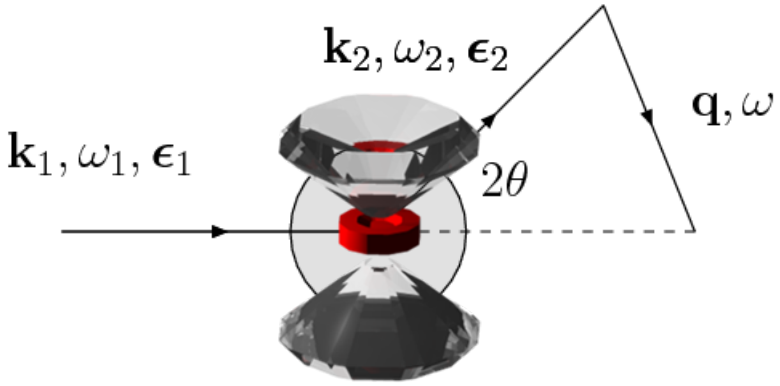
IXS in correlated materials under high pressure

Jean-Pascal RUEFF

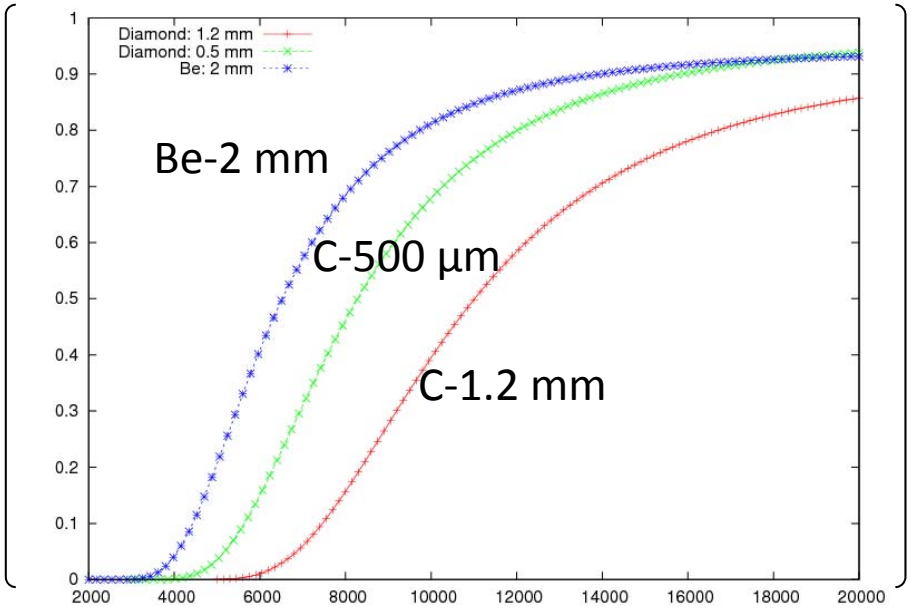
First high pressure RIXS - ID16

Some 10 years ago…

High pressure RIXS

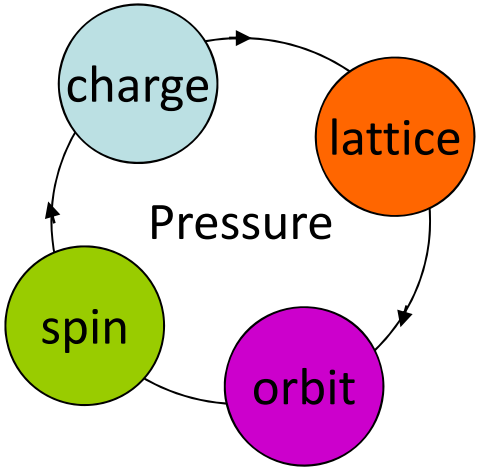


Transmission



^{2} ⊗ scattering volume

Why high pressure ?



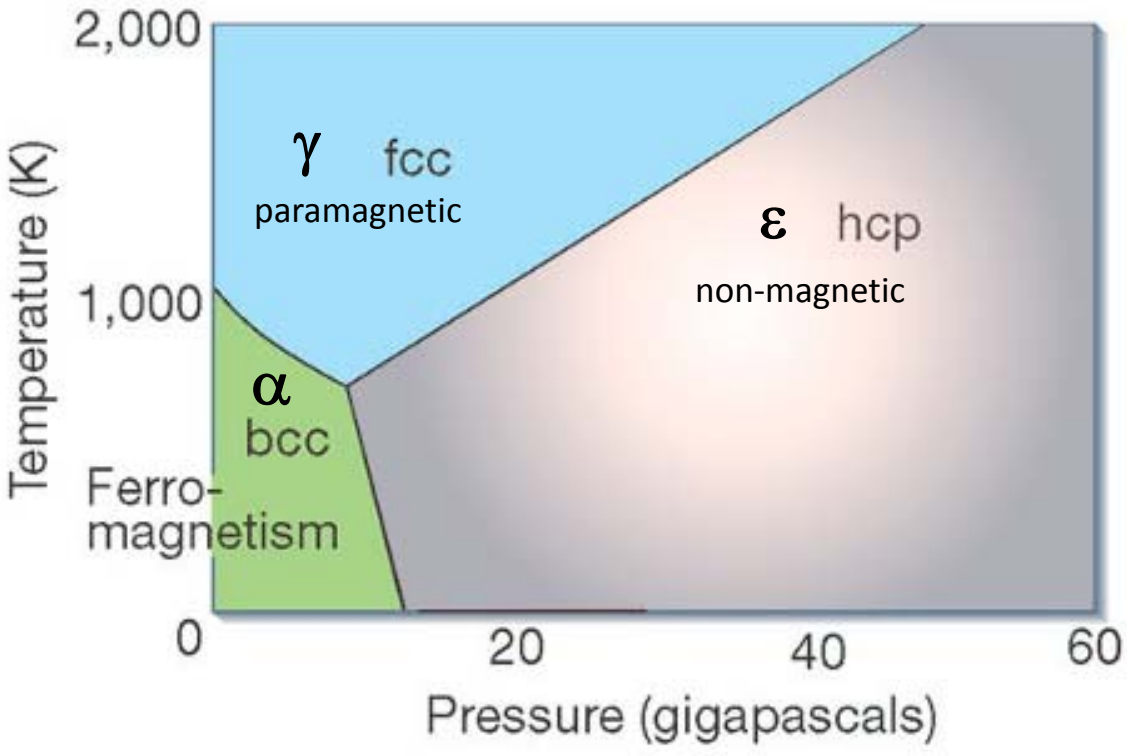
- electron delocalization
- metal-insulator transition
- crystal field enhancement
- magnetic collapse
- structural changes

High pressure spectroscopy: Optical, IR, Raman, neutron,
> hard x-ray spectroscopy [**IXS**]

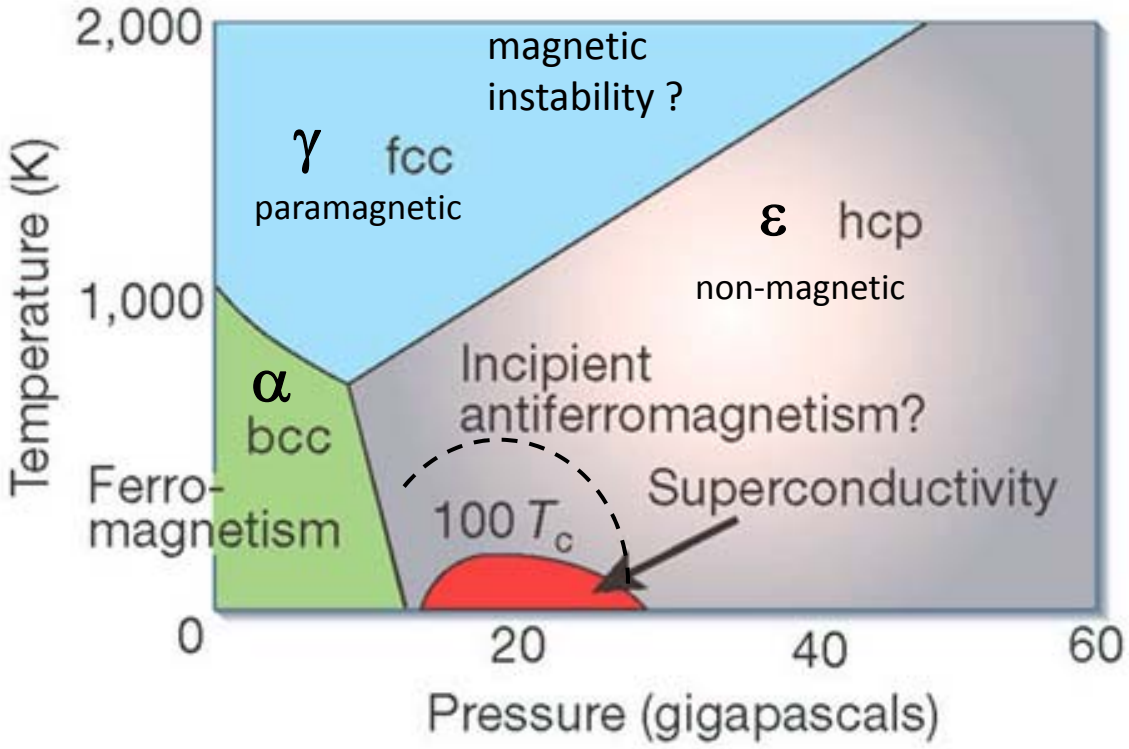
Outline

- Fe, magnetic collapse and more
- Metal insulator transition in V_2O_3
- SmS: valence instabilities in f electrons systems

