Fe superconductor workshop Sep. 4th, 2013

Instantaneous local moment measured with X-ray emission spectroscopy

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Physics
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Acknowledgements

Toronto

H. Gretarsson

A. Lupascu

Argonne+CHESS

Jungho Kim
Diego Casa
Mary Upton
Thomas Gog

Ken Finkelstein

Samples

W. Wu, S. R. Julian (Toronto)

Z. J. Xu, J. S. Wen, G. D. Gu (BNL)

R. H. Yuan Z. G. Chen, N.-L. Wang (IOP Beijing)

S. Khim, K. H. Kim (Seoul Natl. U)

M. Ishikado, I. Jarrige, S. Shamoto (JAEA)

J.-H. Chu, I. R. Fisher (Stanford)

S. R. Saha, T. Drye, J. Paglione (Maryland)

S. Kasahara (Kyoto)

A. Sefat (Oak Ridge)

Outline

- Motivation
- XES - What is measured?
- Local moment in Fe based materials

  Gretarsson et al. PRB 84, 100509 (R) (2012)
- Spin-state transition in RE doped Ca122

  Gretarsson et al. PRL 110, 047003 (2013)
- Spin-state transition in P doped Ca122
- Spin-state change in Cr doped Ba122

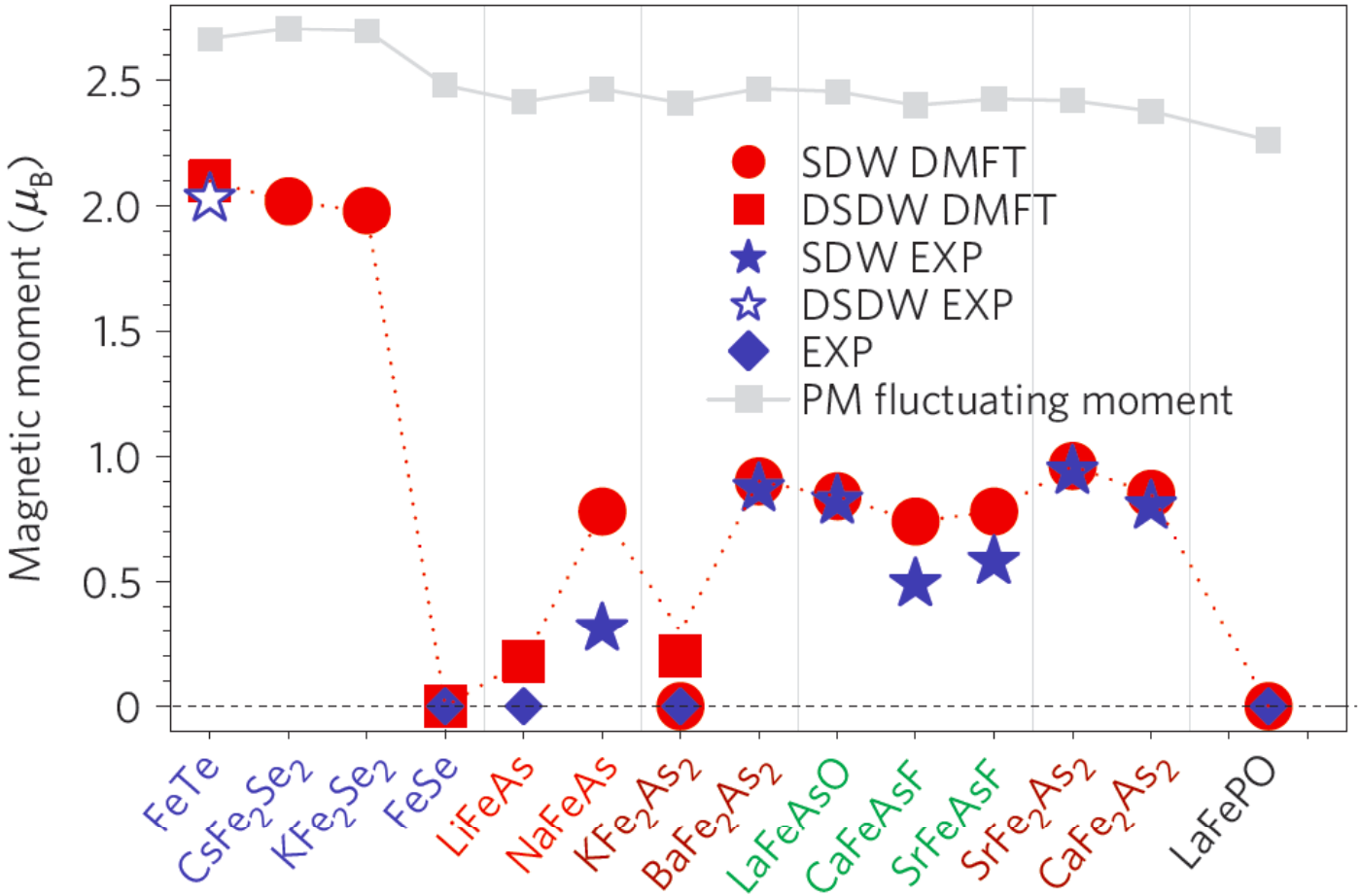
Magnetic moment conundrum

Material	T_N (K)	Wavevector	Moment	Technique
LaFeAsO	137	$(1\ 0\ \frac{1}{2})_O/(\frac{1}{2}\ \frac{1}{2}\ \frac{1}{2})_T$	0.36 μB	Neutrons
	138		4.86 T/23 MHz	Mössbauer/μSR
	145		5.19 T	Mössbauer
	140		5.3 T	Mössbauer
NdFeAsO	141	$(1\ 0\ \frac{1}{2})_O/(\frac{1}{2}\ \frac{1}{2}\ \frac{1}{2})_T$	0.25 μB	Neutrons
	141		5.3 T	Mössbauer
	135		23 MHz	μSR
PrFeAsO	136	$(1\ 0\ 0)_O/(\frac{1}{2}\ \frac{1}{2}\ 0)_T$	0.35 μB	Neutrons
	139		4.99 T	Mössbauer
	123		23 MHz	μSR
CeFeAsO	140	$(1\ 0\ 0)_O/(\frac{1}{2}\ \frac{1}{2}\ 0)_T$	0.8 μB	Neutrons
	136		5.06 T	Mössbauer
	137		~26 MHz	μSR
SmFeAsO	135		23.6 MHz	μSR
$BaFe_2As_2$	90*	$(1\ 0\ 1)_O/(\frac{1}{2}\ \frac{1}{2}\ 1)_T$	0.99 μB	Neutrons
	143	$(1\ 0\ 1)_O/(\frac{1}{2}\ \frac{1}{2}\ 1)_T$	0.87 μB	Neutrons
	140		28.8 MHz	μSR
	140		5.47 T	Mössbauer
$SrFe_2As_2$	220	$(1\ 0\ 1)_O/(\frac{1}{2}\ \frac{1}{2}\ 1)_T$	0.94 μB	Neutrons
	205	$(1\ 0\ 1)_O/(\frac{1}{2}\ \frac{1}{2}\ 1)_T$	1.01 μB	Neutrons
	205		44 MHz	μSR
	205		8.91 T	Mössbauer
$CaFe_2As_2$	173	$(1\ 0\ 1)_O/(\frac{1}{2}\ \frac{1}{2}\ 1)_T$	0.8 μB	Neutrons
$EuFe_2As_2$	200		8.5 T	Mössbauer
$Fe_{1.076}Te$	75	$(\frac{1}{2}\ 0\ \frac{1}{2})$	2.03 μB	Neutrons
$Fe_{1.141}Te$	63	$(0.38\ 0\ \frac{1}{2})$	1.96 μB	Neutrons
$Fe_{1.068}Te$	67	$(\frac{1}{2}\ 0\ \frac{1}{2})$	2.25 μB	Neutrons
$Na_{1-\delta}FeAs$	37	$(1\ 0\ \frac{1}{2})_O/(\frac{1}{2}\ \frac{1}{2}\ \frac{1}{2})_T$	0.09 μB	Neutrons

Lumsden and Christianson J. Phys.: Condens. Matter **22**, 203203 (2010)

- DFT predicts large moments for all Fe pnictides and chalcogenides: ~2 μ_B
- Experimentally observed ordered moment varies widely.

Magnetic Moment



Z. P. Yin, K. Haule, and G. Kotliar, Nature Mater. 10, 932 (2011)

Theoretical approaches

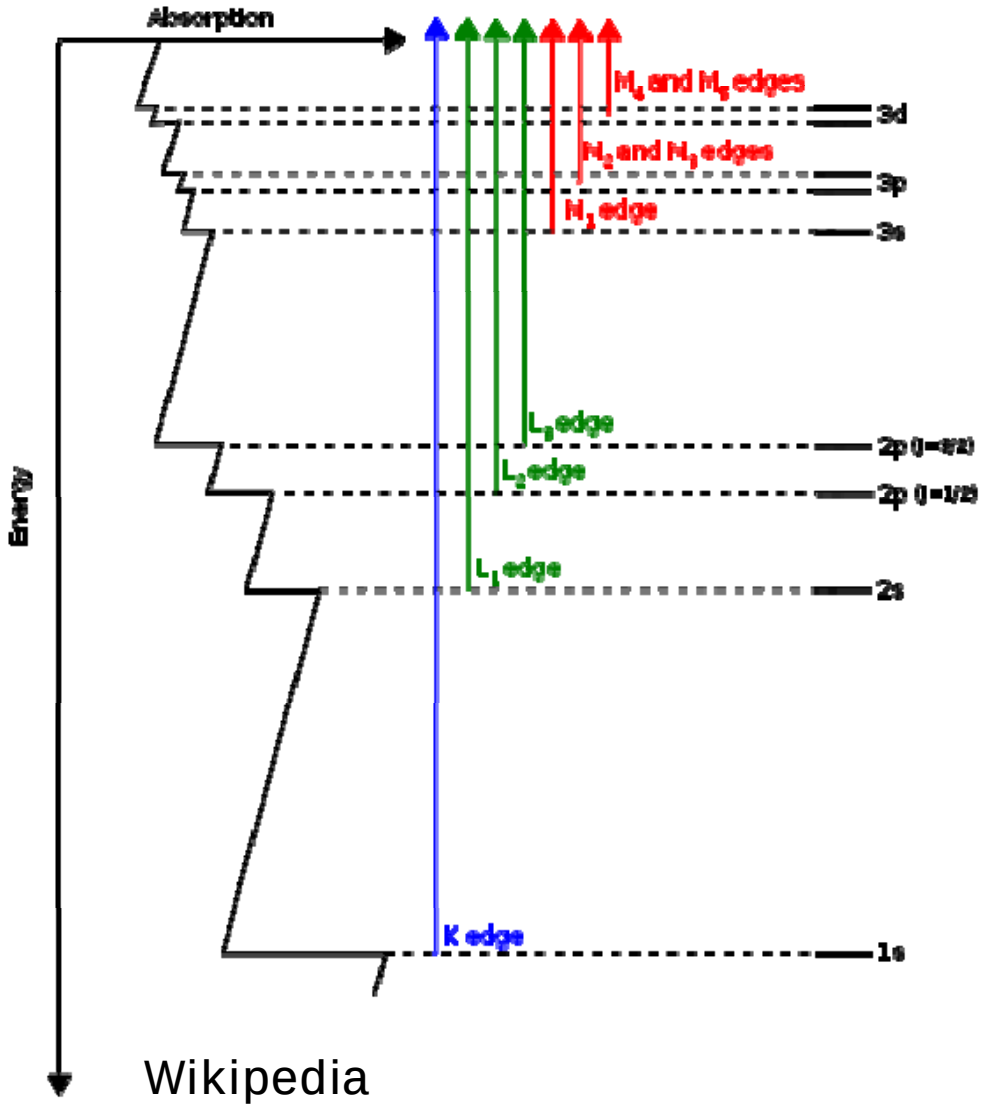
- Dynamic screening of local moment (time-scale)
  - Hansmann et al. PRL 104, 197002 (2010)
- Separation of local and itinerant orbitals
  - Wei-Guo Yin et al. PRL 105, 107004 (2010)
  - Z. P. Yin et al., Nat. Mater. 10, 932 (2011)
- Spin-state crossover (local model)
  - Chaloupka and Khaliullin PRL 110, 207205 (2013)

Need experimental probes for measuring
Local, Fluctuating moment:
➔Q and E integrated Inealstic Neutron Scattering
Harriger et al., PRB 86, 140403 (2012)

