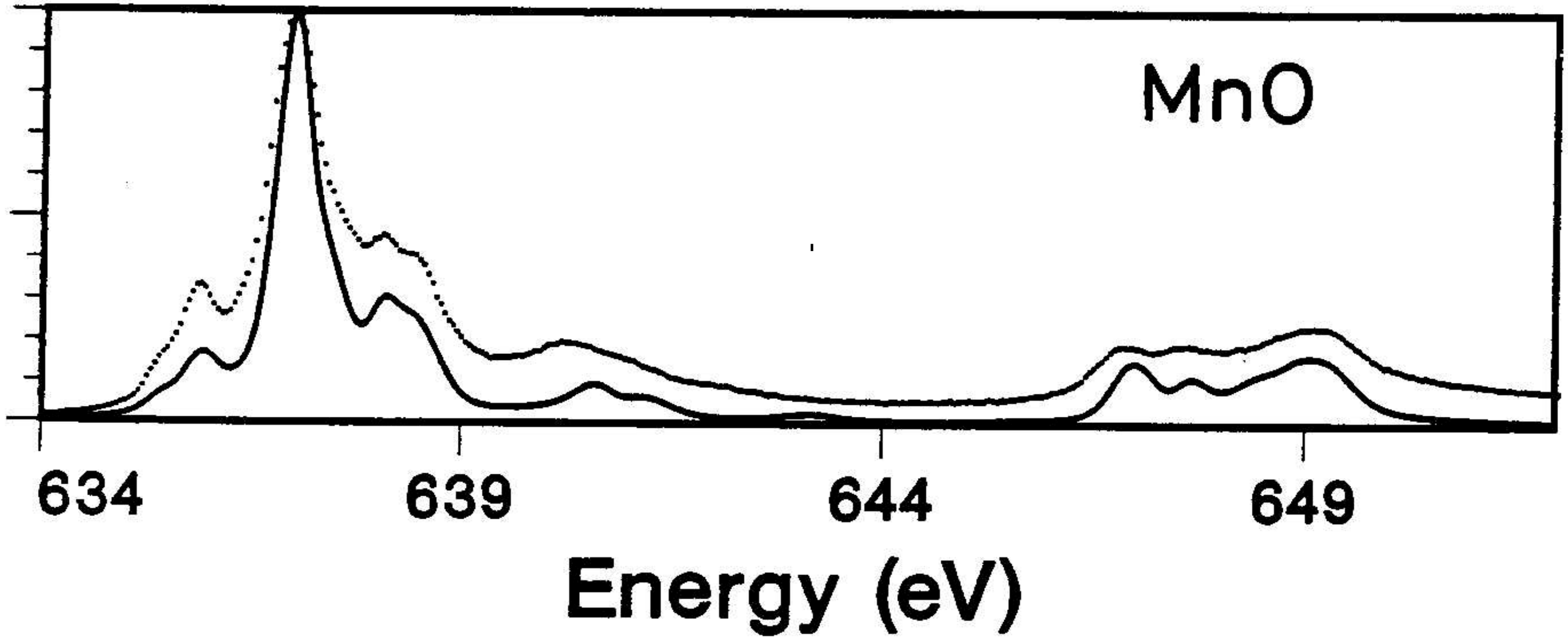
2p3d RIXS

- Some history on modeling RIXS

- Multiplet challenge
- Dispersions in open shell systems
- RIXS-MCD & RIXS Angular Distribution in Fe_3O_4