Fe

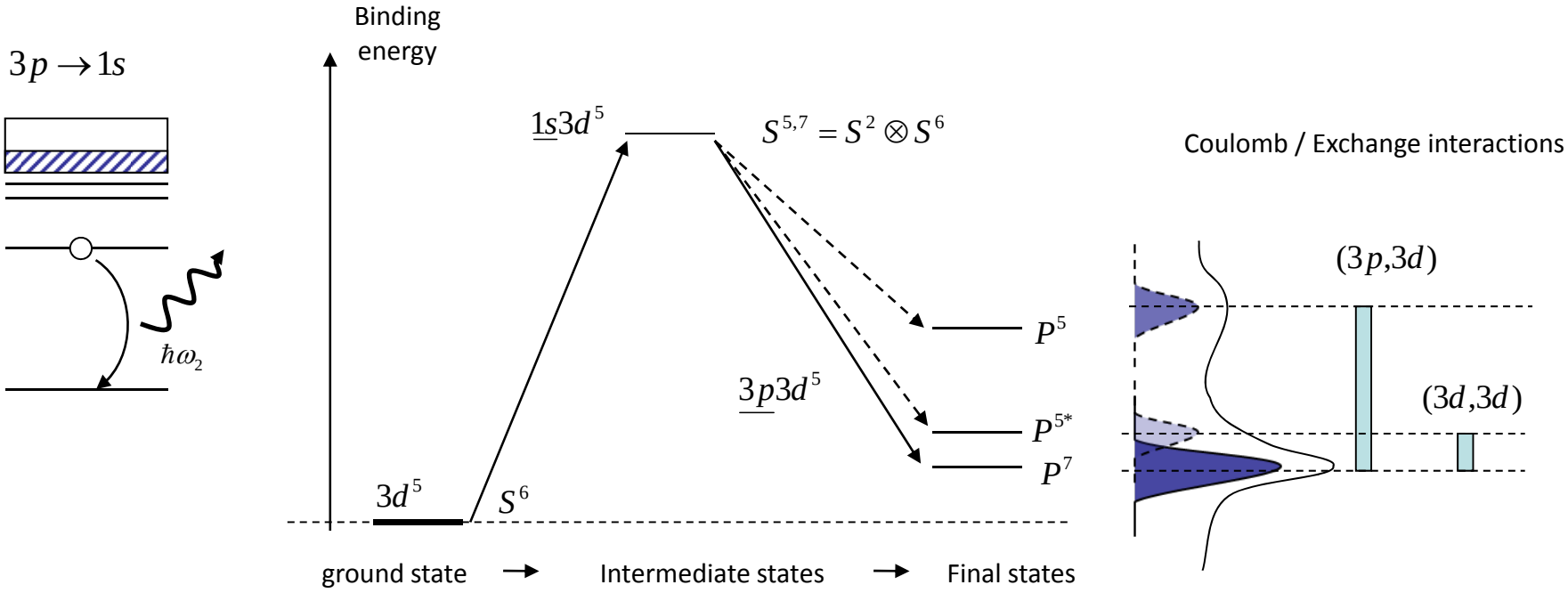

2,000
1,000
0
Temperature (K)
γ fcc
paramagnetic
ε hcp
non-magnetic
α
bcc
Ferro-
magnetism
20
40
60
Pressure (gigapascals)

Fe



*Shimizu et al., Nature, **412**, 316-318 (2001)*
*Merkel et al., Science, **288**, 1626 (2000)*
*Steinle-Neumann et al. Proc. Natl. Acad. Sci., **101**, 33-36 (2004)*
*Thakor et al., Phys. Rev. B, **67**, 180405 (2003)*

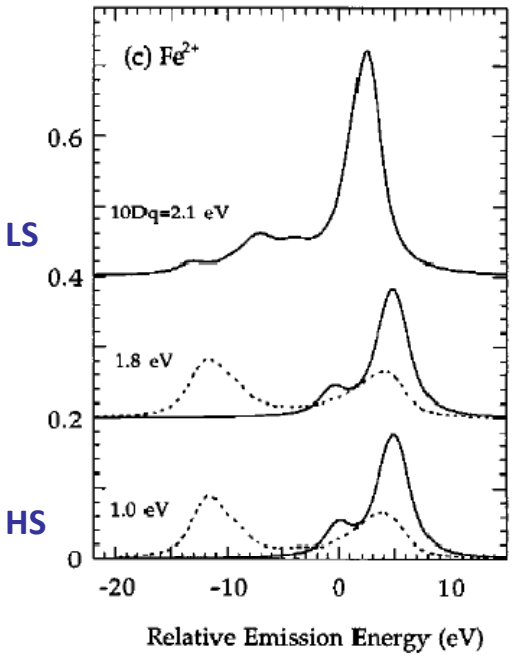
Kβ XES



- local probe of the 3d magnetism in transition metal
- No applied magnetic field
- Compatible with high pressure

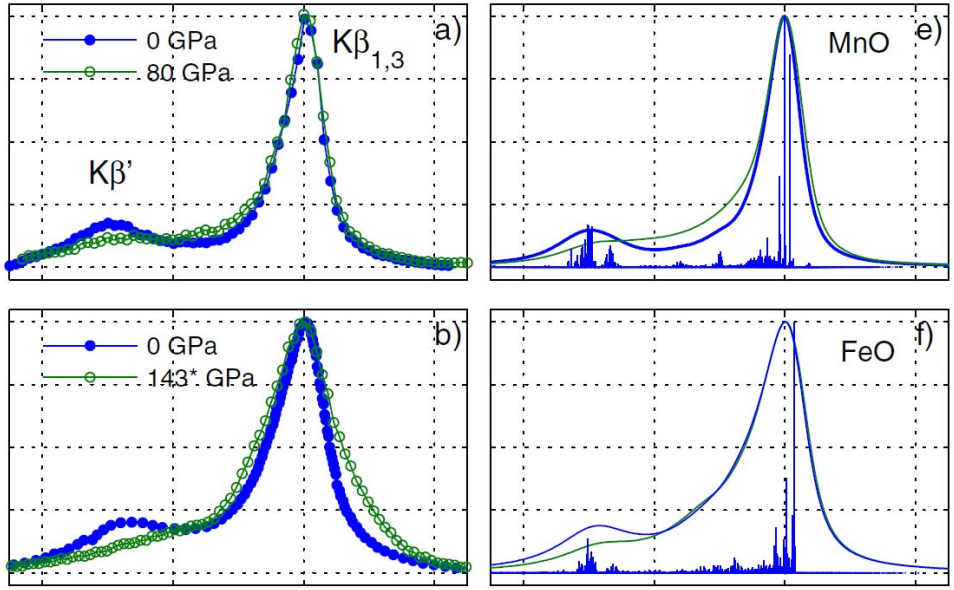
Multiplet calculations

+ ligand field



*X. Wang et al., PRB **56**, 4553 (1997)*

+ charge transfer

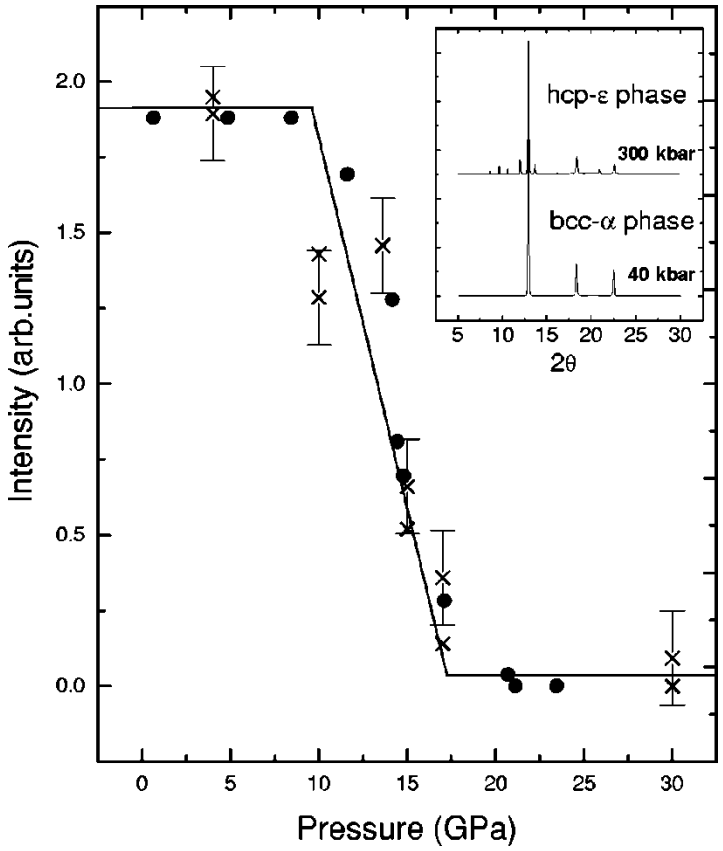


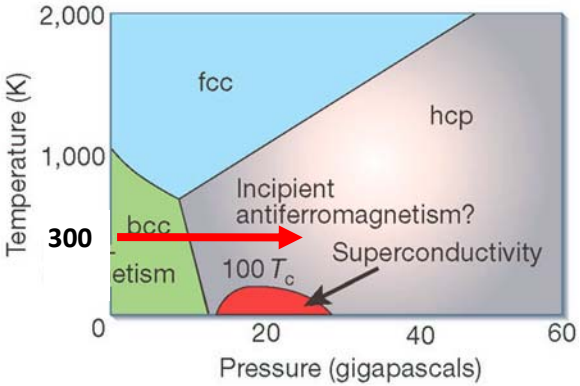
*A. Mattila et al., PRL **98**, 196404 (2007)*

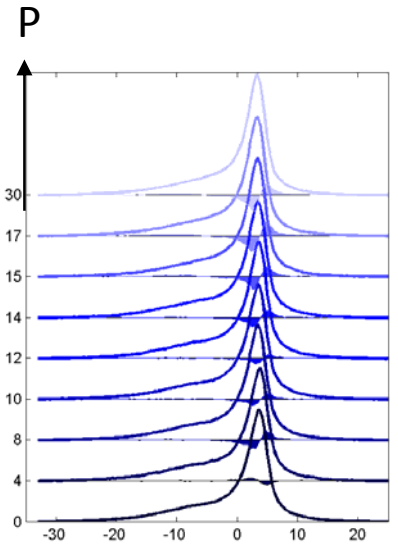
- The spectral lineshape depends (only) on the spin state
- Charge transfer effects affects the spectrum
- Spin state transition is evidenced by a decrease of the satellite intensity

