator-Metal transition





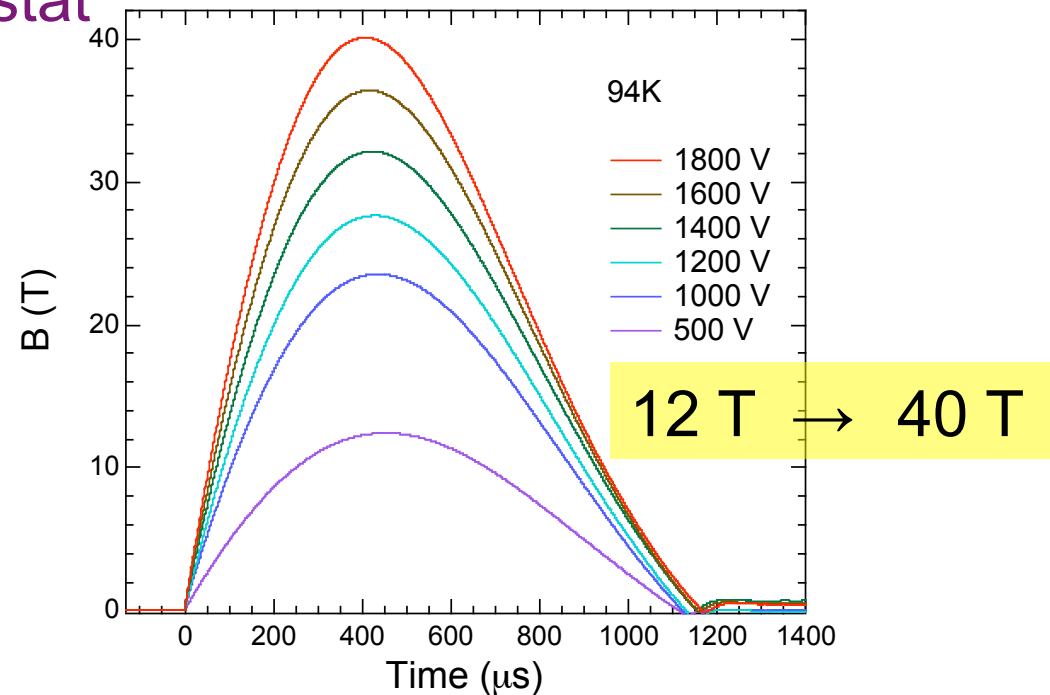
A small absorption change due to the field-induced I-M transition is detected.



Z. W. Ouyang, Y. H. Matsuda, et al.,
 J. Phys. : Condens. Matter **21** (2009) 016006.

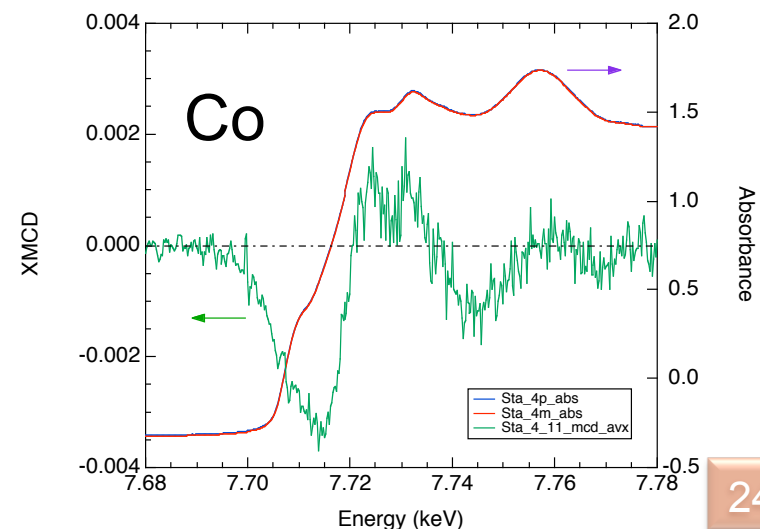
Development of techniques underway

◆ Pulsed magnet & Cryostat



◆ A diamond phase retarder for XMCD

in collaboration with
K. Hirano (KEK-PF)



Summary

- Valence selective XMCD in $\text{EuNi}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ and EuNi_2P_2 were measured up to 40 T using a mini-magnet.
- XMCD of Eu^{3+} ($J=0$) possibly reflects the magnetism of itinerant states through the c-f hybridization, while XMCD of Eu^{2+} ($J=7/2$) reflects localized states.
- DXAFS experiment in high magnetic fields were made. At the field-induced I-M transition of $\text{Pr}_{0.6}\text{Ca}_{0.4}\text{MnO}_3$, recovering the local lattice distortion induces an enhancement of the Mn K-edge absorption intensity.

Other collaborators

Y. Murakami (Tohoku Univ.) Discussions

K. Yoshimura (Kyoto Univ.) YbInCu_4

A. Mitsuda (Kyushu Univ.) $\text{EuNi}_2(\text{Si}_{1-x}\text{Ge}_x)_2$

H. Ueda (Univ. Tokyo) CdCr_2O_4

T. Matsumura (Hiroshima Univ.) TbB_4

T. Arima (Tohoku Univ.) $\text{Pr}_{0.6}\text{Ca}_{0.4}\text{MnO}_3$

M. Suzuki, N. Kawamura (JASRI/SPring-8)

XMCD / BL39XU

Y. Inada, Y. Niwa, M. Nomura (KEK PF)

DXAFS / PF-AR NW2A