X-RAY EMISSION SPECTROSCOPY

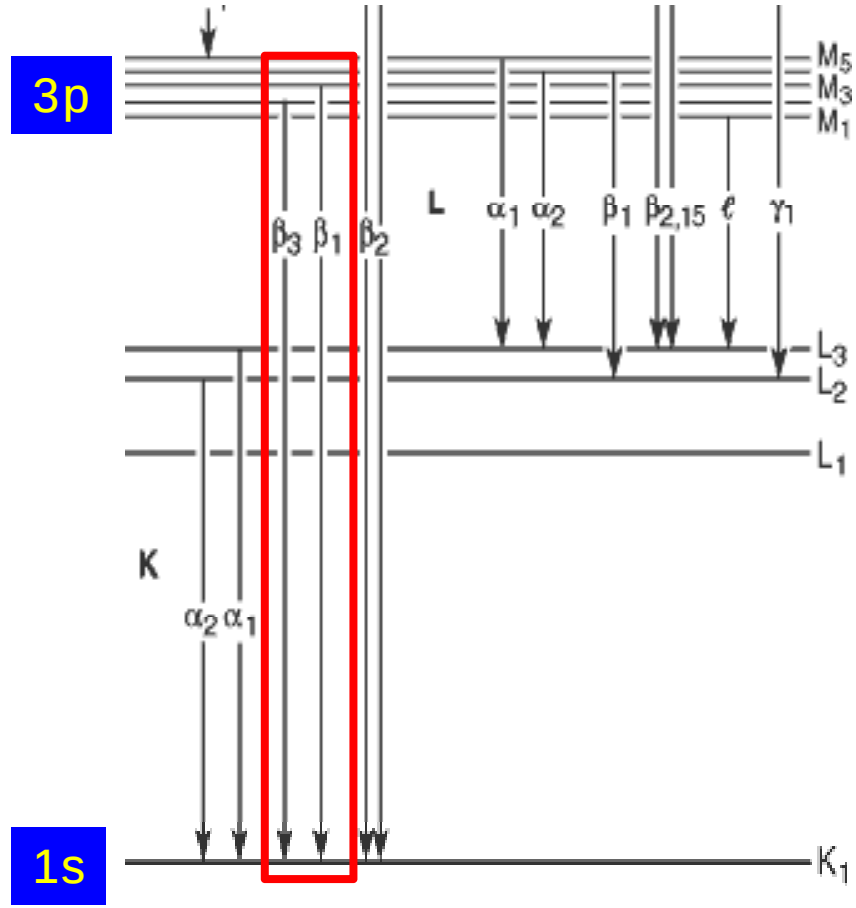
X-ray physics primer

Absorption



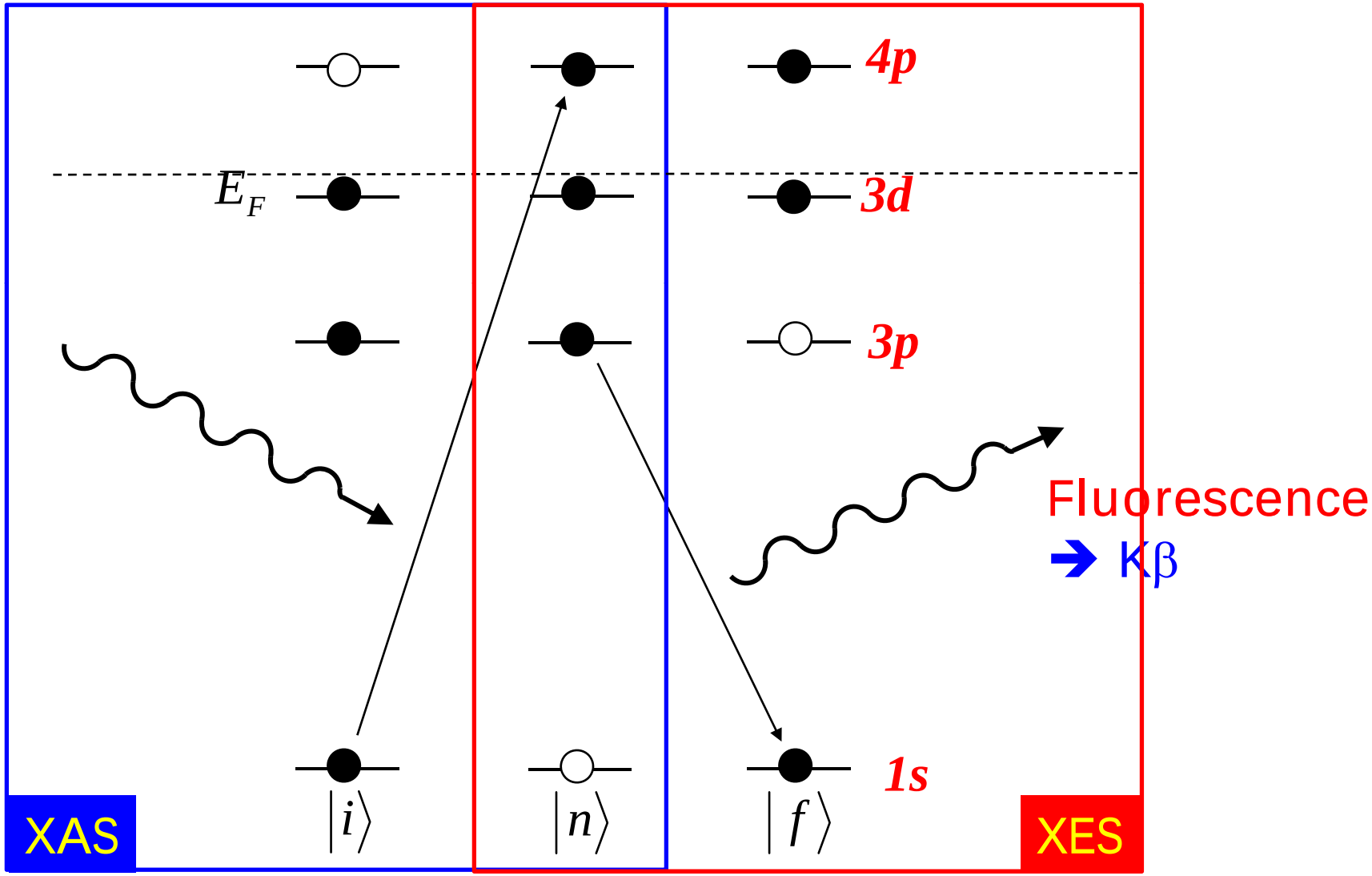
Wikipedia

Emission



X-ray Data booklet
(Center for X-ray Optics)

X-ray Absorption vs. Emission

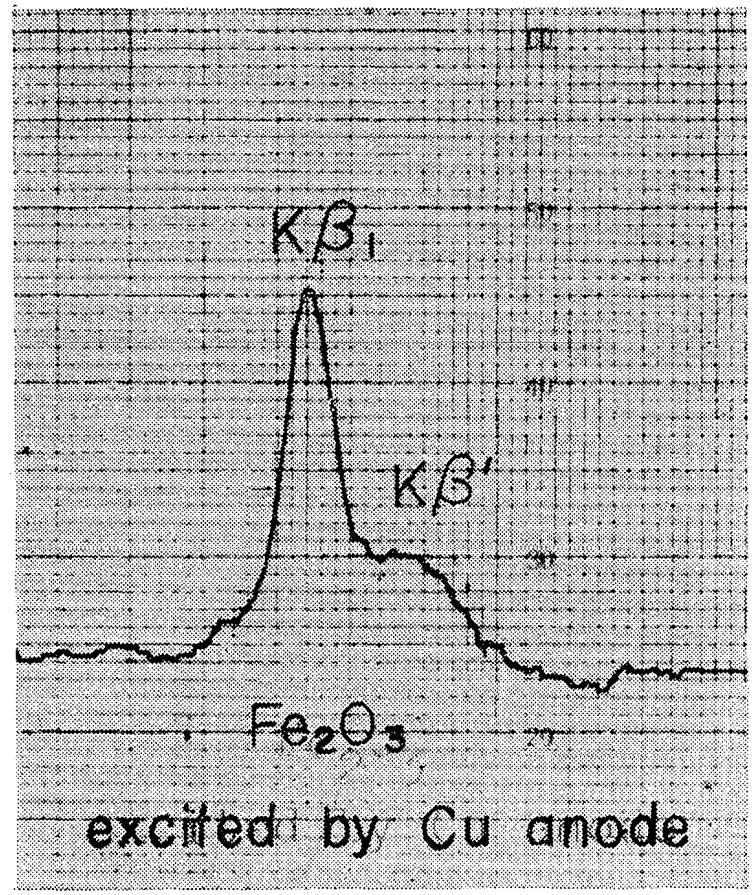


JOURNAL OF THE PHYSICAL SOCIETY OF JAPAN, Vol. 14, No. 12, DECEMBER 1959

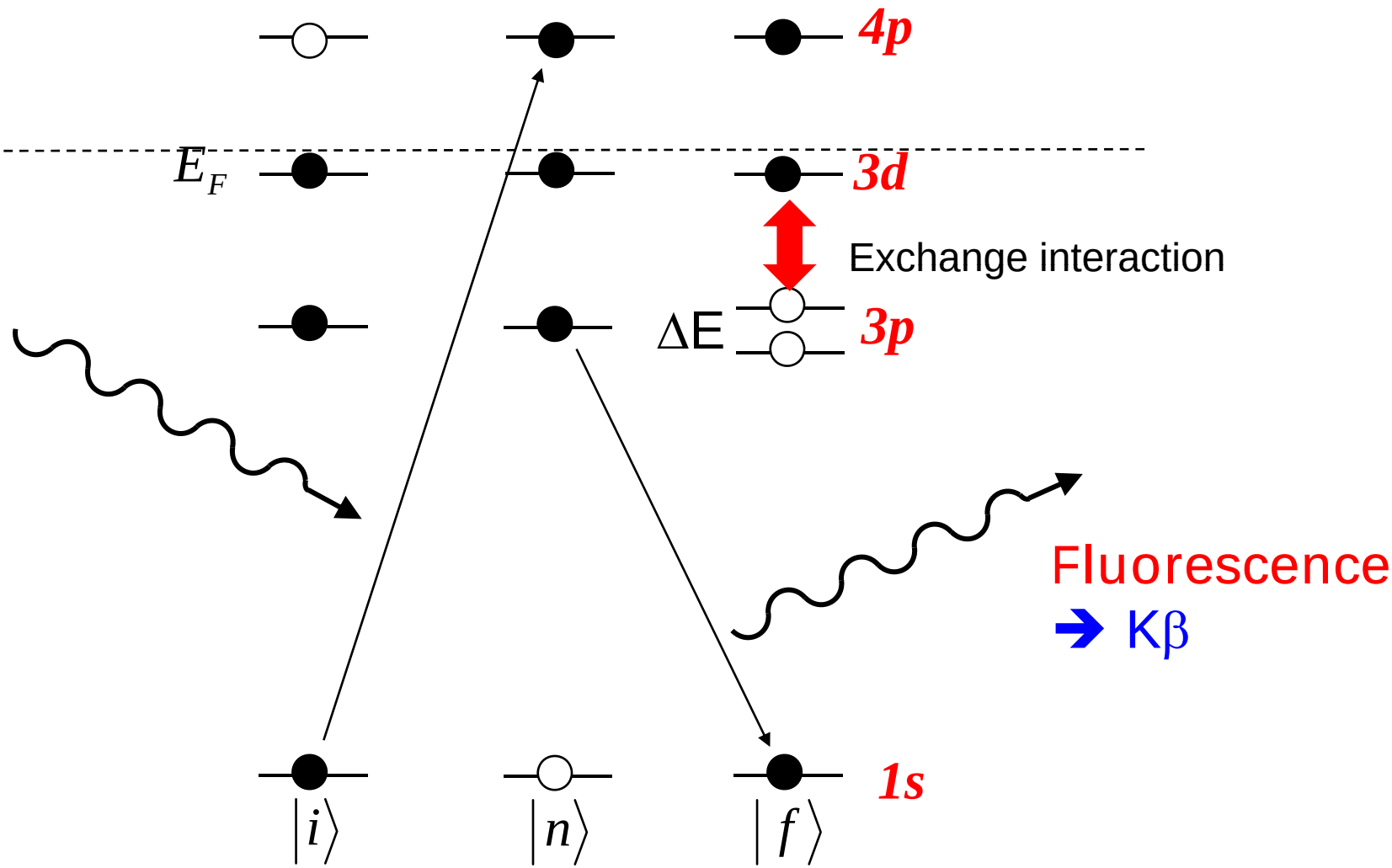
The X-ray Non-diagram Lines $K\beta'$ of Some Compounds of the Iron Group