Fe – HP, 300 K

Fe α-ε phases





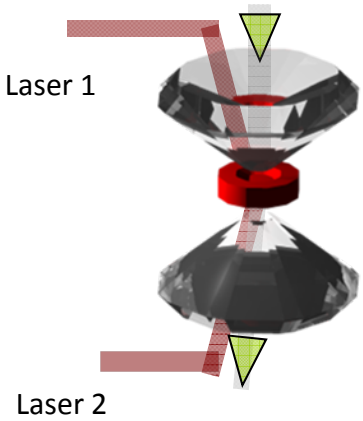


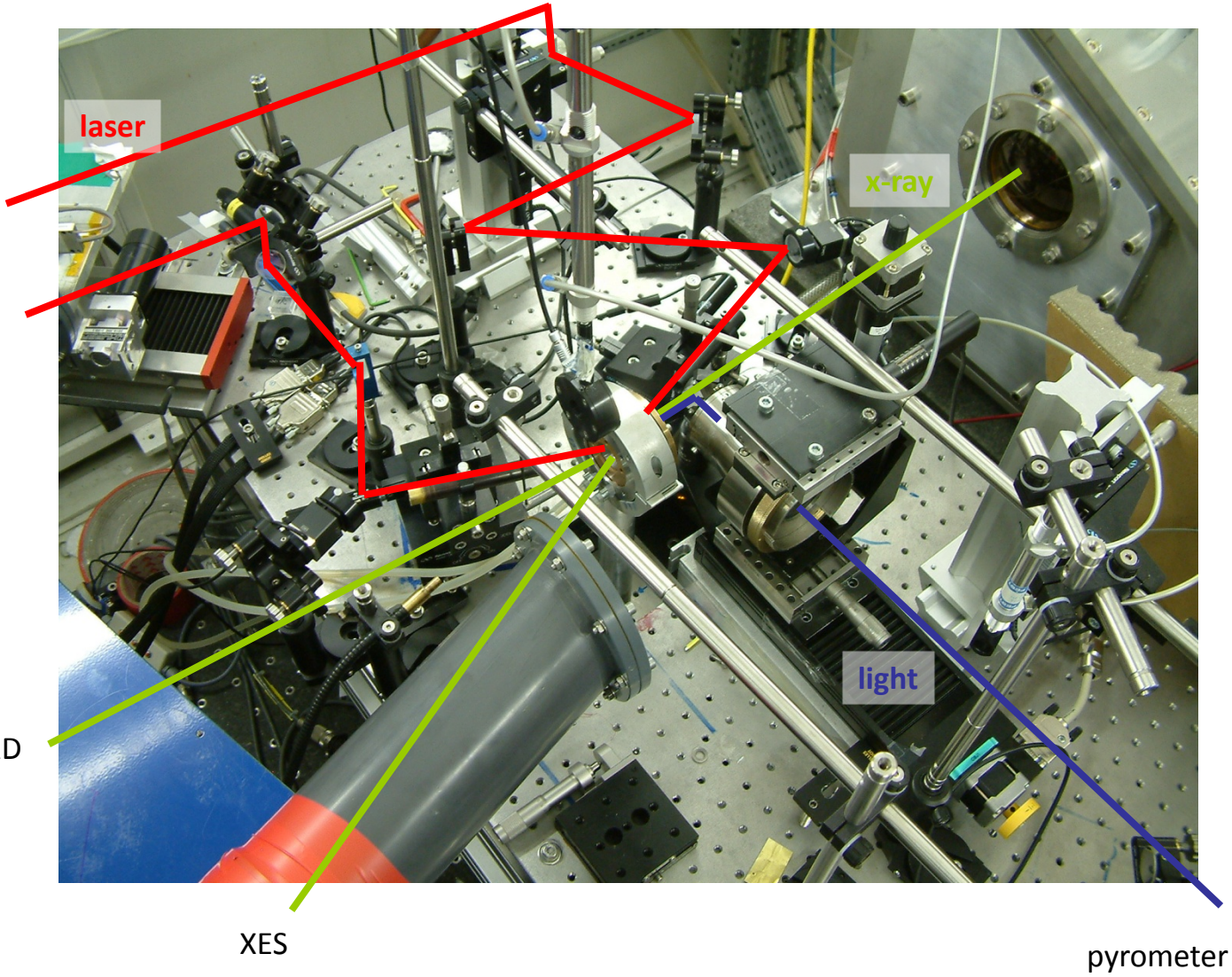
- no formal sum rule
- use of the difference spectra to extract the spin state

JPR et al., Phys. Rev. B, **60**, 14510 (1999)

XES - Laser heating

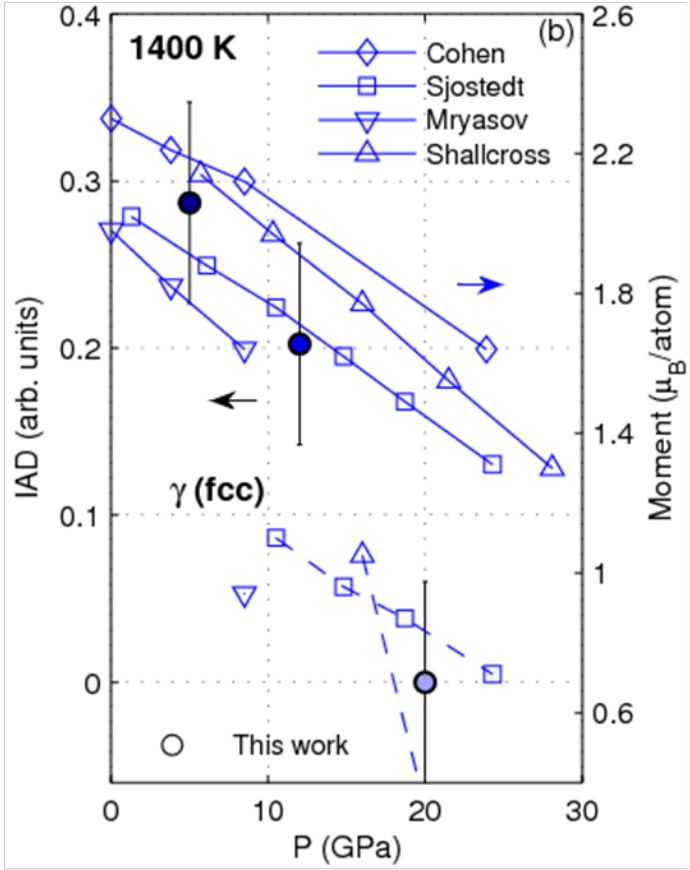
2 microns x-ray spot
50 microns laser spot

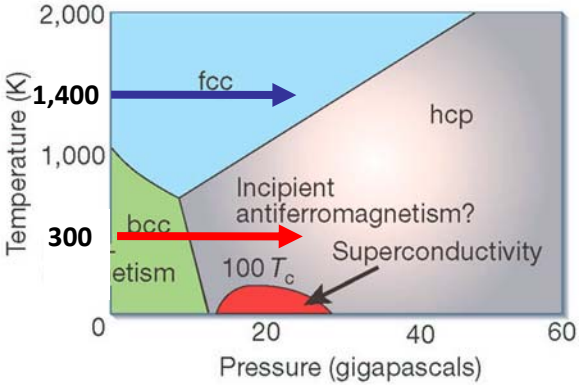




Fe – HP, 1400 K

Fe γ phase

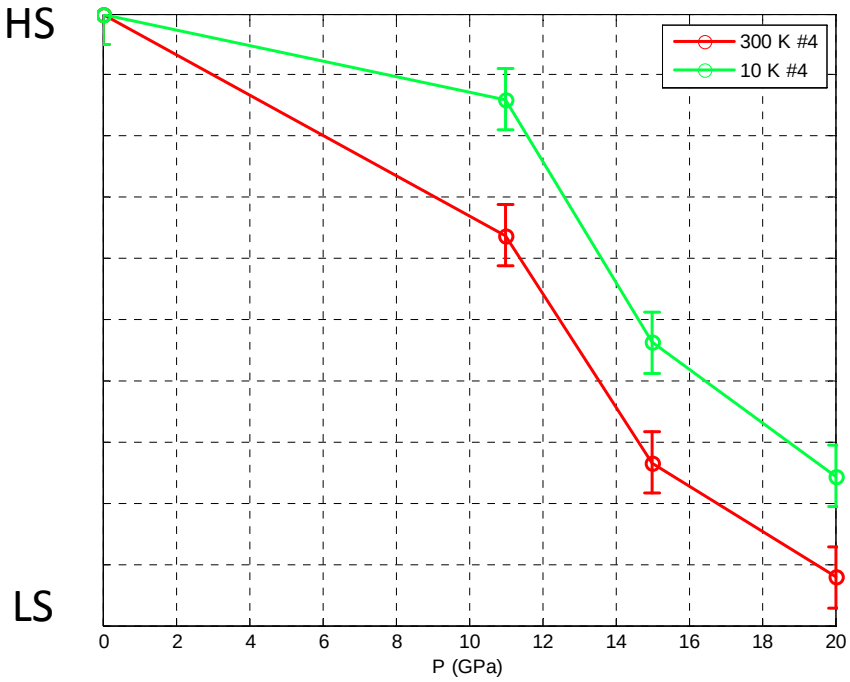


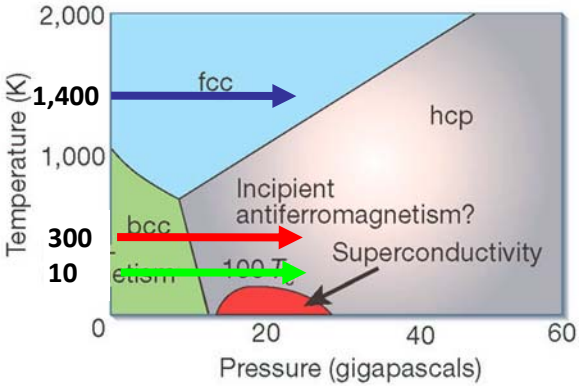


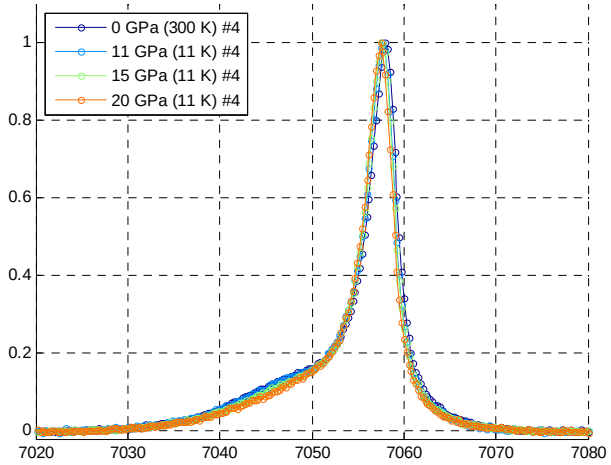
- Evidence of a magnetic transition in the γ phase
- non magnetic state obtained at high pressure
- influence of non collinear structure ?