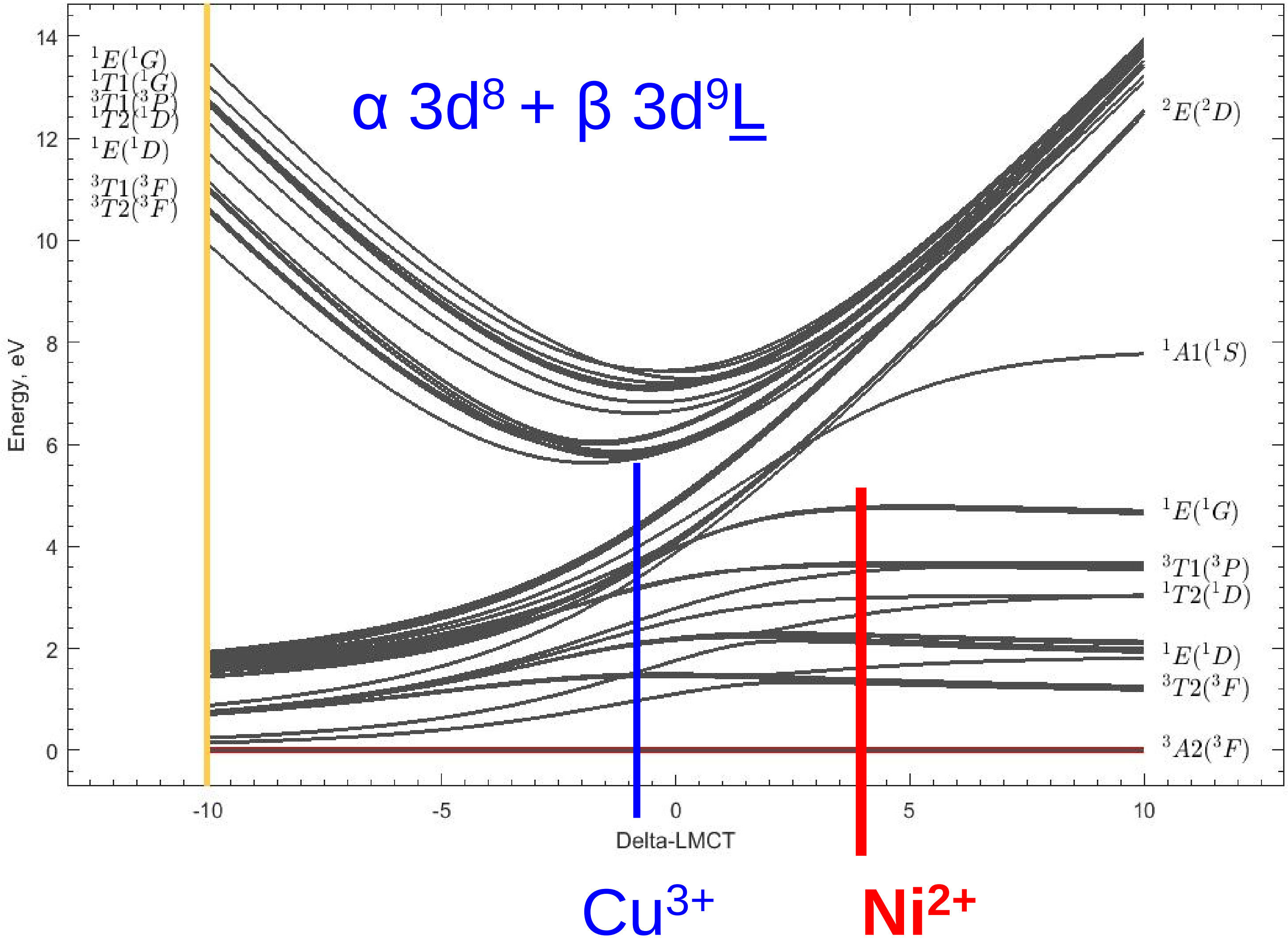
1990: 2p XAS of MnO



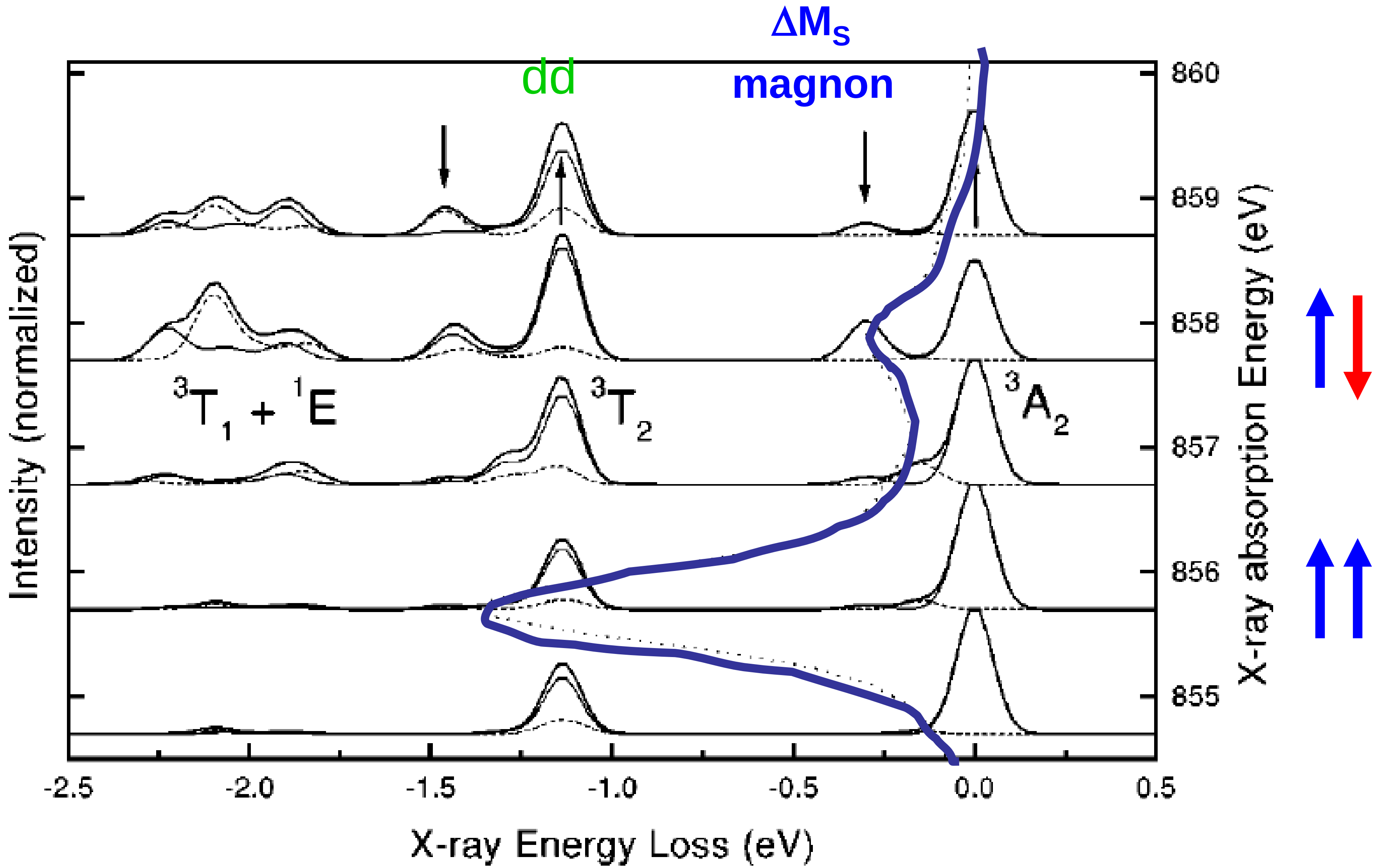
- Spectrum did NOT improve in last 30 years
- Neutral (self-screened) excitation
- Local $3d^5 > 2p^5 3d^6$ simulation is sufficient

[Phys. Rev. B. 42, 5459 (1990)]

(Local) low energy states of NiO



1998: 2p3d RIXS of NiO



- Self-screened excitation and decay

[Phys. Rev. B. 57, 14584 (1998)]

Multiplet calculations

Calculated for an atom/ion

- Valence and core **spin-orbit coupling**
- Core and valence **electron-electron interactions**

Comparison with experiment

- Core hole **potential and lifetime**
- Local and global symmetry (**crystal field)**
- Spin-spin interactions (**molecular field**)
- Core hole screening effects (**charge transfer**)

First Principle Multiplet calculations

<u>Calculated for an atom/ion</u>

- Valence and core **spin-orbit coupling**
- Core and valence **electron-electron interactions**

<u>Comparison with experiment</u>

- Core hole **potential and lifetime**
- Local and global symmetry (**crystal field)**
- Spin-spin interactions (**molecular field**)
- Core hole screening effects (**charge transfer**)

2p3d RIXS (2p XAS) first-principle codes

SOLIDS

- Band structure multiplet (Haverkort, Hariki/Kunes)
- Cluster DFT multiplet (Ikeno, Ramanantoanina, Delley)

MOLECULES

- Restricted Active Space CI (Odelius, Lundberg, Kuhn)
- Restricted Open-shell CI (Neese)

TDDFT/BSE

- *Time-Dependent DFT (Joly, Ambrosh-Draxl)*
- *Bethe-Salpeter (Rehr, Shirley, Prendergast)*
- *Multi-channel Multiple-scattering (Kruger)*