By Kenjiro TSUTSUMI

Department of Physics, Faculty of Science, Osa



X-ray Absorption vs. Emission



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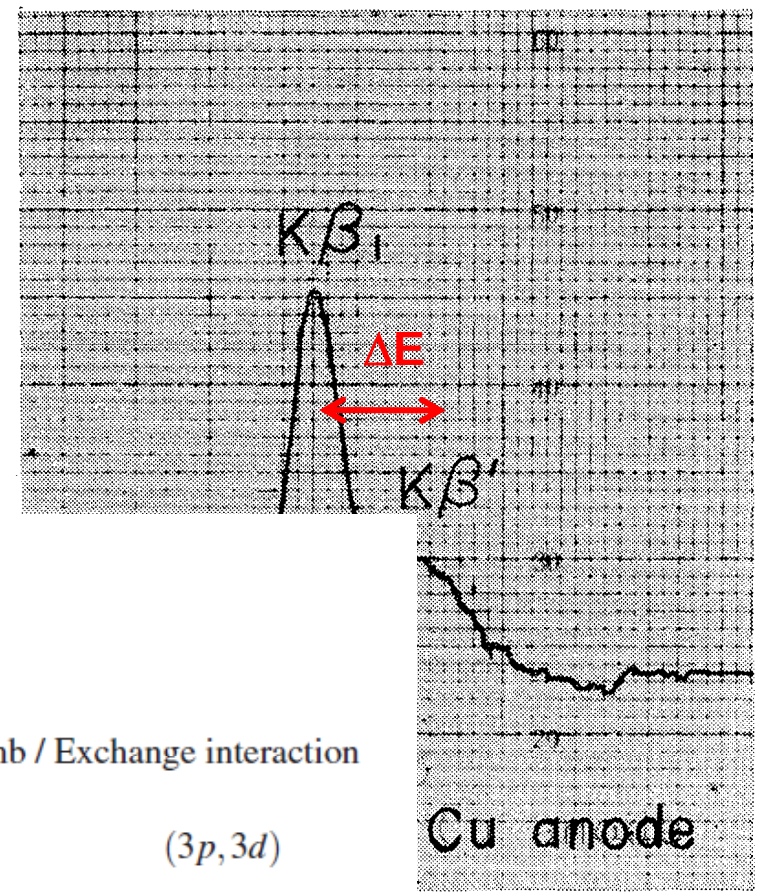
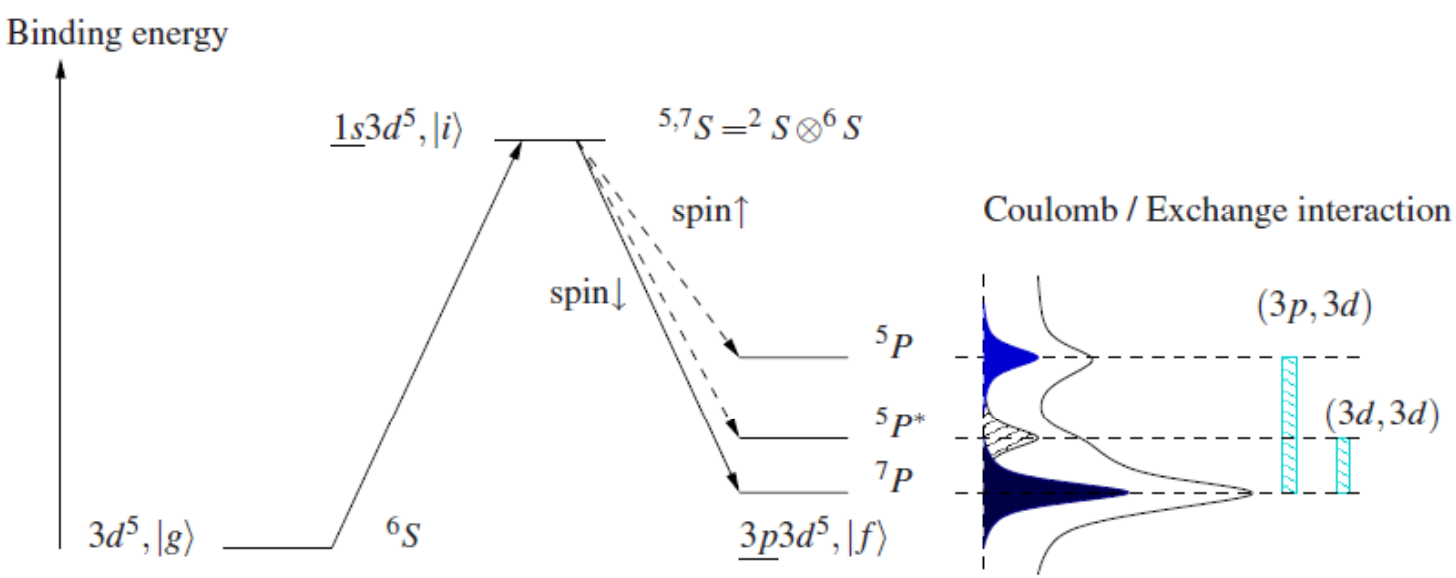
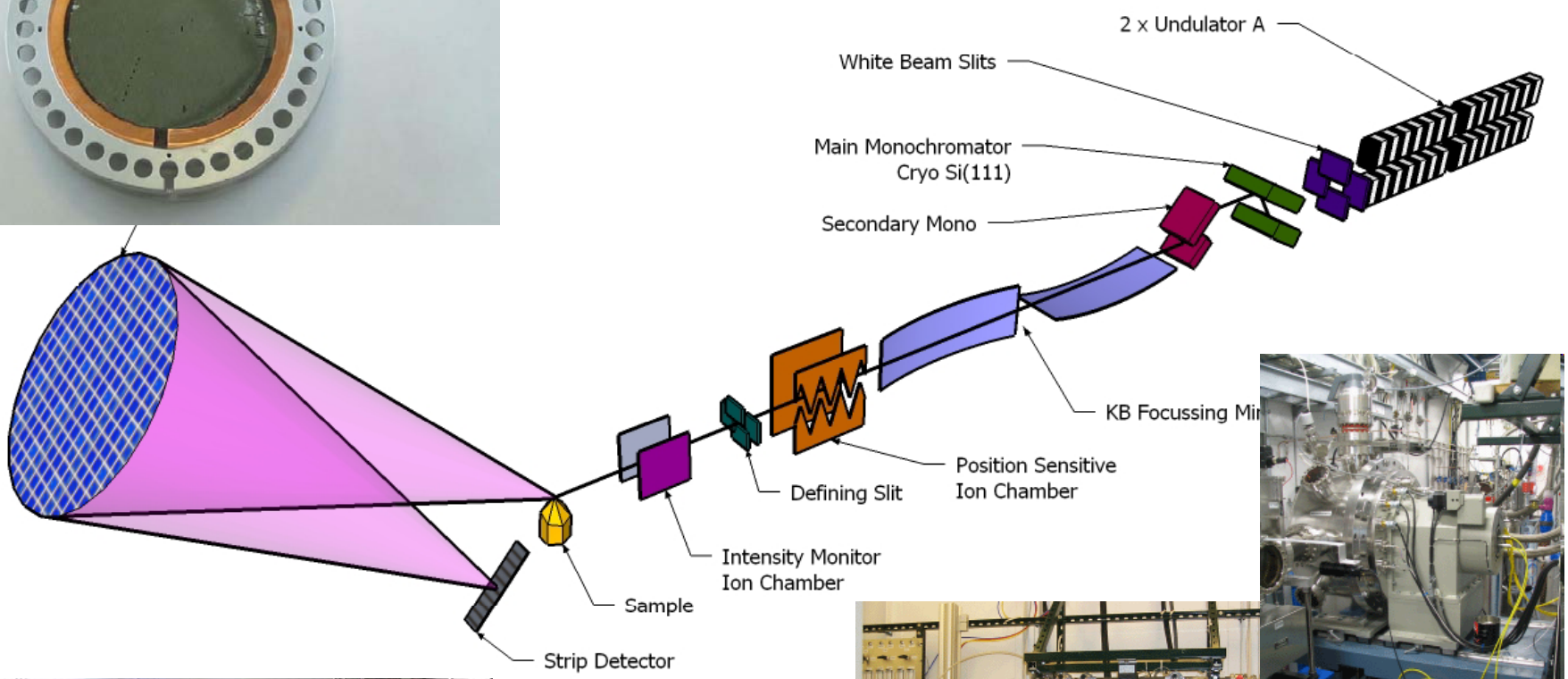
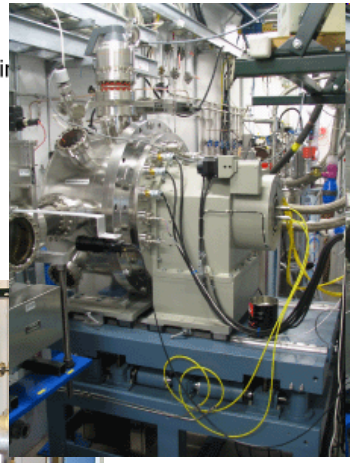
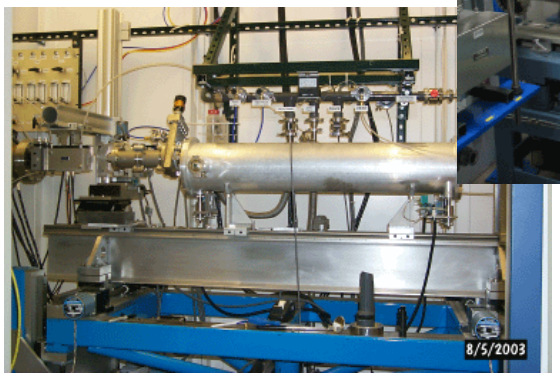


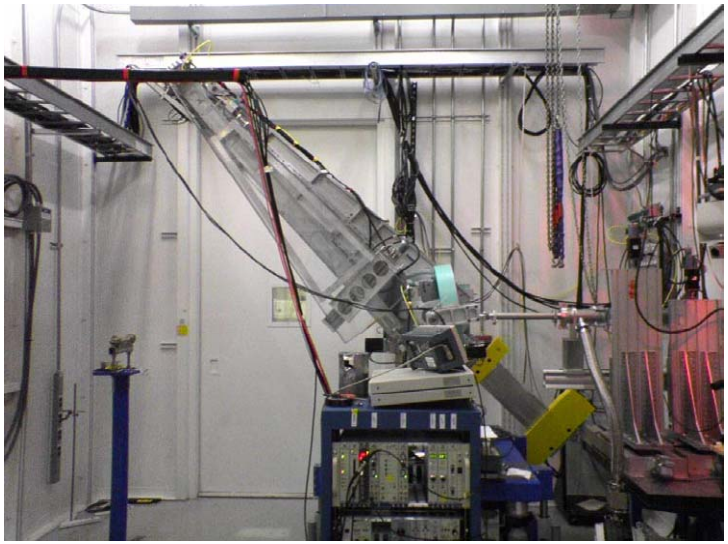
J-P Rueff and A. Shukla, RMP 2010




2 x Undulator A
White Beam Slits
Main Monochromator
Cryo Si(111)
Secondary Mono
KB Focussing Mi
Position Sensitive
Ion Chamber
Defining Slit
Intensity Monitor
Ion Chamber
Sample
Strip Detector