*JPR et al., PRB **78**, 100405(R) (2008)*

Fe – HP, 10 K

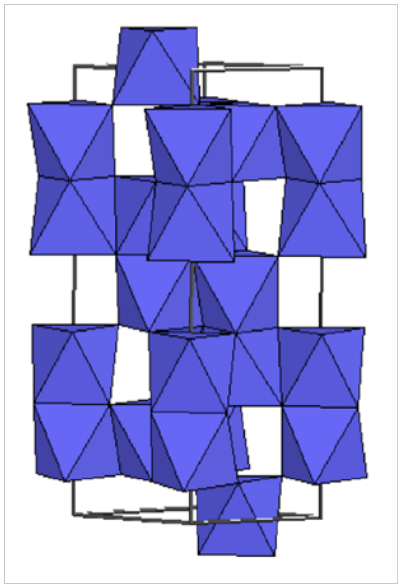


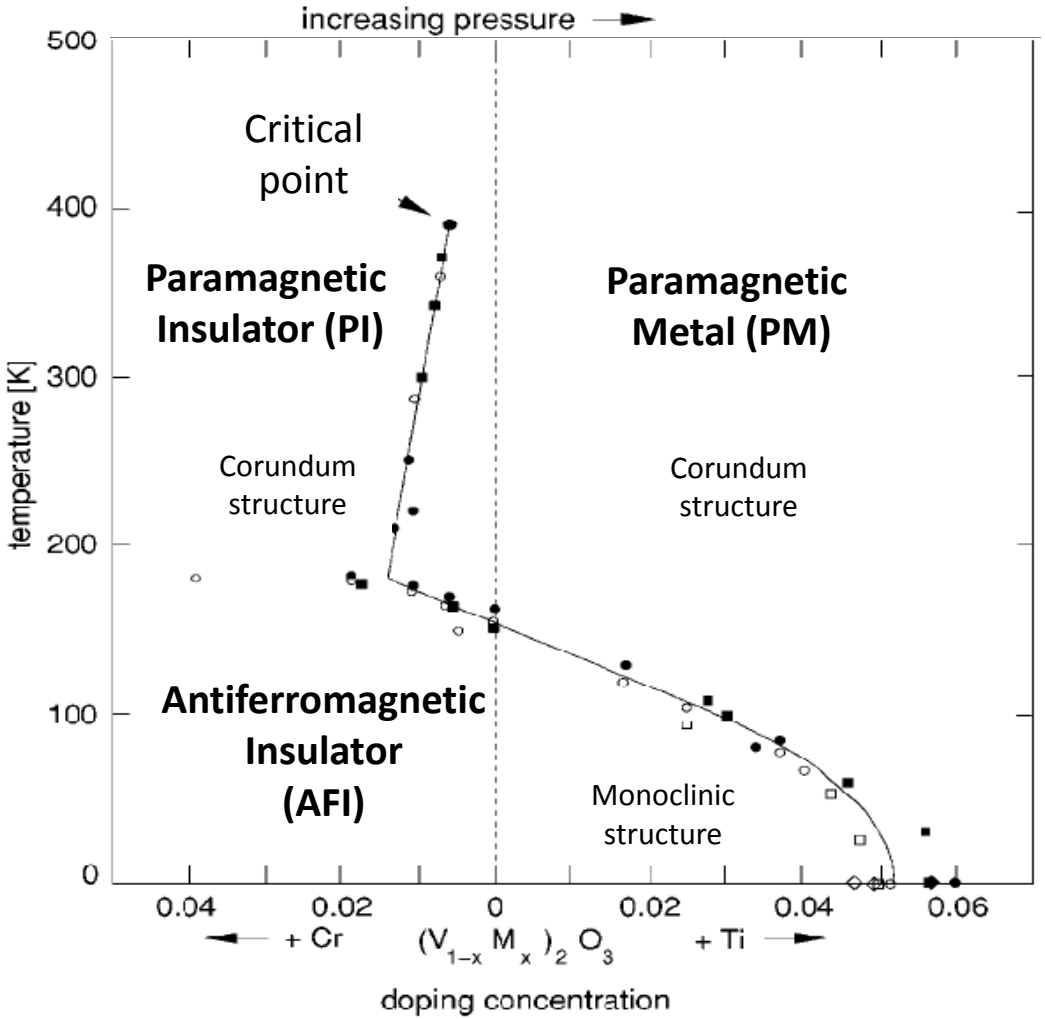




- non evidence of a low temperature anomaly
- but few data points

Metal insulator transition in V_2O_3



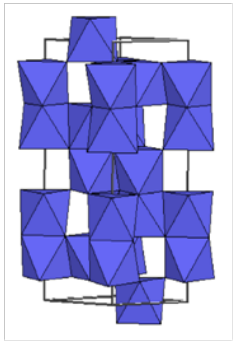


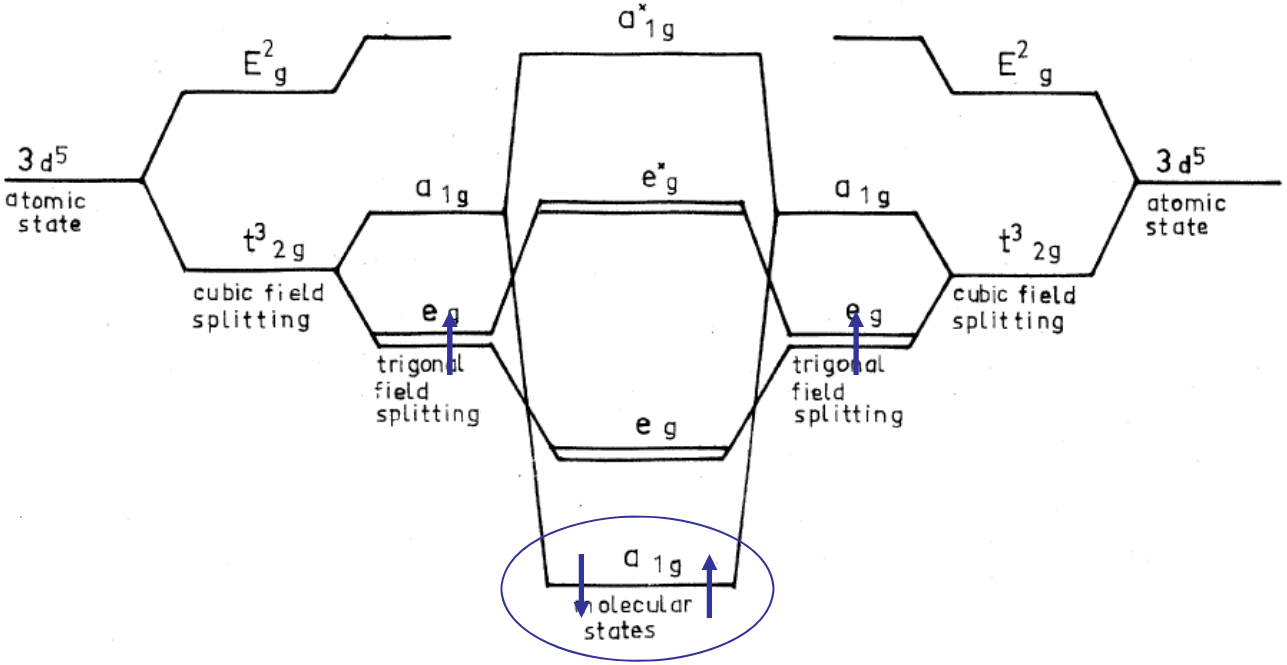
D. B. McWhan et al., PRB (1973)

Electron pairs ?

- V^{3+}-V^{3+} pairs: a_{1g} molecular singlet formation : effective S=1/2

Castellani et al., Phys. Rev. B, 18, 4945 (1978)

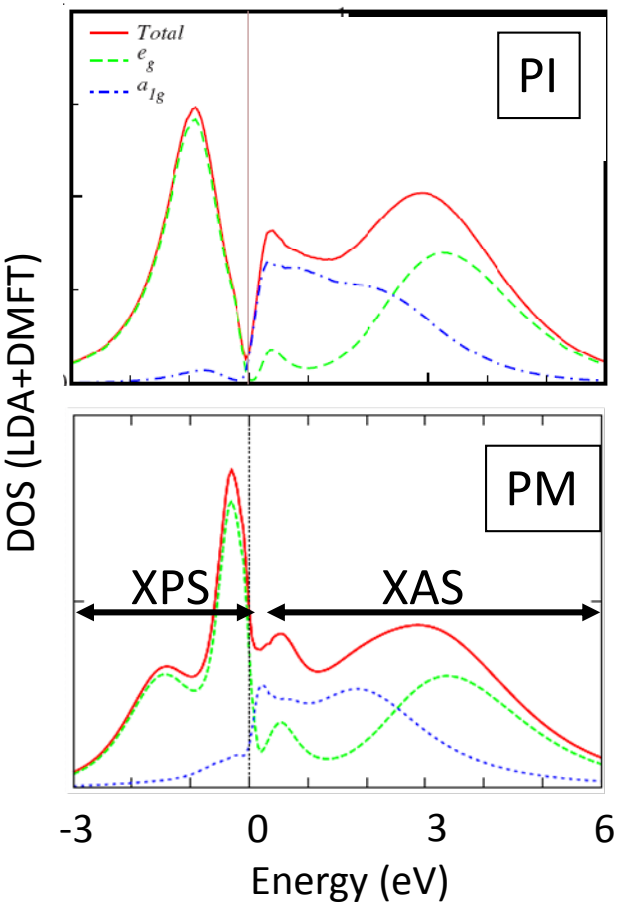




- $L_{2,3}$ XAS data : evidence for S=1, $(e_g^{1/4}; e_g^{1/4}); (e_g^{1/4}; a_{1g})$

Park et al., Phys. Rev. B, 61, 11506 (2000)

XPS + DMFT

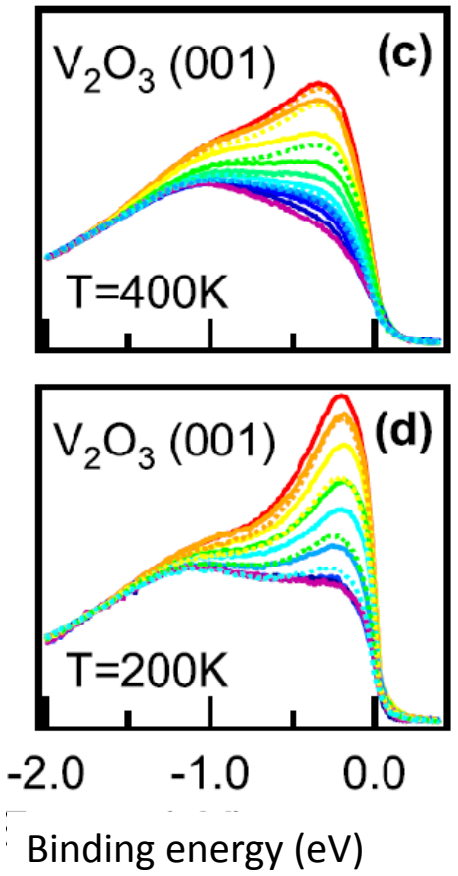


$(e_g^{\pi}; e_g^{\pi})$: $(e_g^{\pi}; a_{1g})$

65%:35%

50%:50%

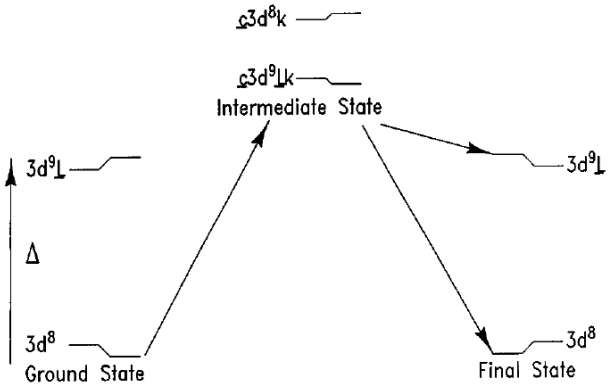
Valence band XPS



A.I. Poteryaev et al., Phys. Rev. B 76, 085127 (2007)