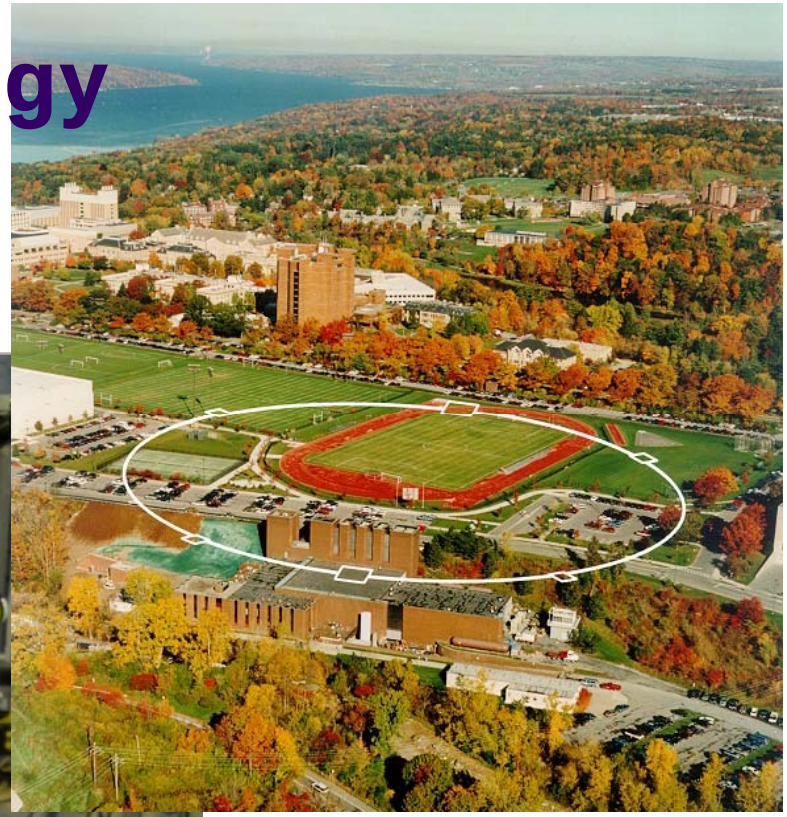


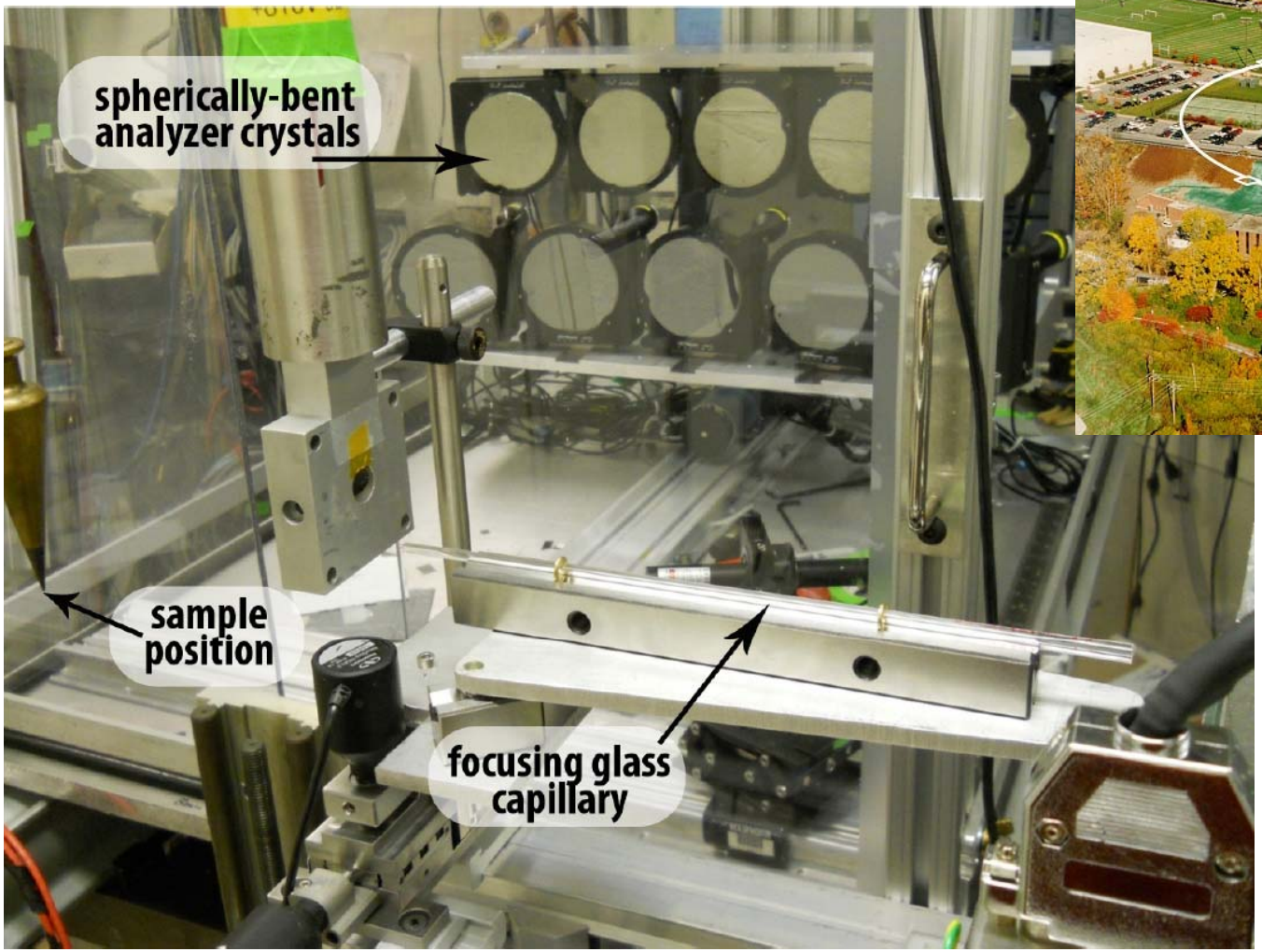

8/5/2003



APS 9ID

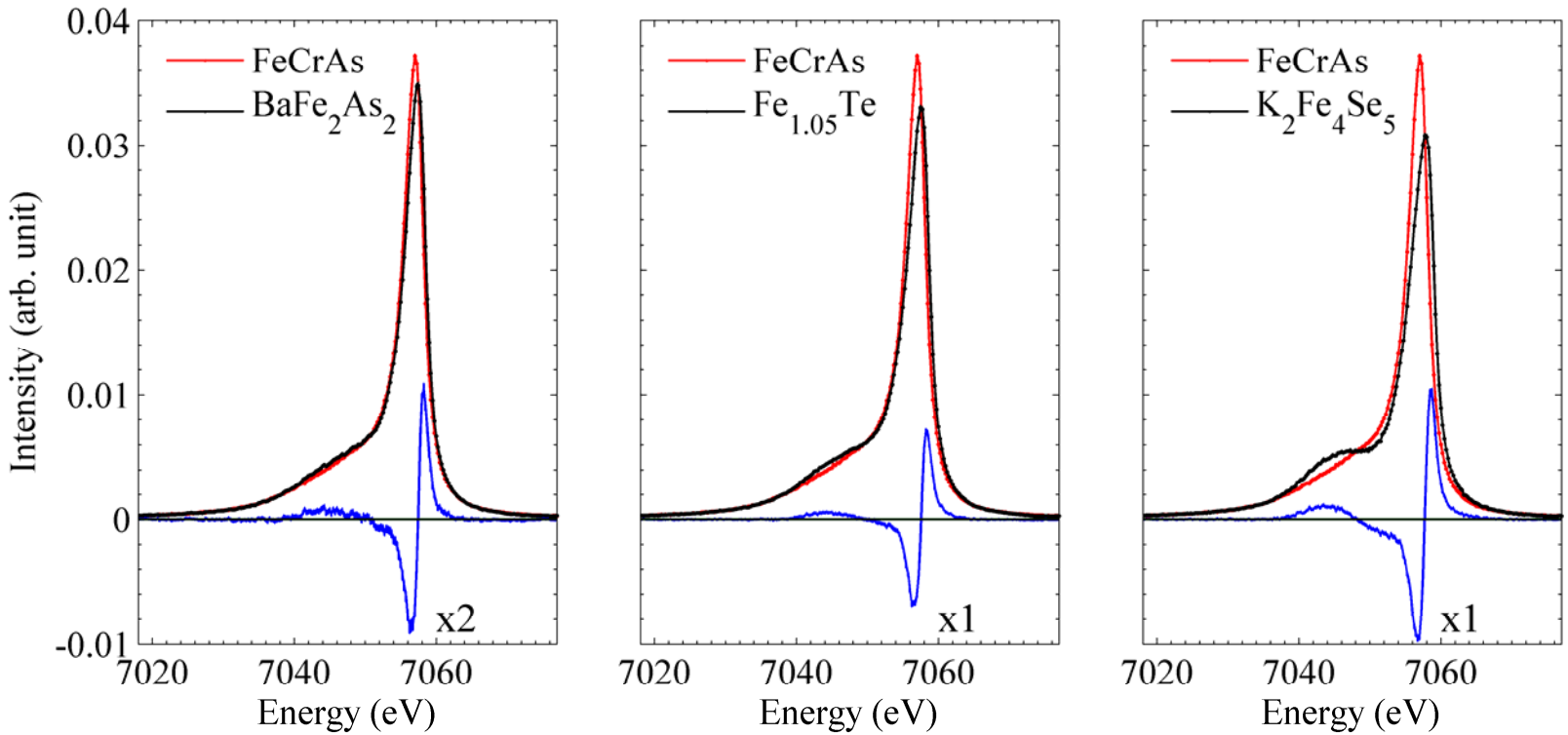
CHESS (Cornell High Energy Synchrotron Source)





C1 XES station
Ken Finkelstein

Spectra comparison



How to obtain Quantitative information?

$$IAD = \int \left| I(E) - I_{ref}(E) \right| dE$$

Vanko, de Groot et al. J. Phys. Chem. B 110, 11647 (2006)

Multiplet structure

Vanko and de Groot, Phys. Rev. B 75, 177101 (2007)

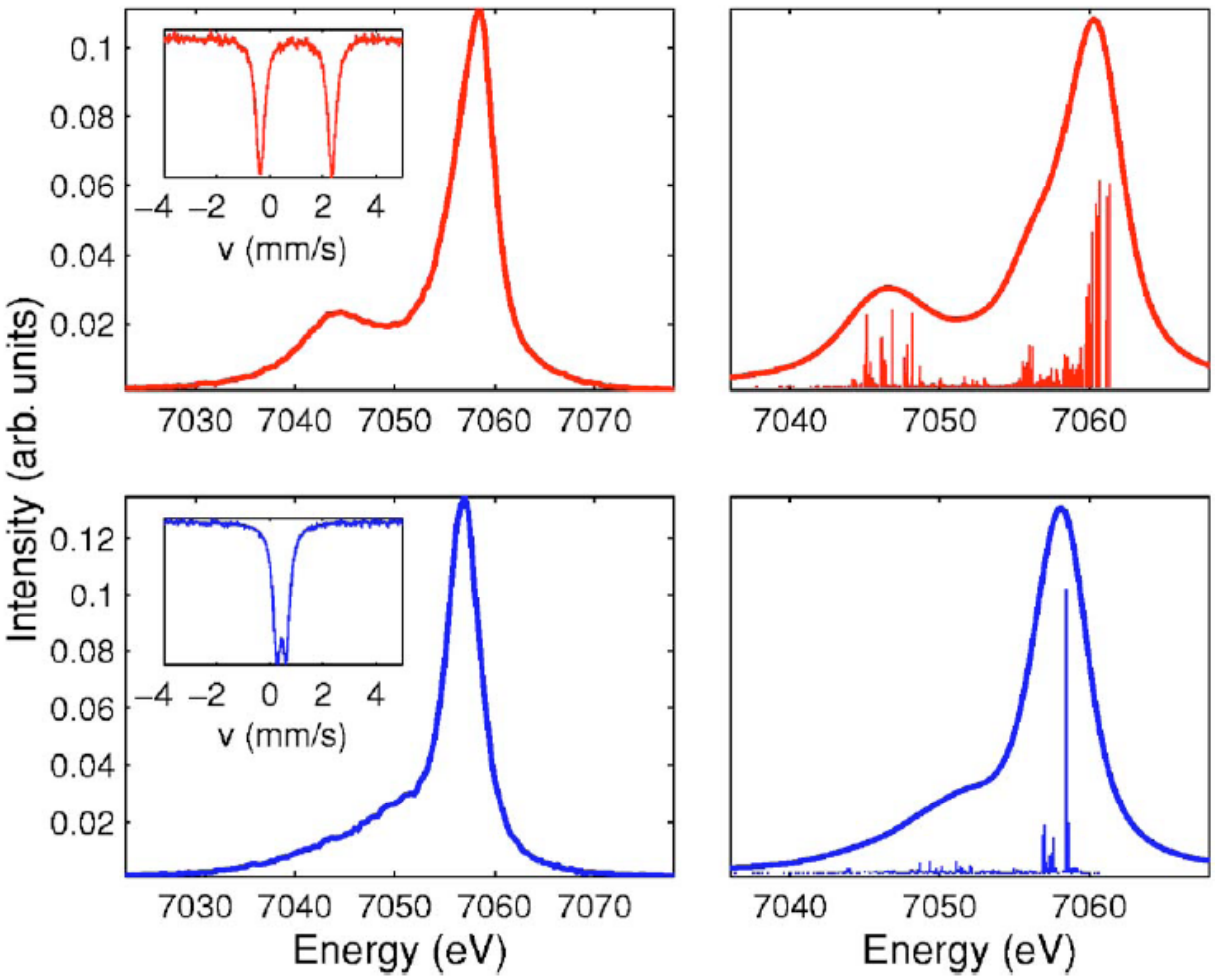
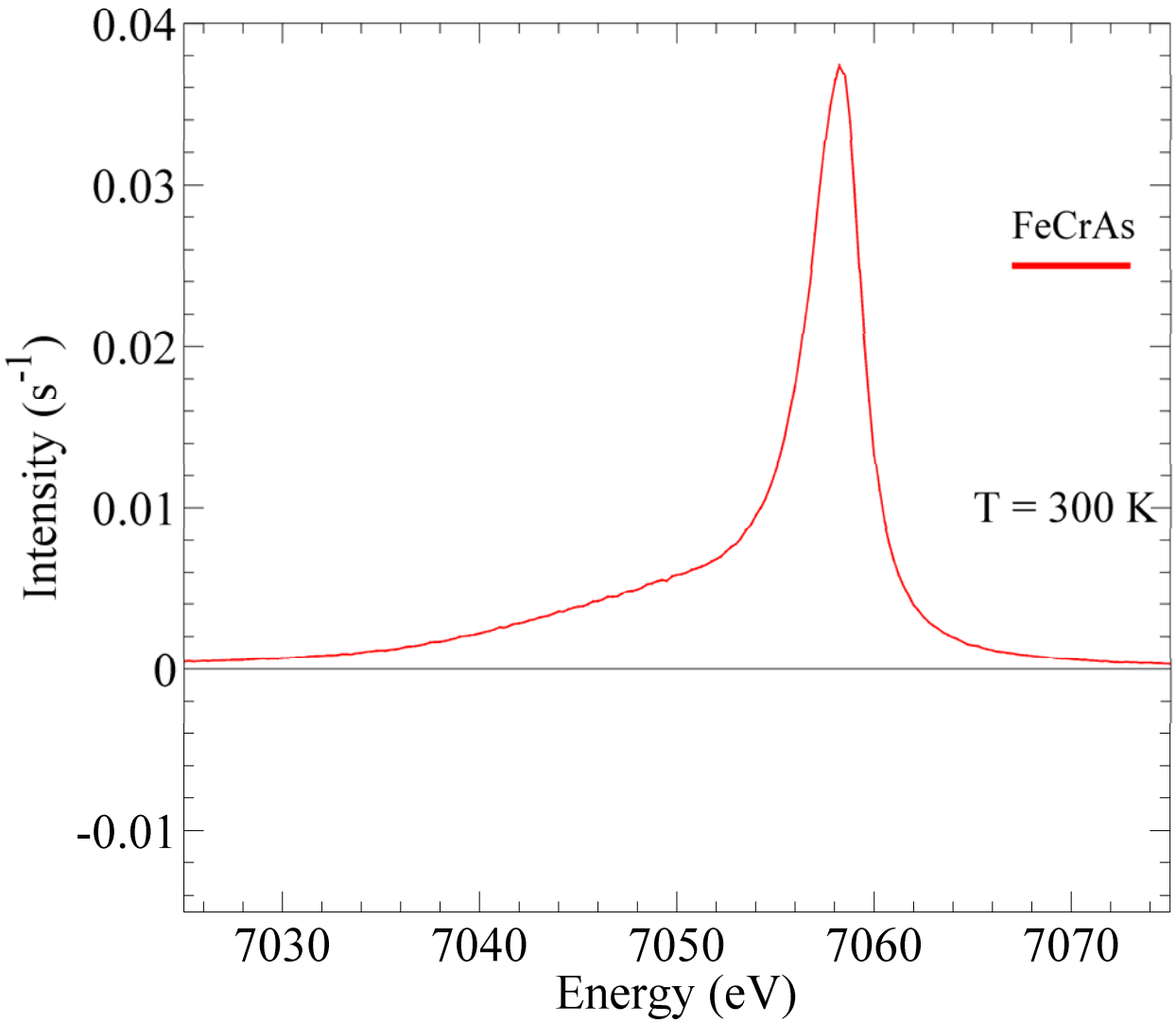


FIG. 1. (Color online) Experimental (left panel) and theoretical (right panel) $K\beta$ spectra of $[Fe(phen)_2(NCS)_2]$, in its HS (top) and LS (bottom) forms. Insets in the experimental part display the Mössbauer spectra.

IAD analysis method



A reference sample required

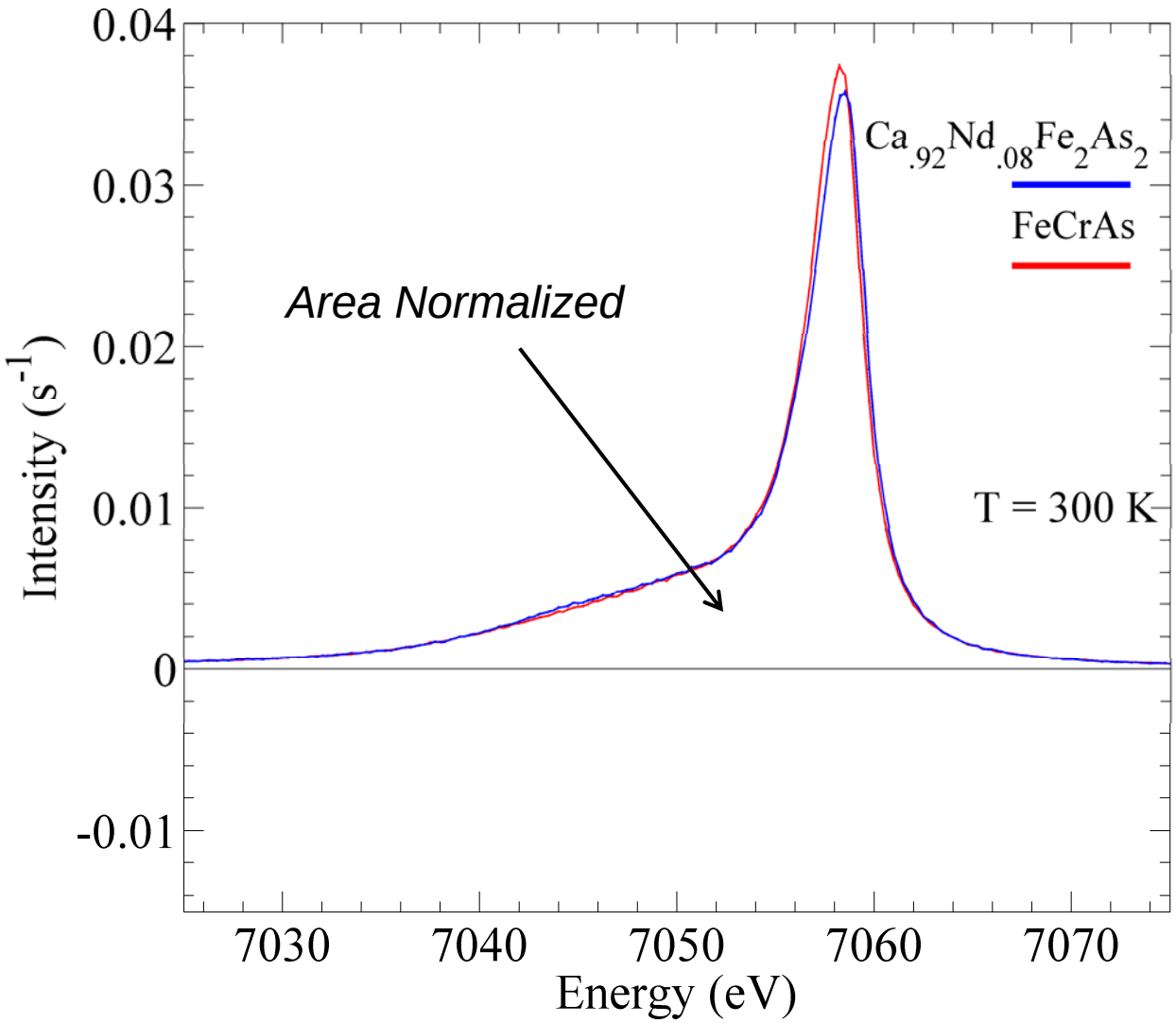
FeCrAs:

1. Same local environment as Fe-based SC (tetrahedral)
2. No magnetic moment (neutron scattering and Mossbauer)

W. Wu, S. Julian et al., EPL 2009

I. Swainson et al., Can. J. Phys. 2011

IAD analysis method



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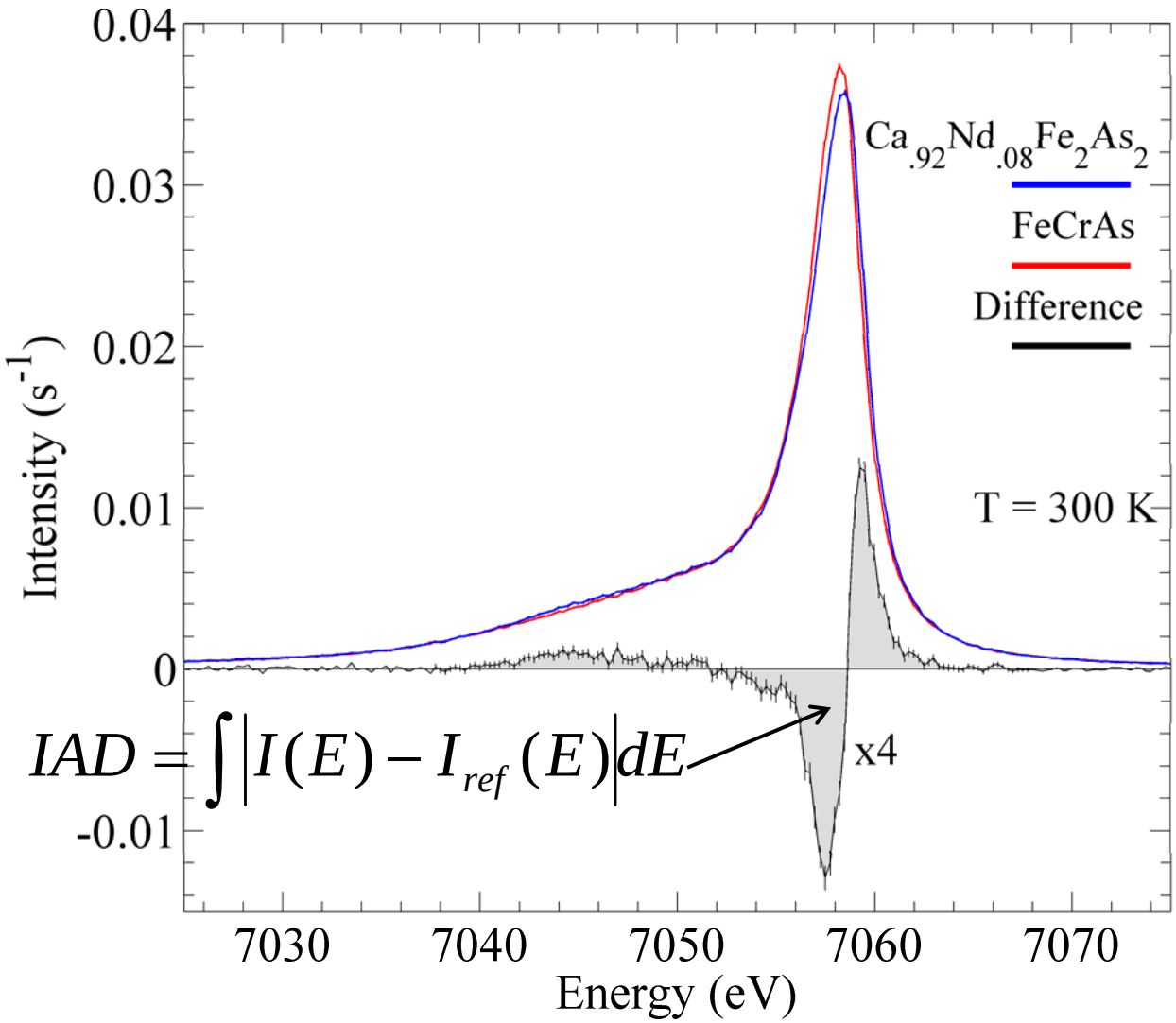

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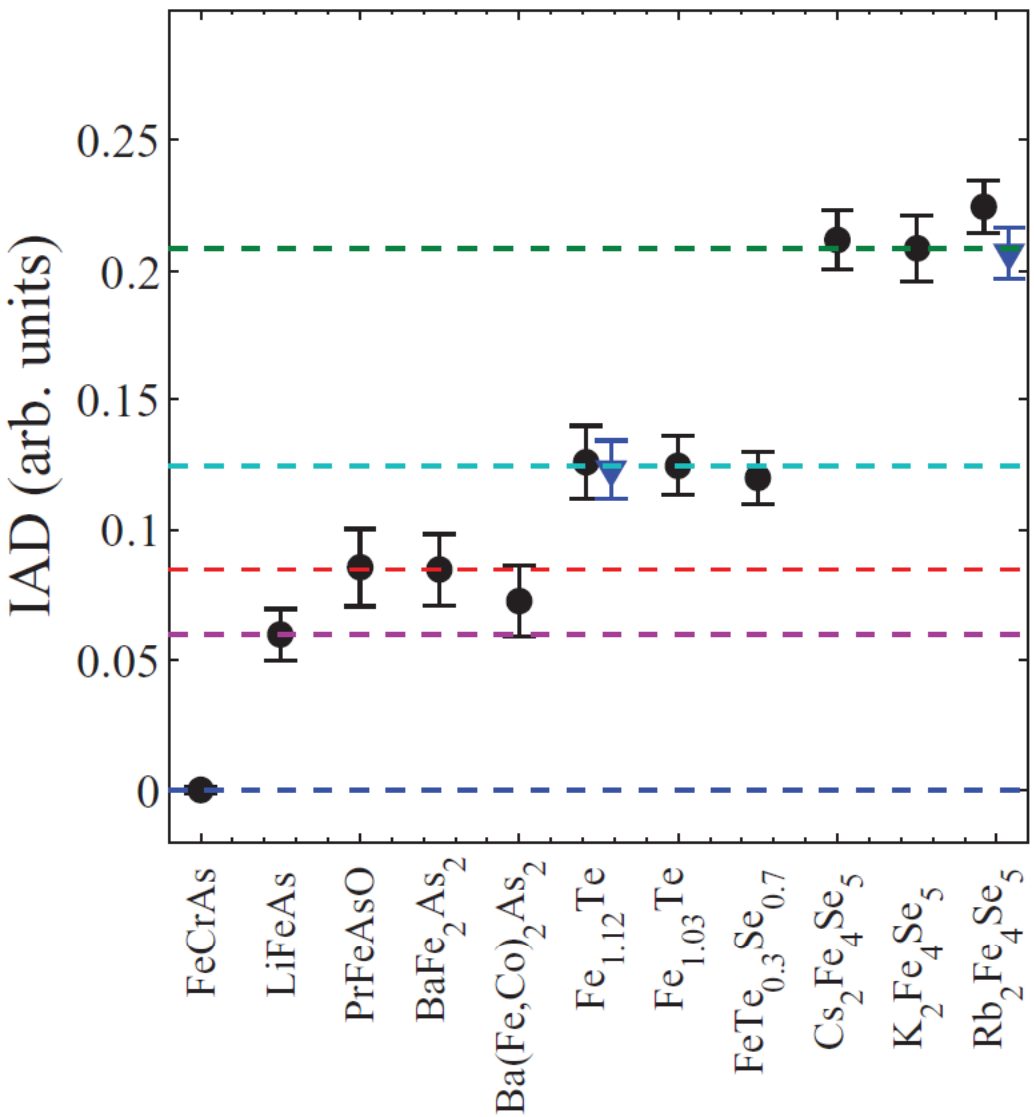
W. Wu, S. Julian et al., EPL 2009

I. Swainson et al., Can. J. Phys. 2011

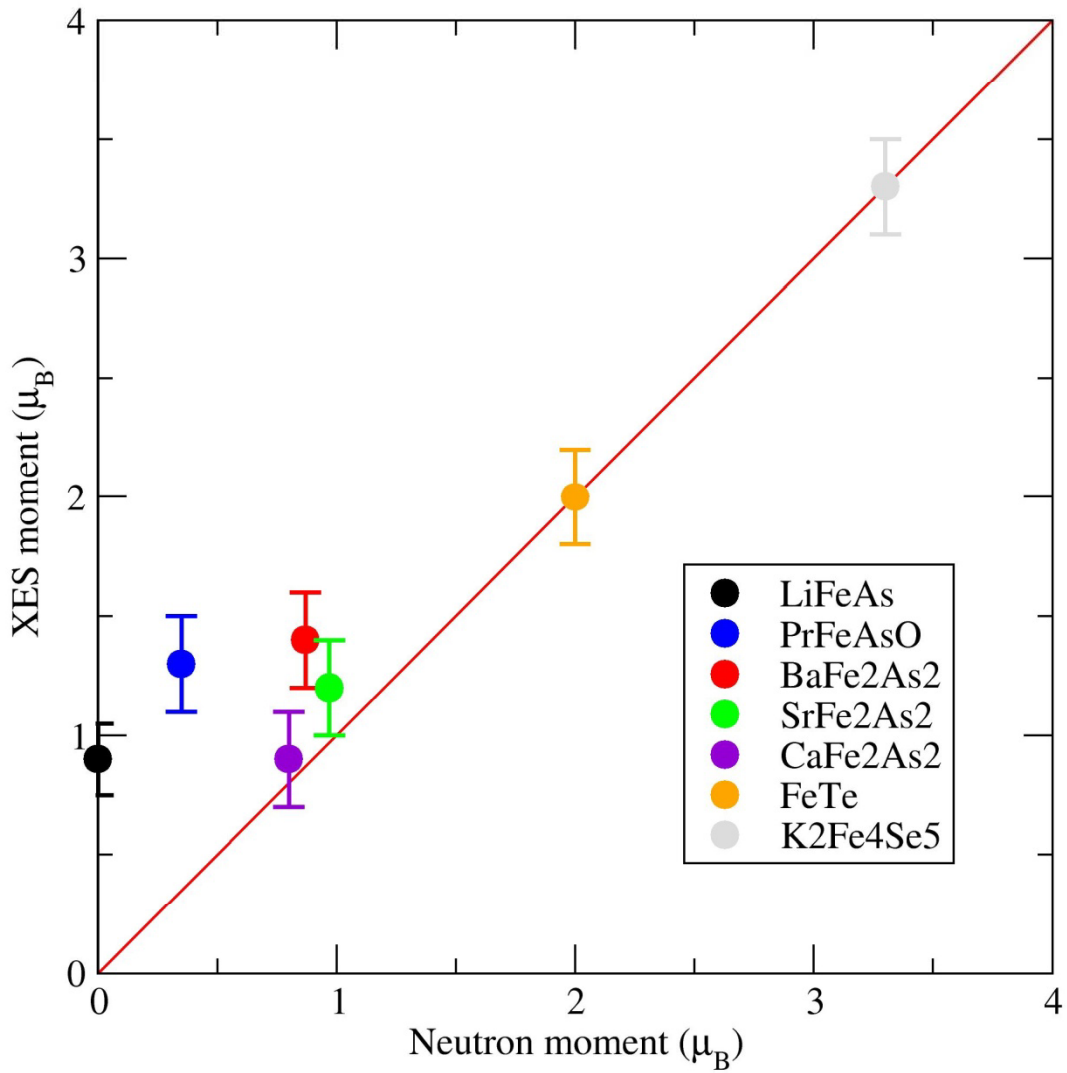
IAD is a reliable measure of the Fe spin state