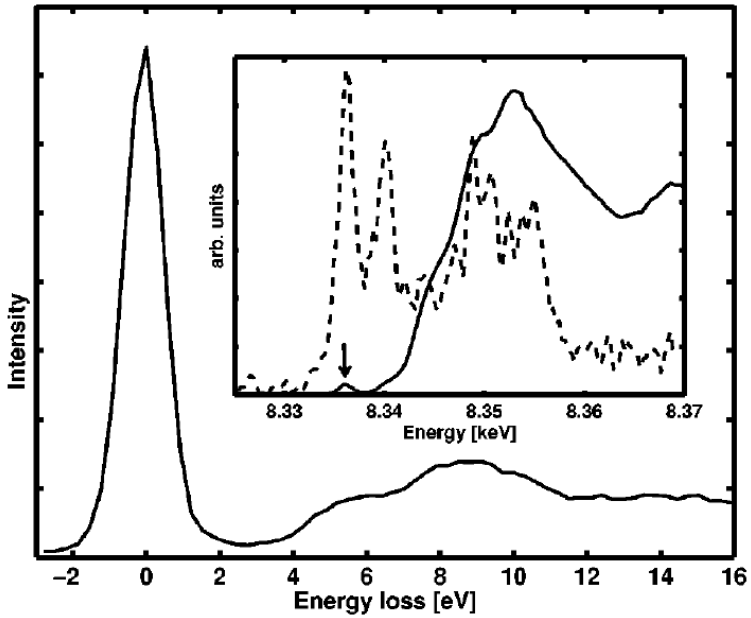
*F. Rodolakis, Phys. Rev. Lett., **102**, 066805 (2009)*