Instantaneous Local Moment

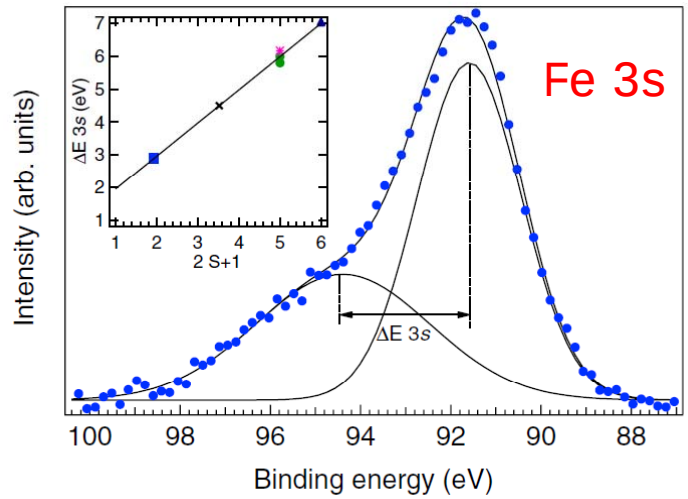


- Instantaneous : femtosecond time-scale
- Local: atomic length-scale

Ordered moment size vs. XES



CeFeAsOF ~ 1μB

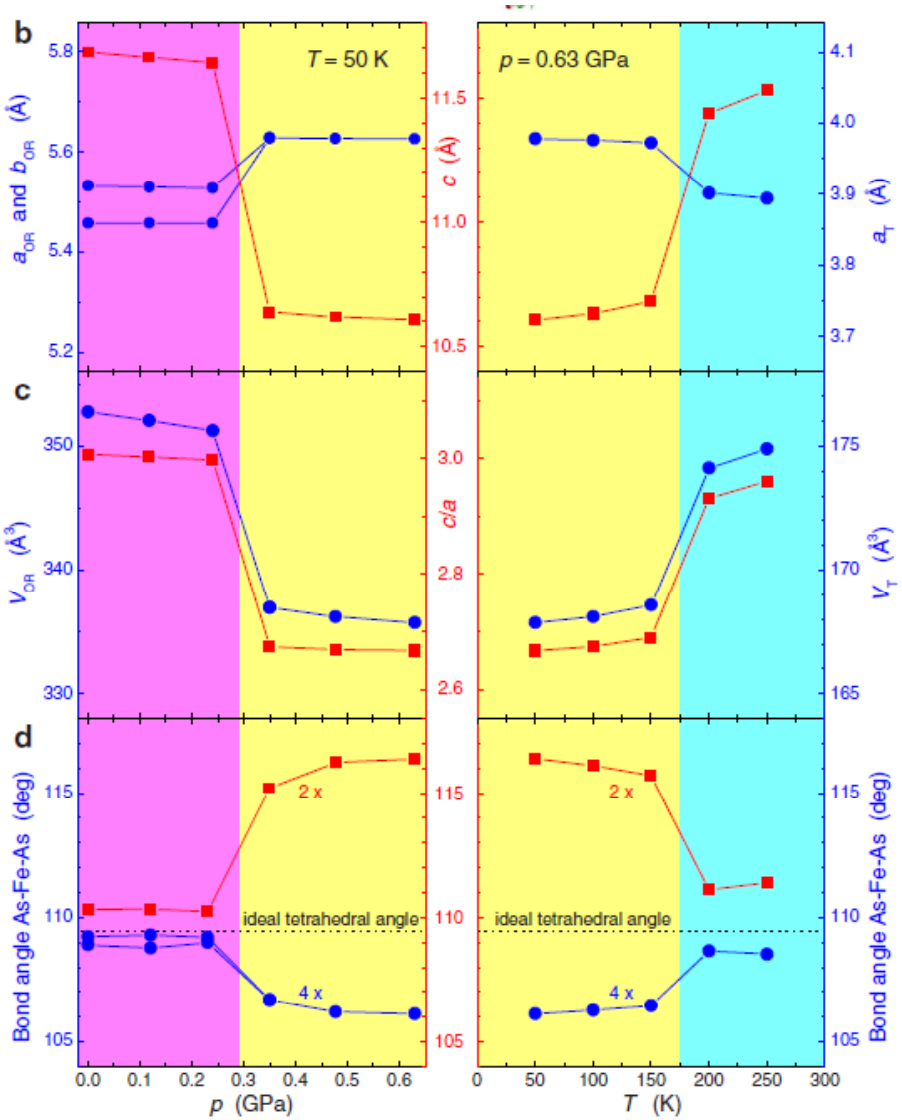


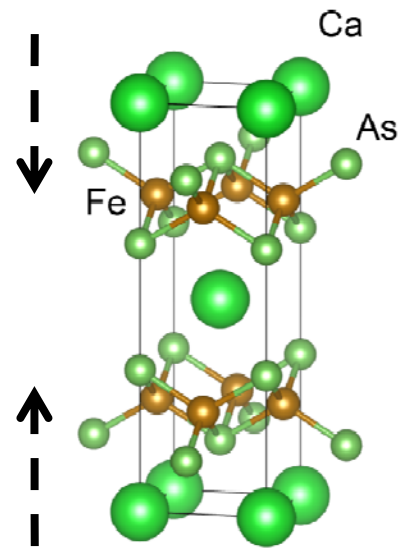
Bondino et al. PRL 2008

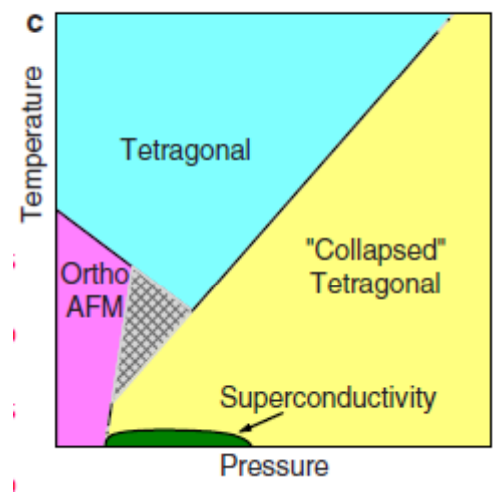
SPIN-STATE TRANSITION

Collapsed Tetragonal (cT) phase

$CaFe_2As_2$



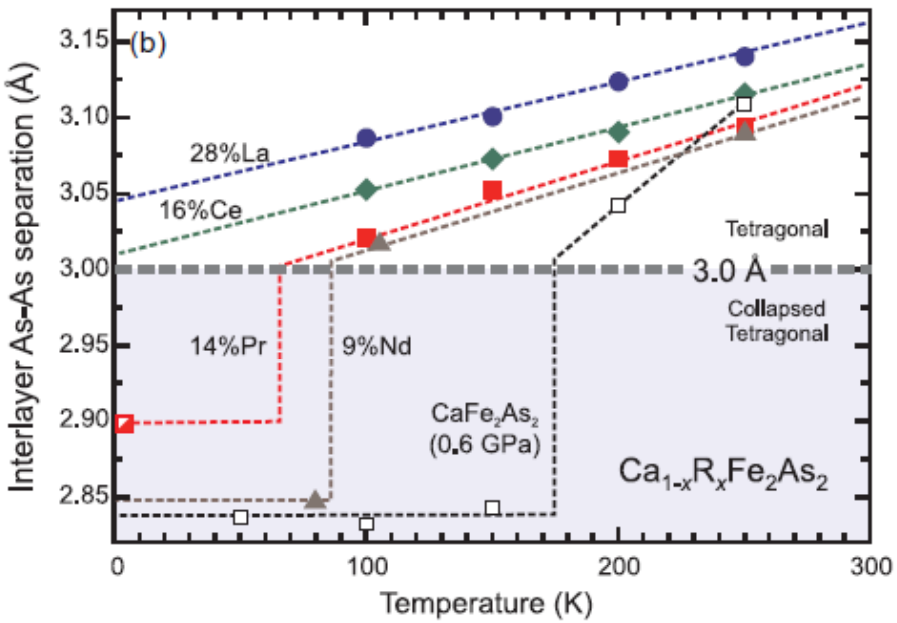




Kreyssig et al. Phys. Rev. B 78, 184517 (2008)

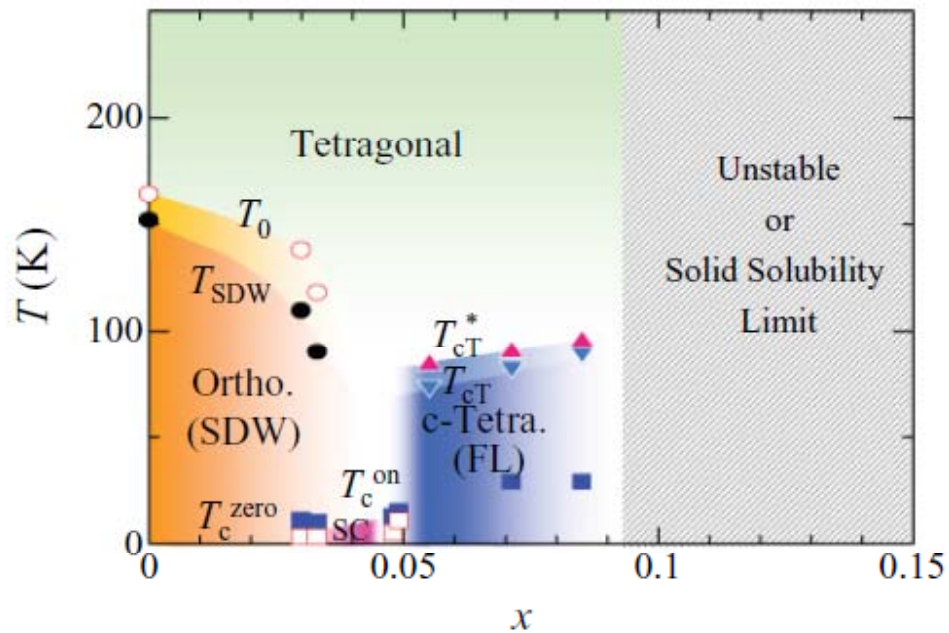
cT phase in ambient pressure

Rare-Earths doping



Saha, Paglione et al.
Phys. Rev. B
85, 024525 (2012)

P doping (As site)



Kasahara et al.
Phys. Rev. B
83, 060505 (R) (2011)

Loss of magnetism

PHYSICAL REVIEW B **79**, 024513 (2009)

Lattice collapse and quenching of magnetism in $CaFe_2As_2$ under pressure: A single-crystal neutron and x-ray diffraction investigation

A. I. Goldman,[1,2] A. Kreyssig,[1,2] K. Prokeš,[3] D. K. Pratt,[1,2] D. N. Argyriou,[3] J. W. Lynn,[4] S. Nandi,[1,2] S. A. J. Kimber,[3] Y. Chen,[4,5] Y. B. Lee,[1,2] G. Samolyuk,[1,2] J. B. Leão,[4] S. J. Poulton,[4,5] S. L. Bud'ko,[1,2] N. Ni,[1,2] P. C. Canfield,[1,2] B. N. Harmon,[1,2] and R. J. McQueeney[1,2]

PHYSICAL REVIEW B **79**, 060510(R) (2009)

Suppression of antiferromagnetic spin fluctuations in the collapsed phase of $CaFe_2As_2$

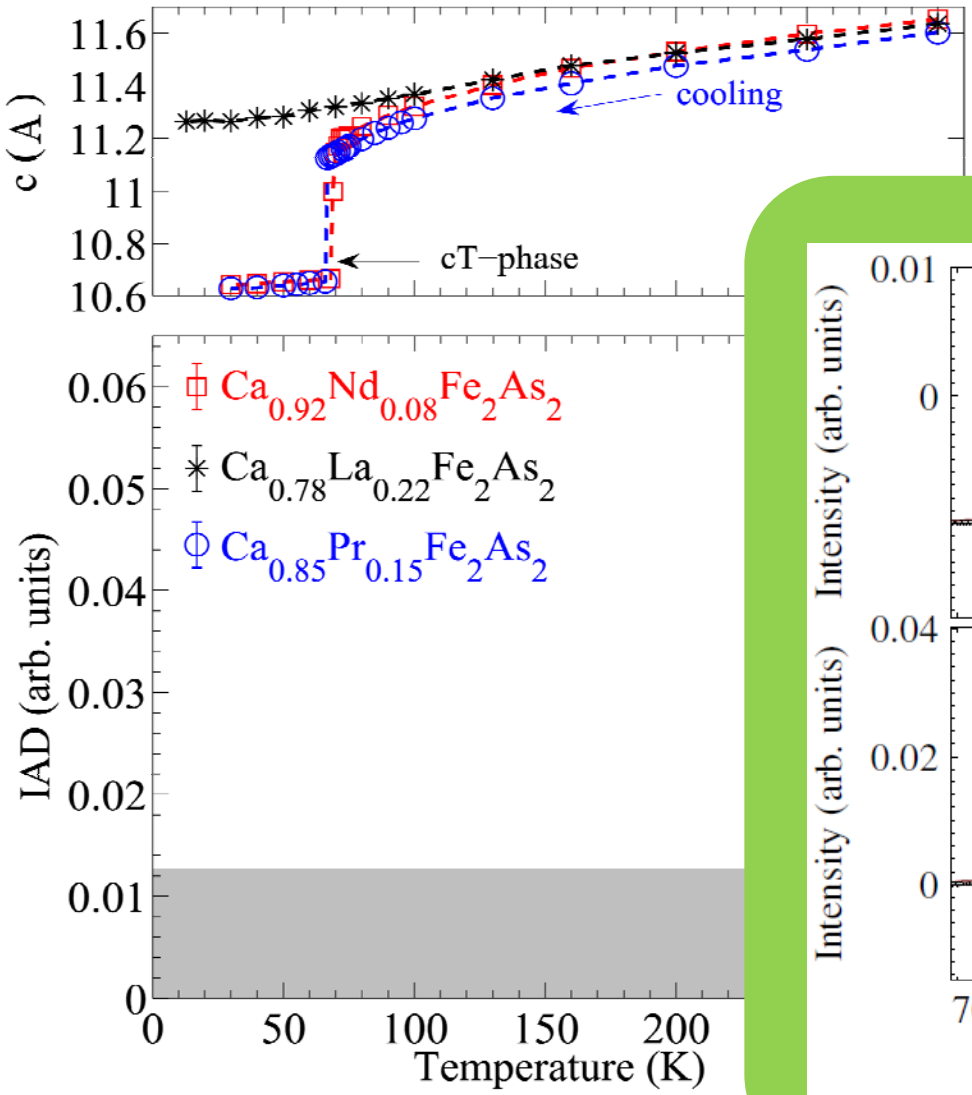
D. K. Pratt,[1,2] Y. Zhao,[3] S. A. J. Kimber,[4] A. Hiess,[5] D. N. Argyriou,[4] C. Broholm,[3] A. Kreyssig,[1,2] S. Nandi,[1,2] S. L. Bud'ko,[1,2] N. Ni,[1,2] P. C. Canfield,[1,2] R. J. McQueeney,[1,2] and A. I. Goldman[1,2]

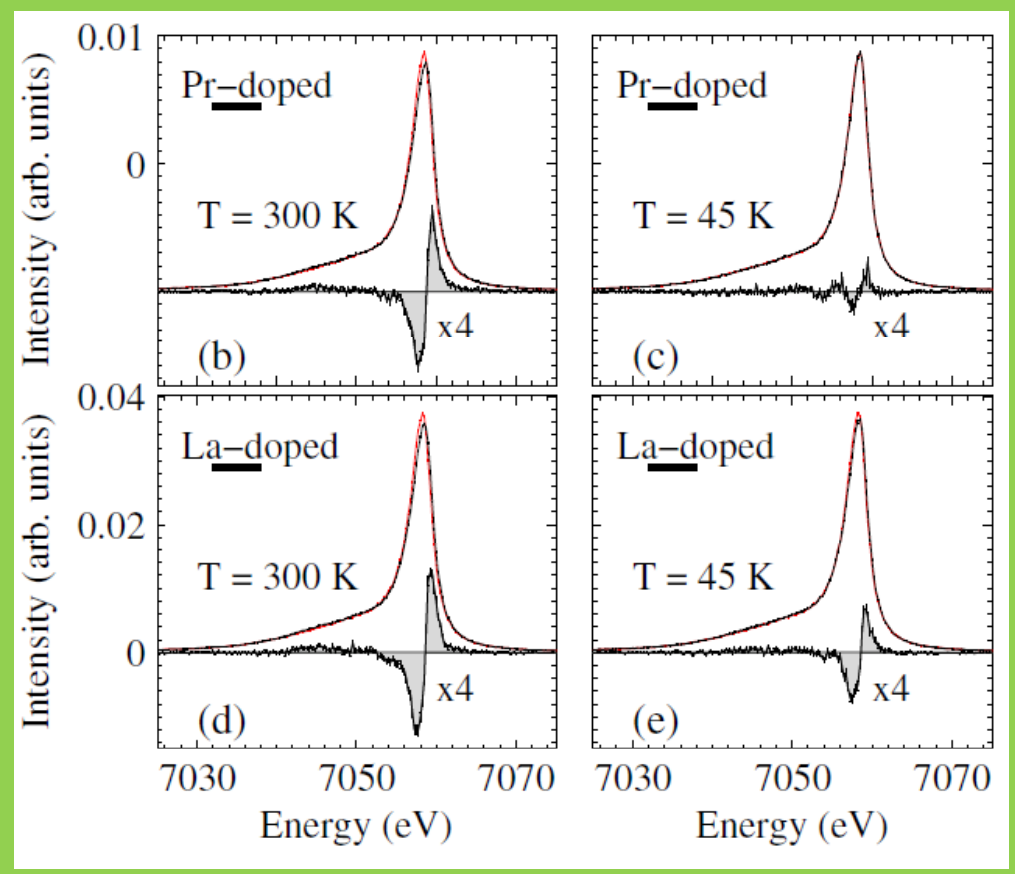
PRL **102,** 037003 (2009) PHYSICAL REVIEW LETTERS week ending 23 JANUARY 2009

Strong Coupling of the Fe-Spin State and the As-As Hybridization in Iron-Pnictide Superconductors from First-Principle Calculations

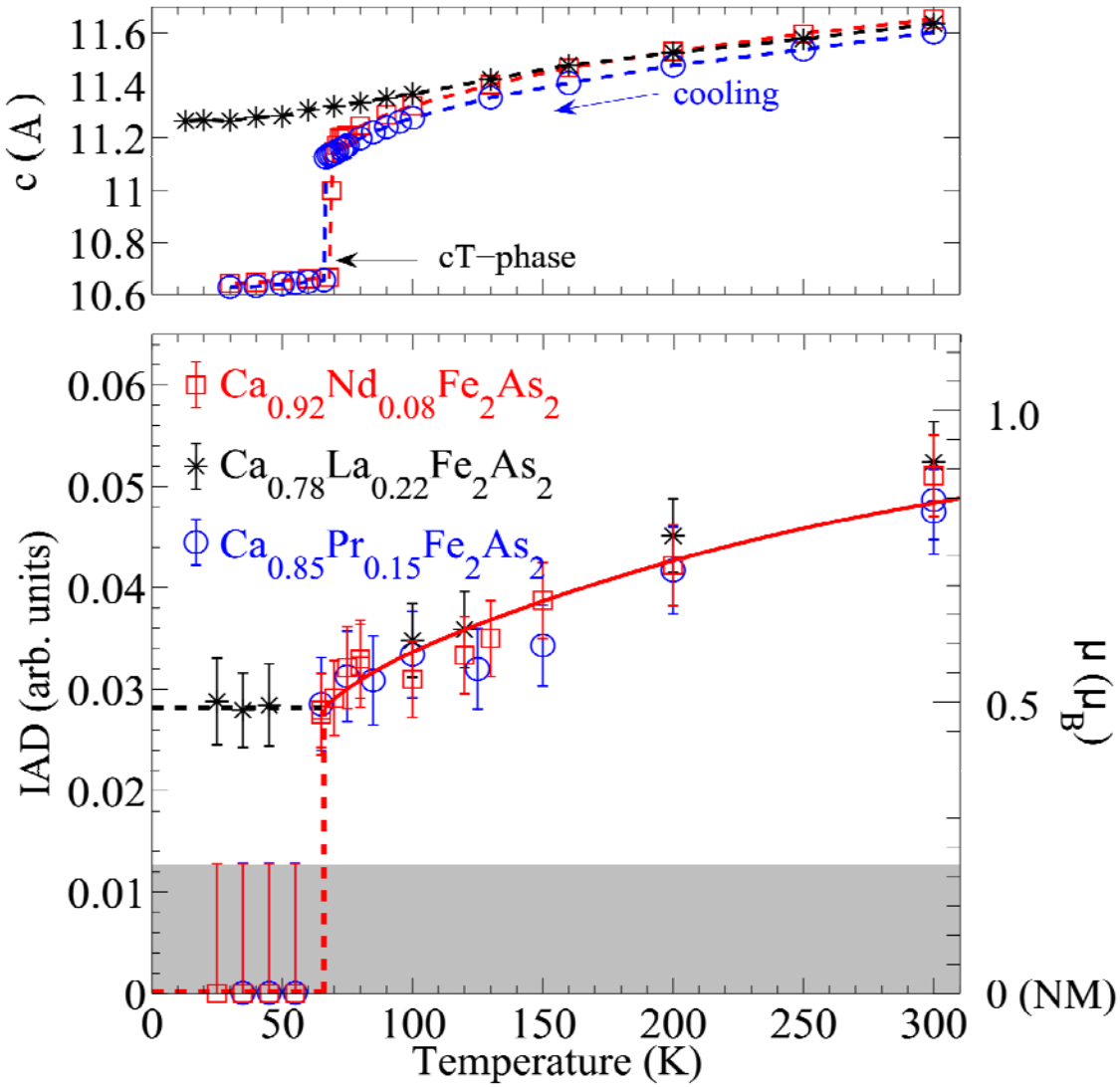
T. Yildirim[1,2,*]

RE doped $CaFe_2As_2$





RE doped $CaFe_2As_2$



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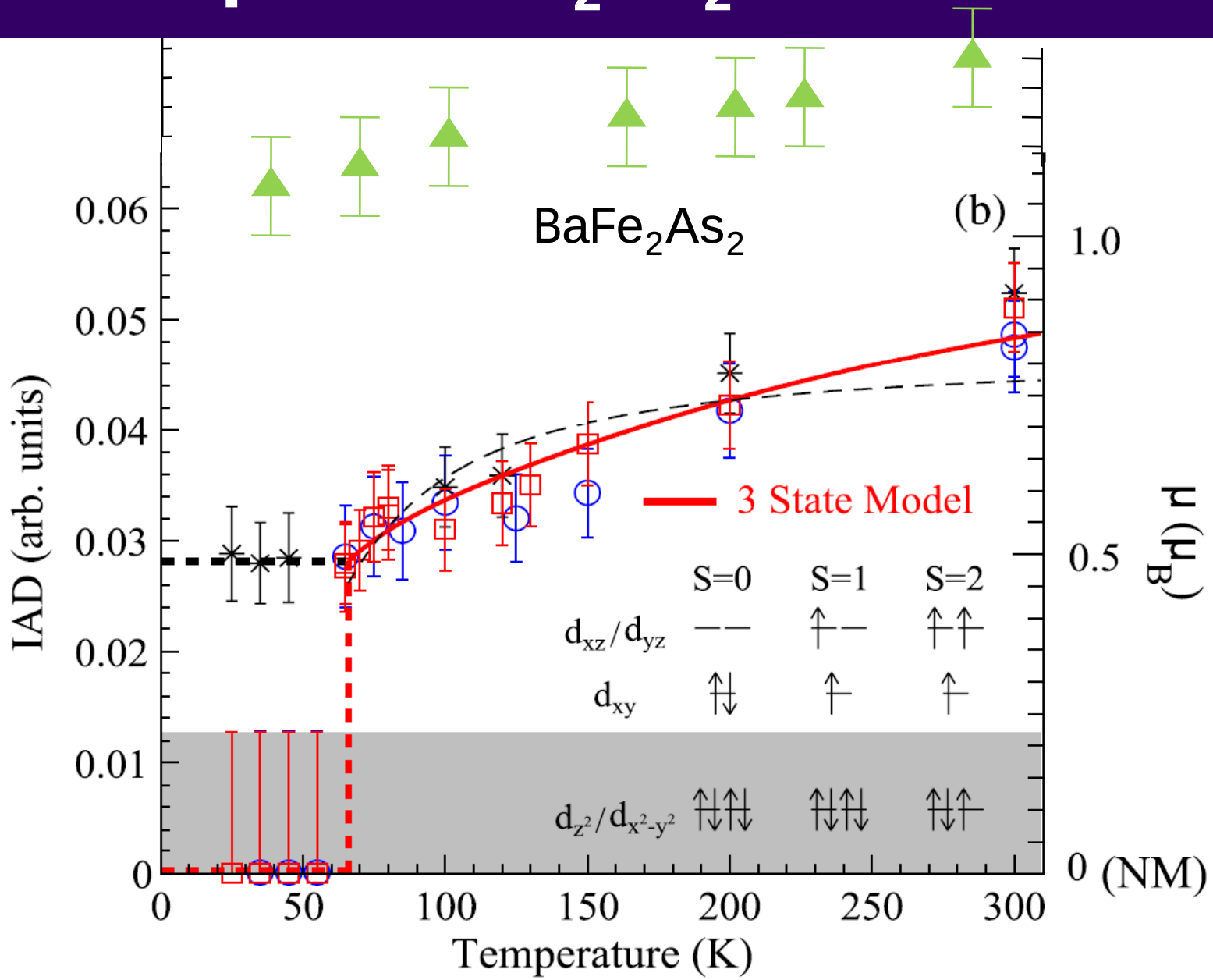
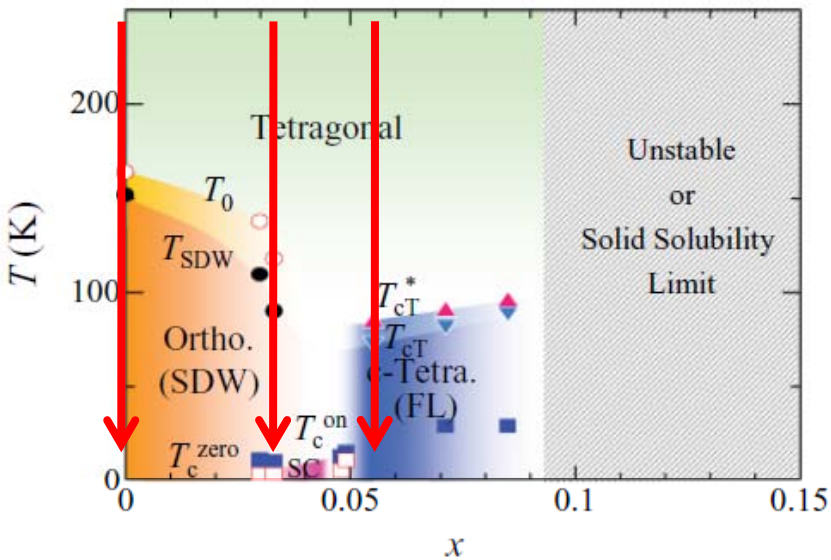
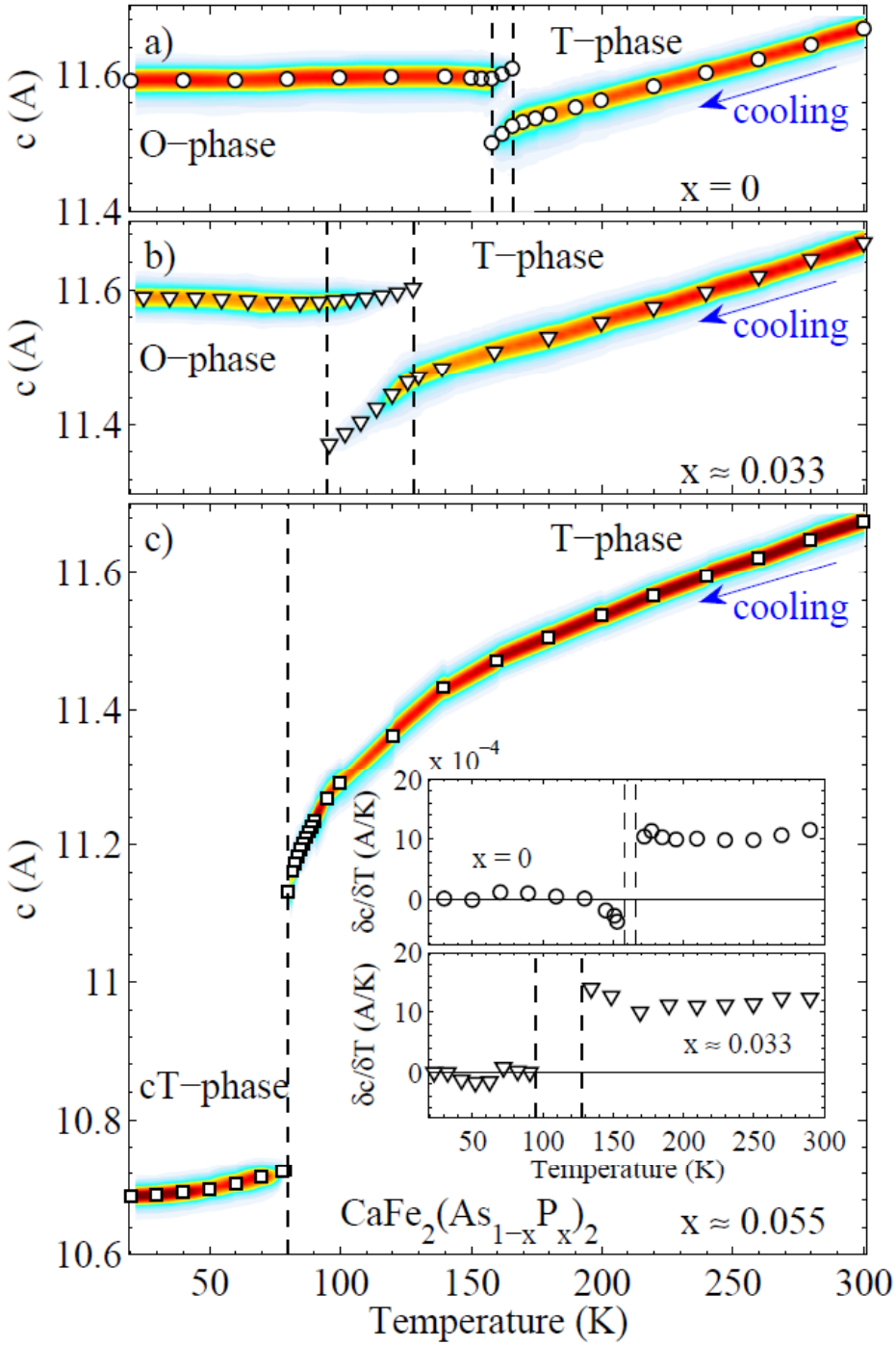