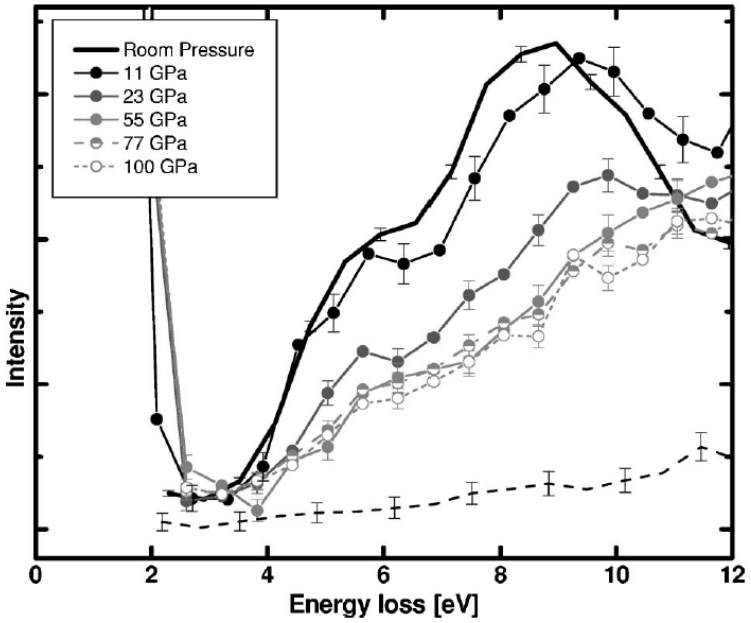
RIXS : Large gap insulator



C.C. Kao, Phys. Rev. B, ***54****, 16361 (1996)*

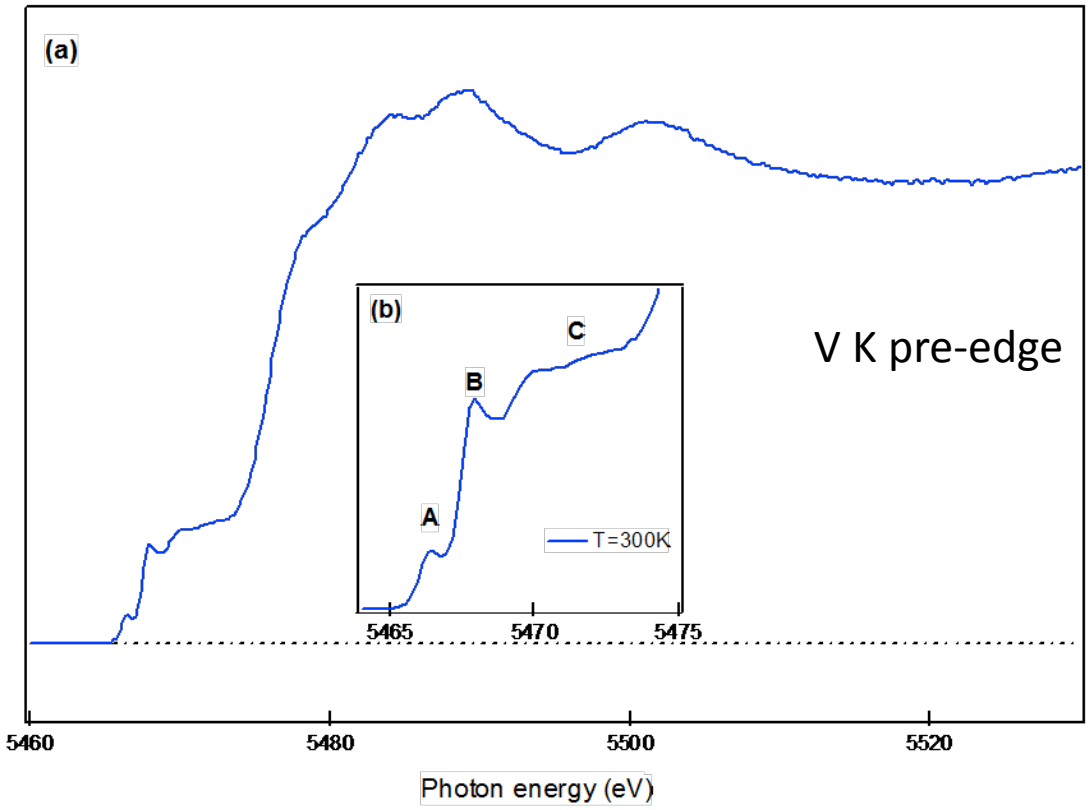
NiO under pressure



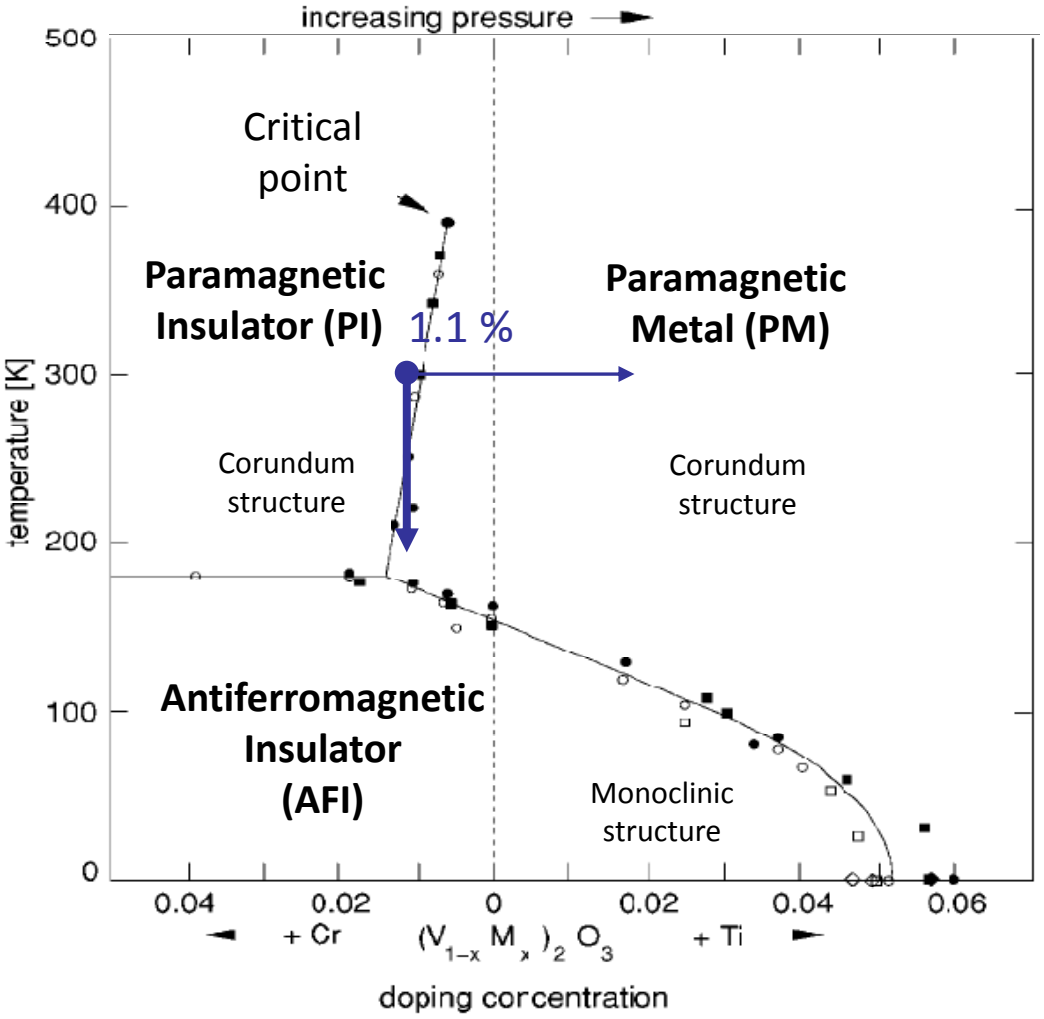


A. Shukla, Phys. Rev. B, **67**, 81101 (2003)

V K-edge in V_2O_3



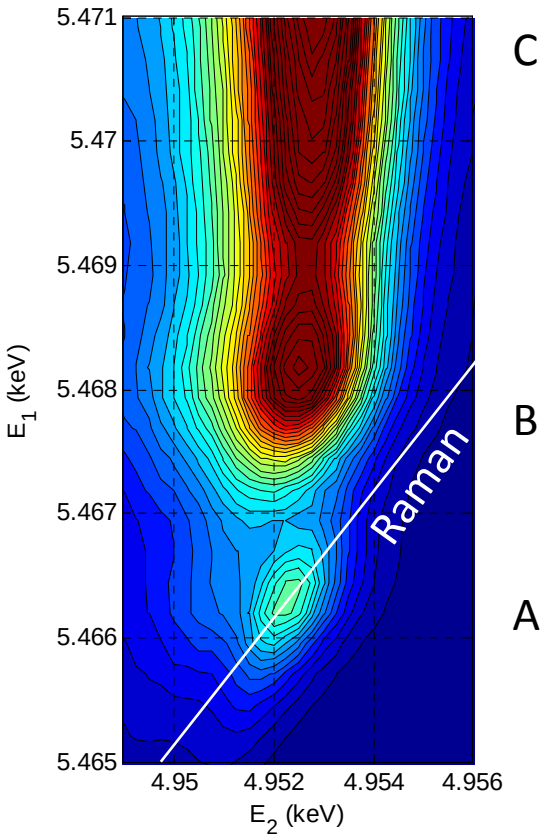
Metal Insulator transition in V_2O_3



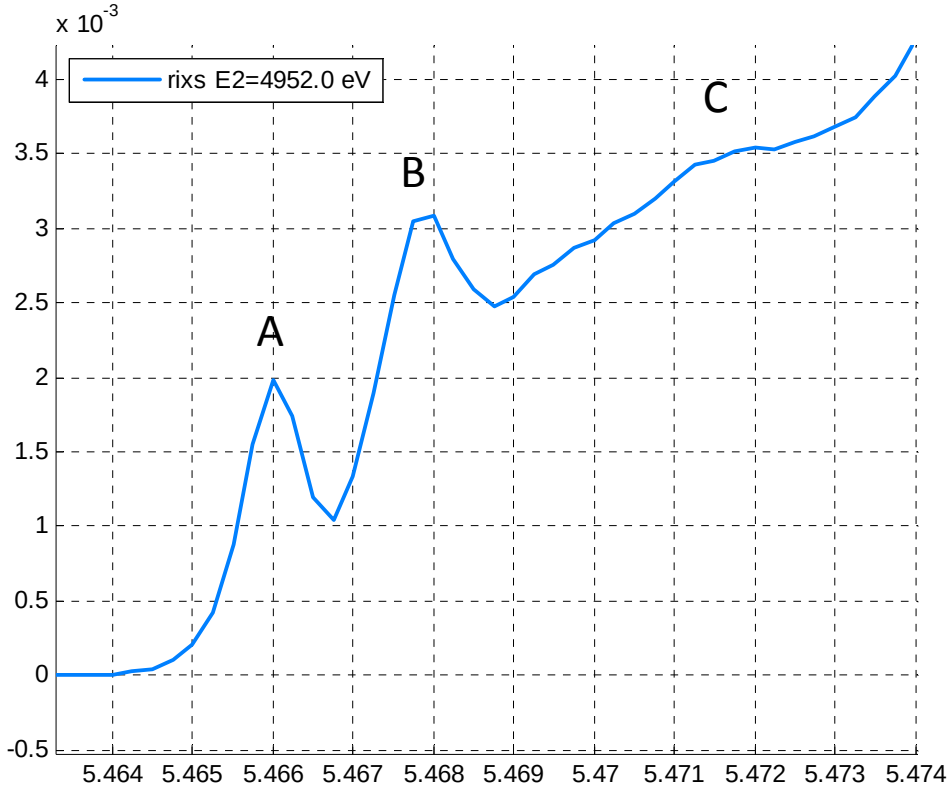
D. B. McWhan et al., PRB (1973)

PFY-XAS

1s2p-RXES

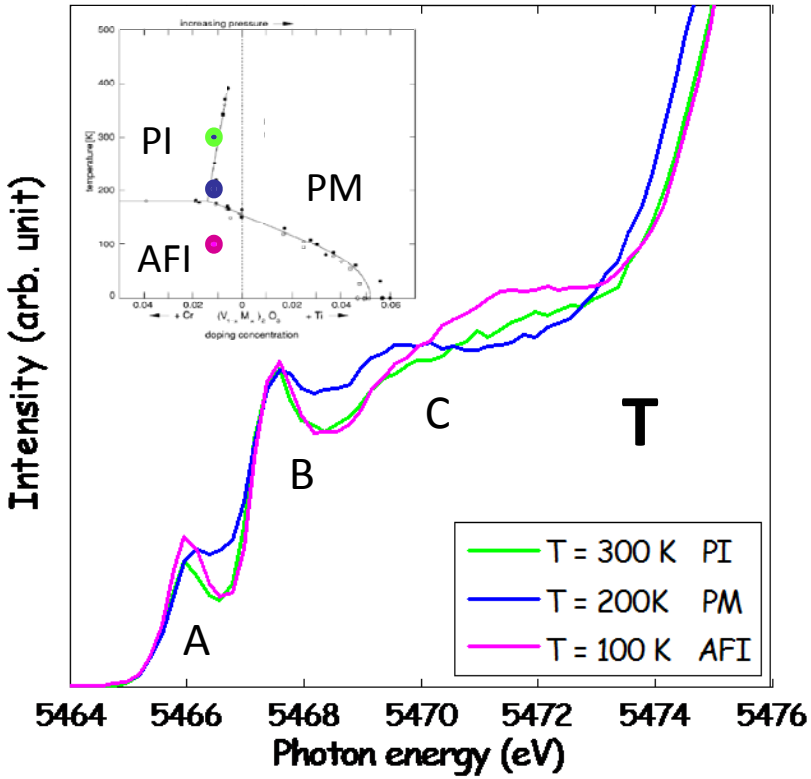


PFY-XAS V K-edge

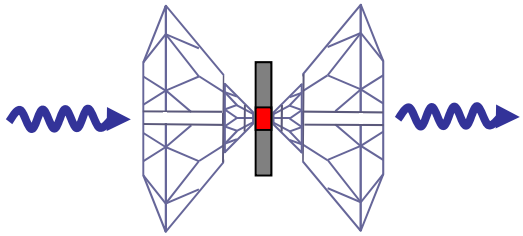


Temperature vs Pressure

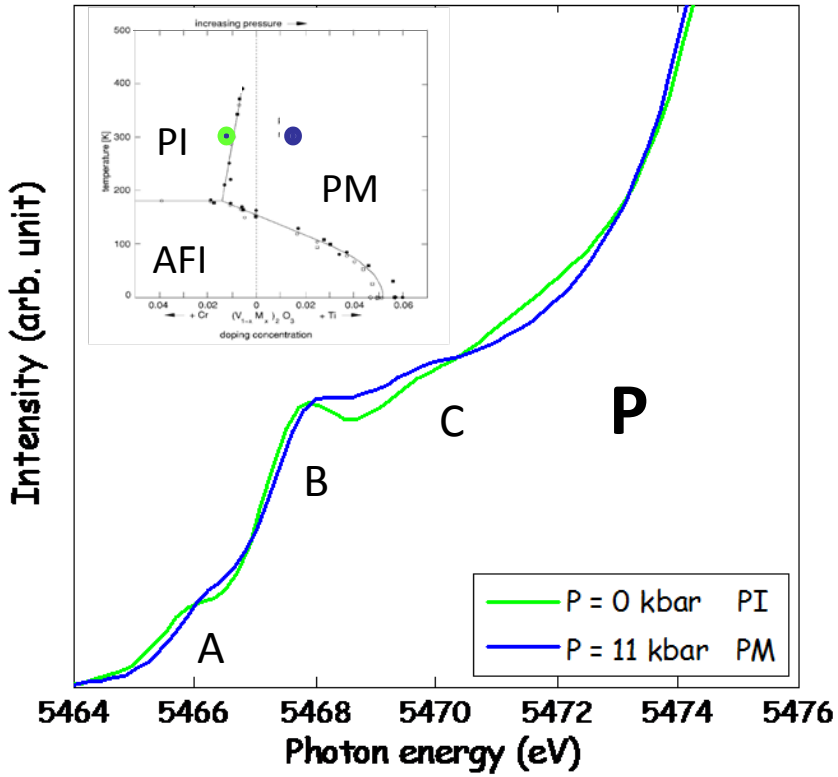
PFY-XAS



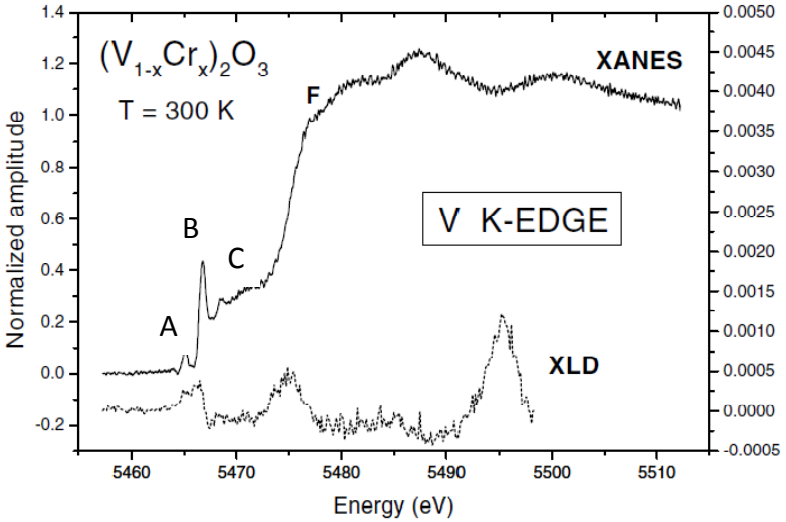
Perforated diamonds



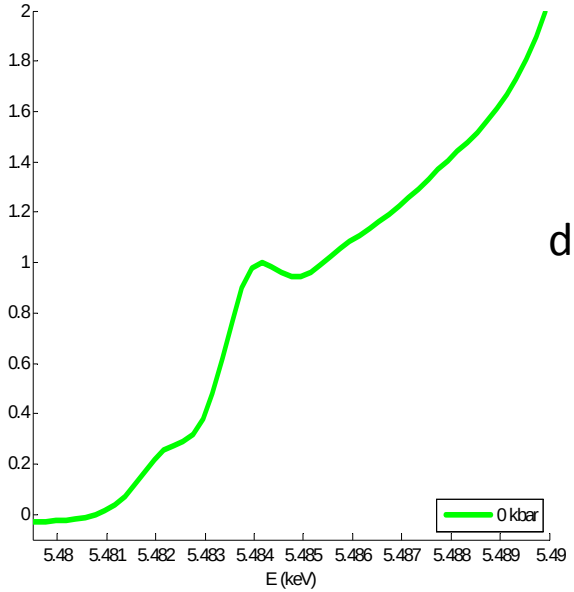
XAS - transmission



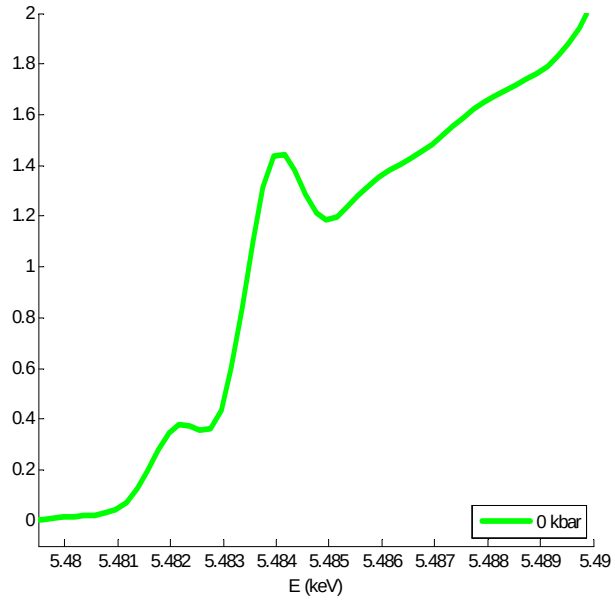
Deconvolution



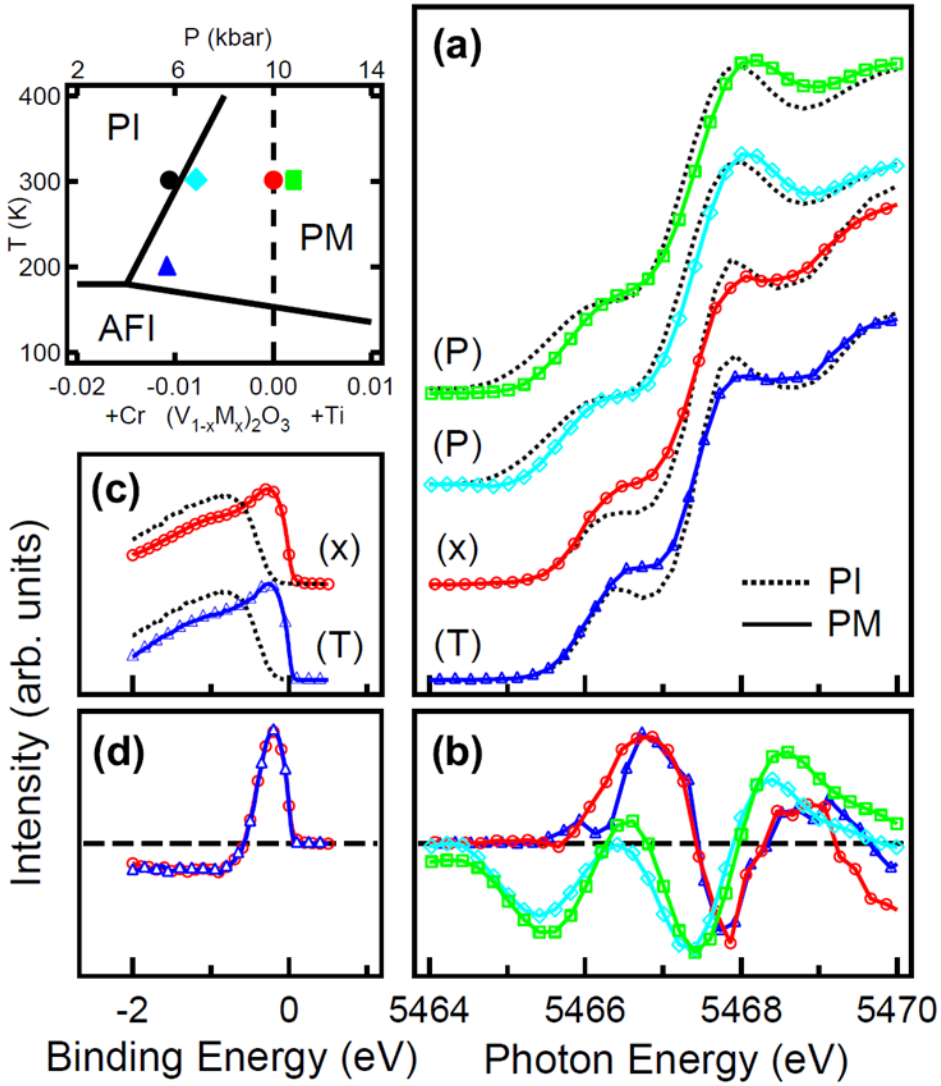
J. Goulon, Phys. Rev. Lett., **85**, 4385 (2000)



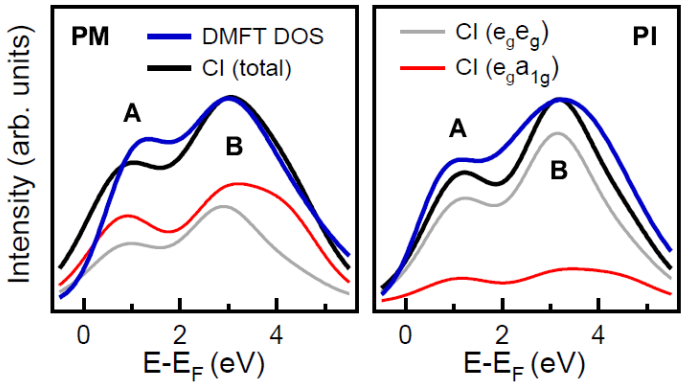
deconv (GNXAS)

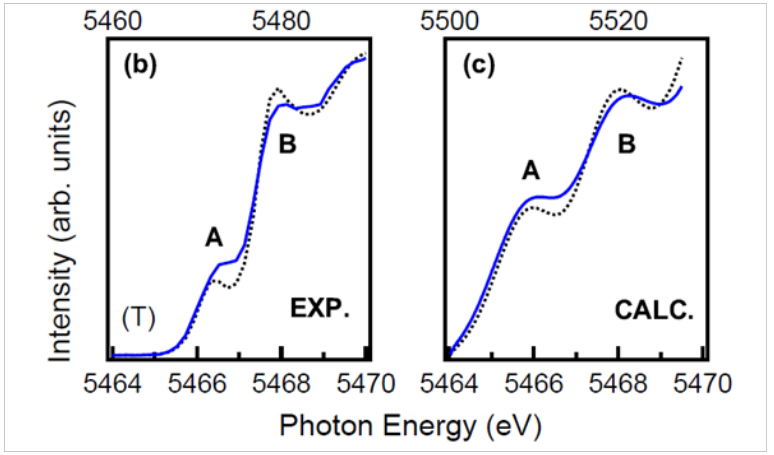


Multiplet + DMFT



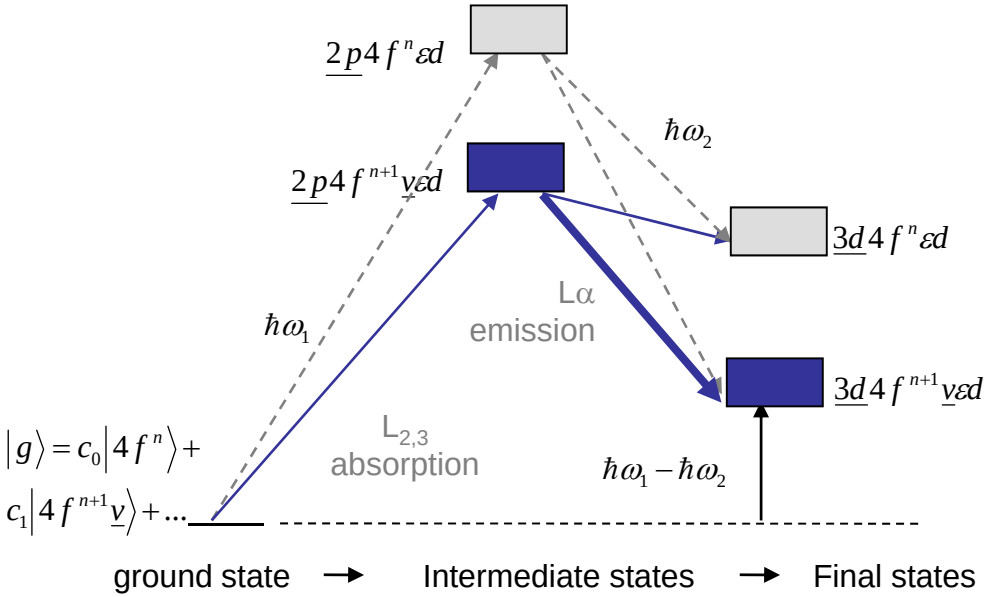
DMFT Incoherent part



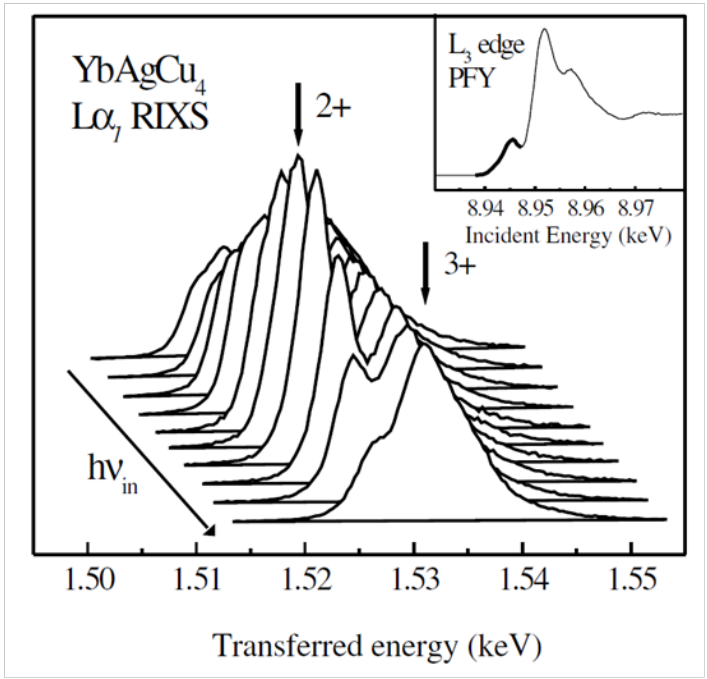


F. Rodolakis et al.

Mixed valent state: 2p3d-RXES



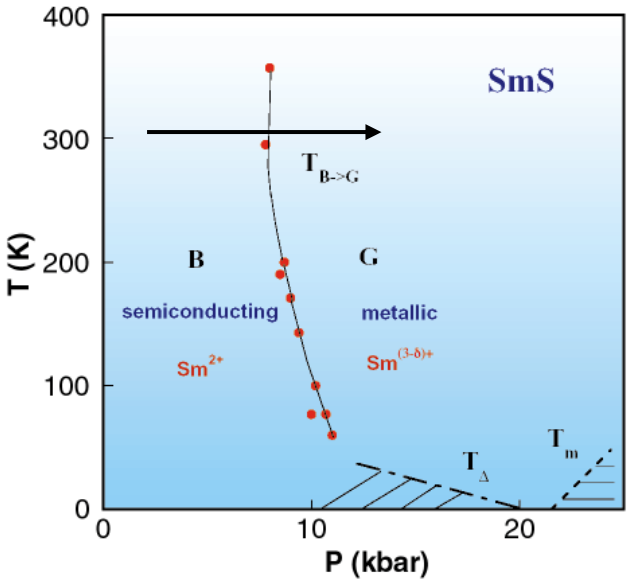
C. Dallera et al., Phys. Rev. Lett., **88**, 196403 (2002)