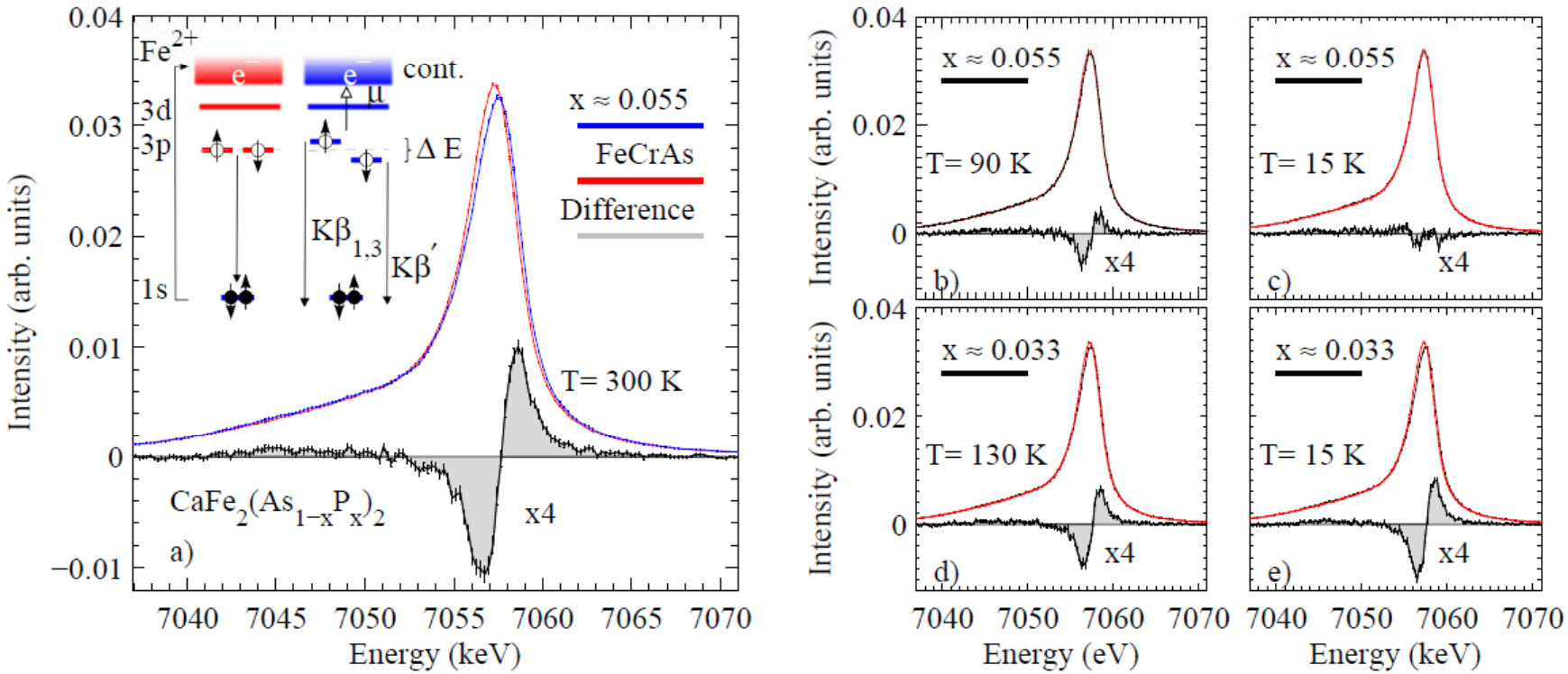
$CaFe_2(As,P)_2$



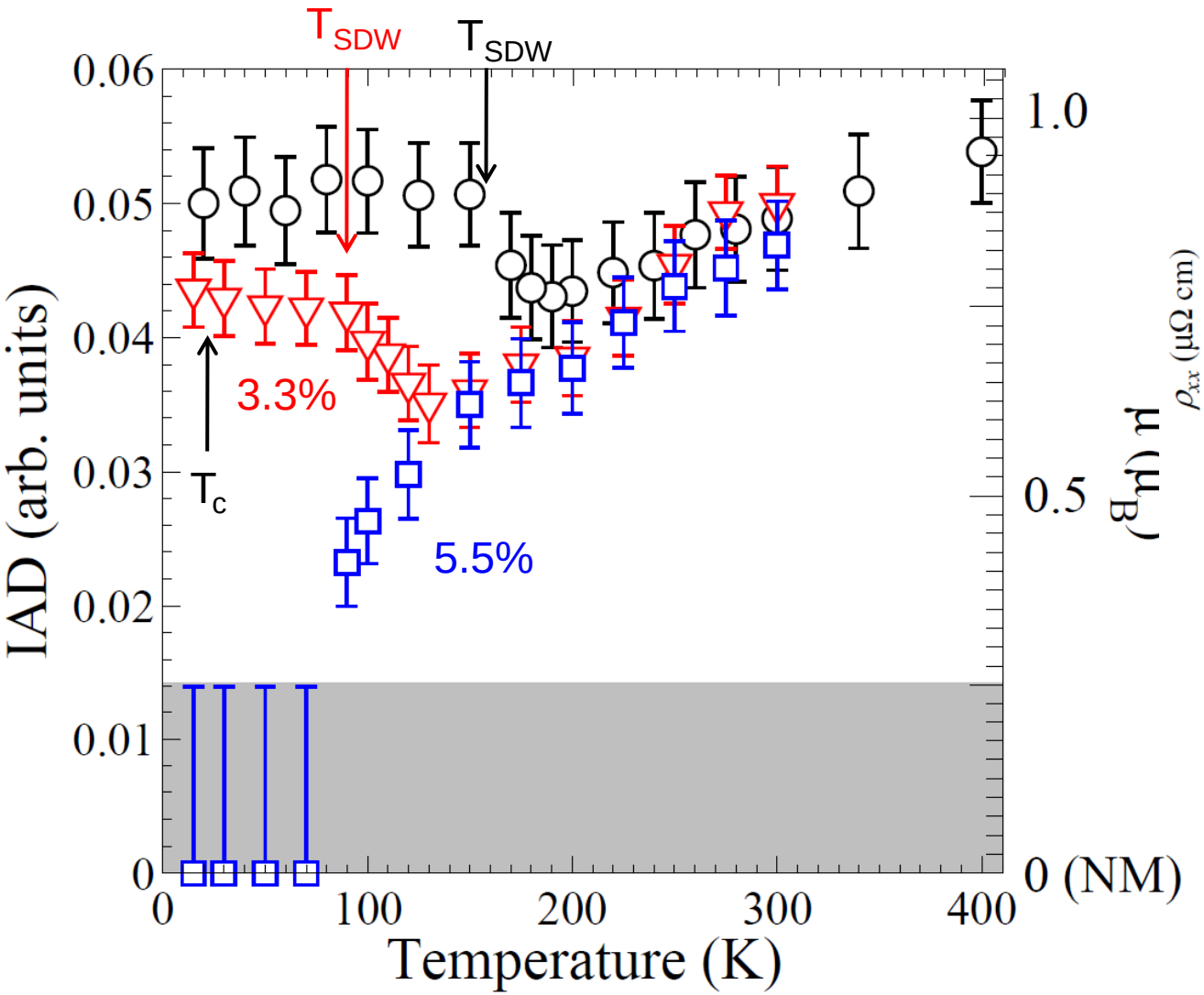
Kasahara et al.
Phys. Rev. B
83, 060505 (R) (2011)

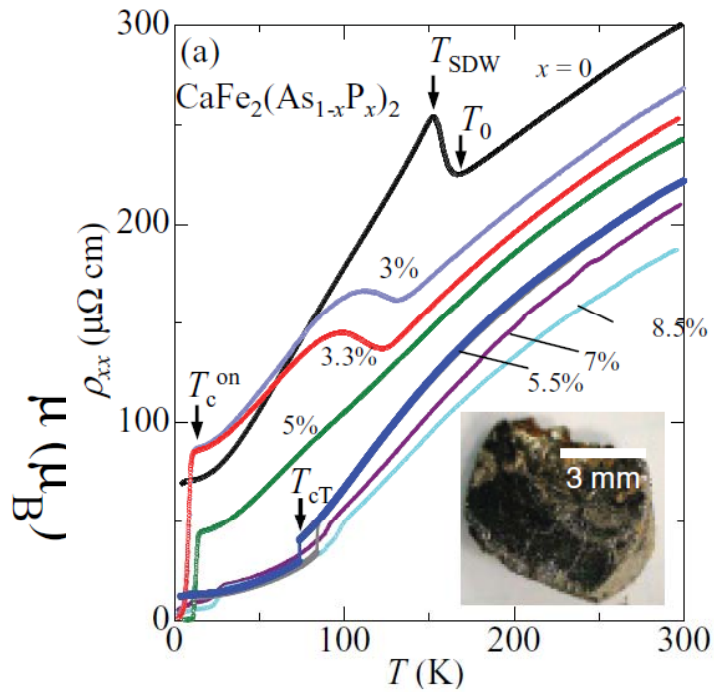


XES spectra

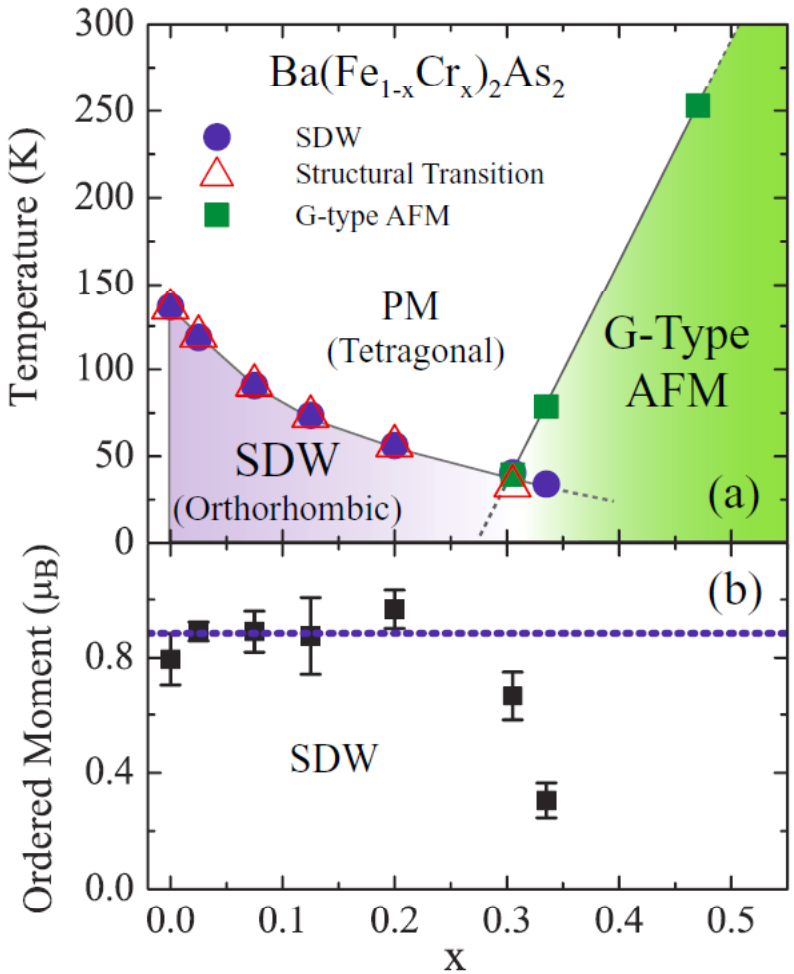

Fe2+
cont.
3d
3p
1s
μ
}Δ E
Kβ1,3
Kβ′
x ≈ 0.055
FeCrAs
Difference
T= 300 K
CaFe2(As1−xPx)2
x4
a)
Intensity (arb. units)
Energy (keV)
x ≈ 0.055
T= 90 K
b)
T= 15 K
c)
x ≈ 0.033
T= 130 K
d)
T= 15 K
e)
Energy (eV)

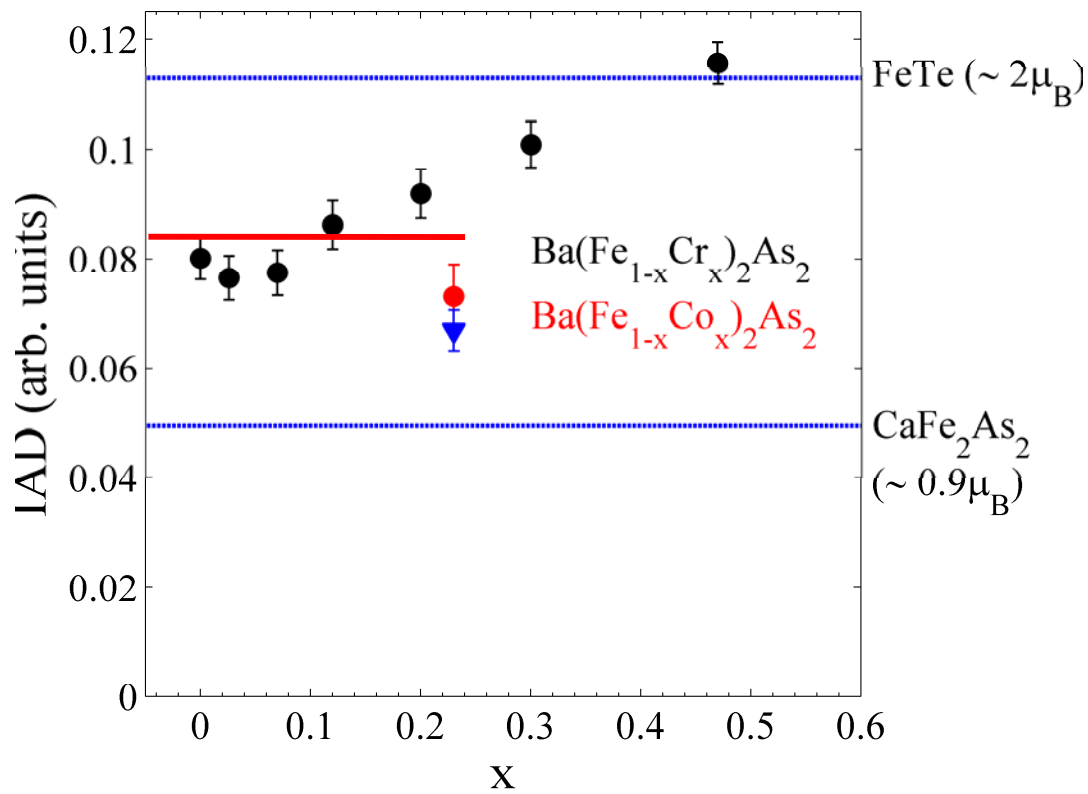
Absence of SC in low-spin state





Cr doped $BaFe_2As_2$





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Conclusions

- X-ray Emission Spectroscopy provides
  - Instantaneous measurement of
  - Fluctuating local moment
- All pnictides have about ~1 μ_B fluctuating local moment at all temperatures
- Except for $CaFe_2As_2$ in collapsed tetragonal phase (through Rare-Earths or P doping)
- Loss of fluctuating local moment in cT phase coincides with the absence of SC in 5.5% P doped $CaFe_2As_2$.
- Spin-crossover model applicable to $CaFe_2As_2$.