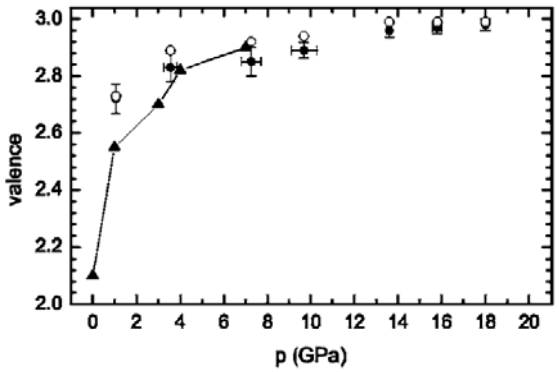


> Core hole potential separates the different mixed states
> Sharpening effect due to resonant effects
> Great accuracy in the determination of the valent state

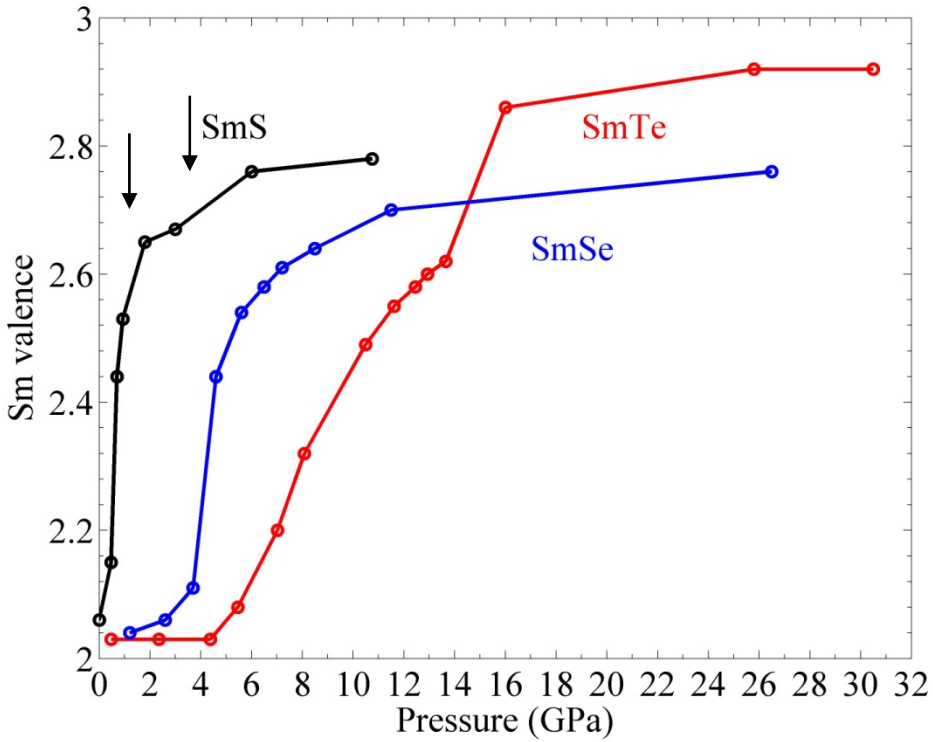
$$v = 2 + \frac{I_{RIXS}(3+) + I_{RIXS}(2+)}{I_{RIXS}(2+)}$$

SmS



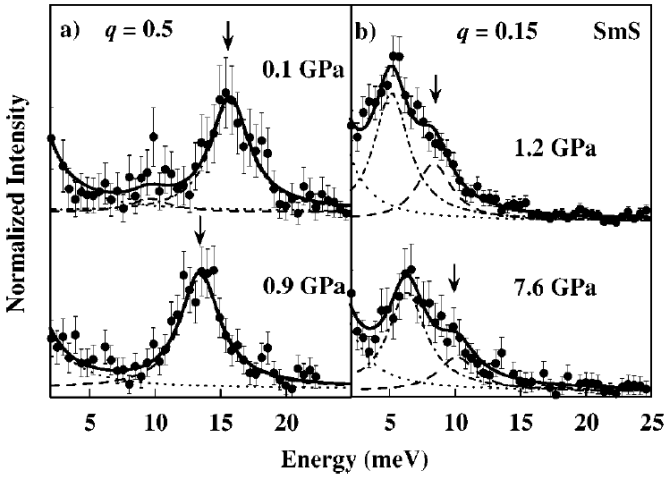


I. Jarrige et al. (2009)

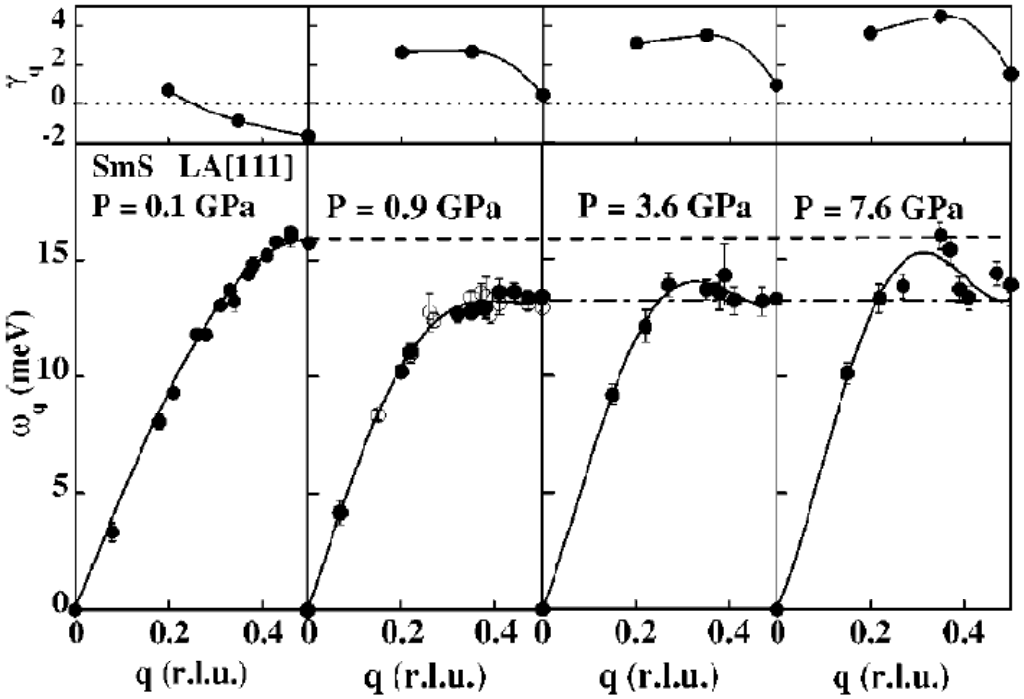


*Annese et al., PRB **70** 075117 (2004)*

SmS : Phonons under pressure

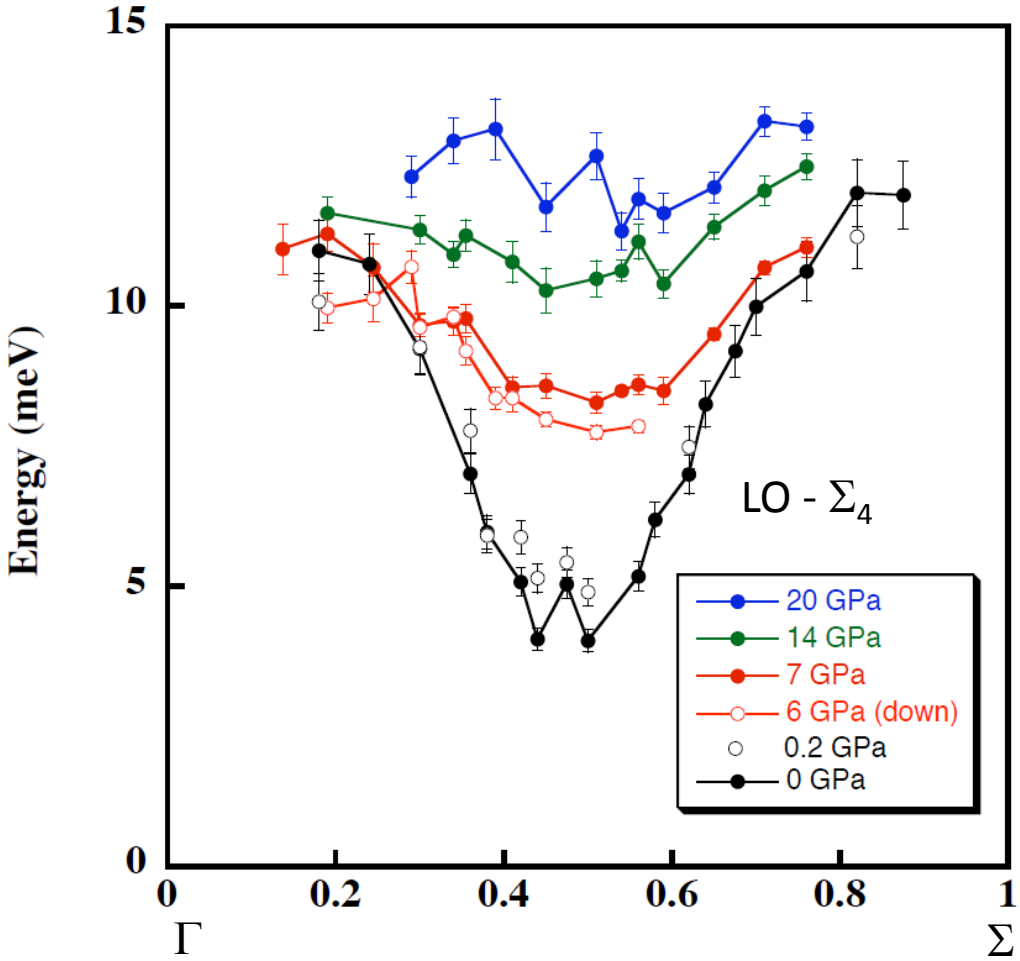


S. Raymond et al., Phys. Rev. B, **66**, 220301(R) (2002)



U phonon softening

Q=(h+1/2, 0, 1)



S. Raymond et al.

Conclusions

- RIXS is well adapted to high pressure studies
- Electronic, magnetic properties
- Phonons
- K-edge of light elements, …

J.-P. Rueff & A. Shukla, Inelastic X-ray Scattering by Electronic Excitations in Solids at High Pressure, arXiv:0812.0538 – submitted to Rev. Mod. Phys.

> GALAXIES beamline at SOLEIL : Hard x-ray IXS and Photoemission spectroscoy

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M. Haverkort, Ph. Hansman
K. Held

C. Hague, J.-M. Mariot

M. Grioni

C. Dallera, E. Annese, L. Braicovich