2p XAS & 2p3d RIXS

1989

XAS: 200 meV

RIXS:500 meV

→

2019

XAS: 200 meV

RIXS: 20 meV

200 meV

Valence, spin

Crystal-field

Charge transfer

20 meV

Spin-orbit

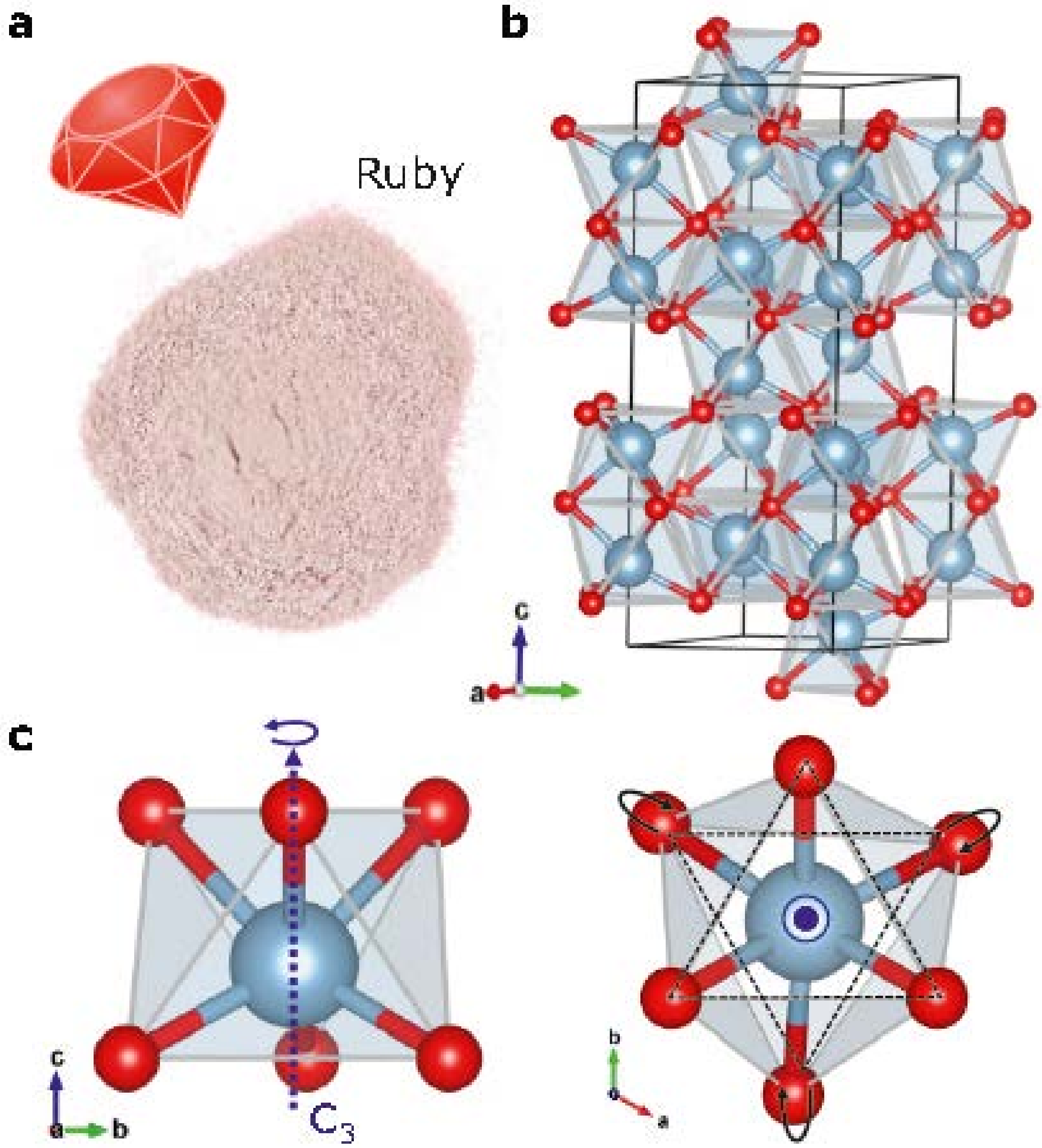
Magnons

phonons

polarization, direction

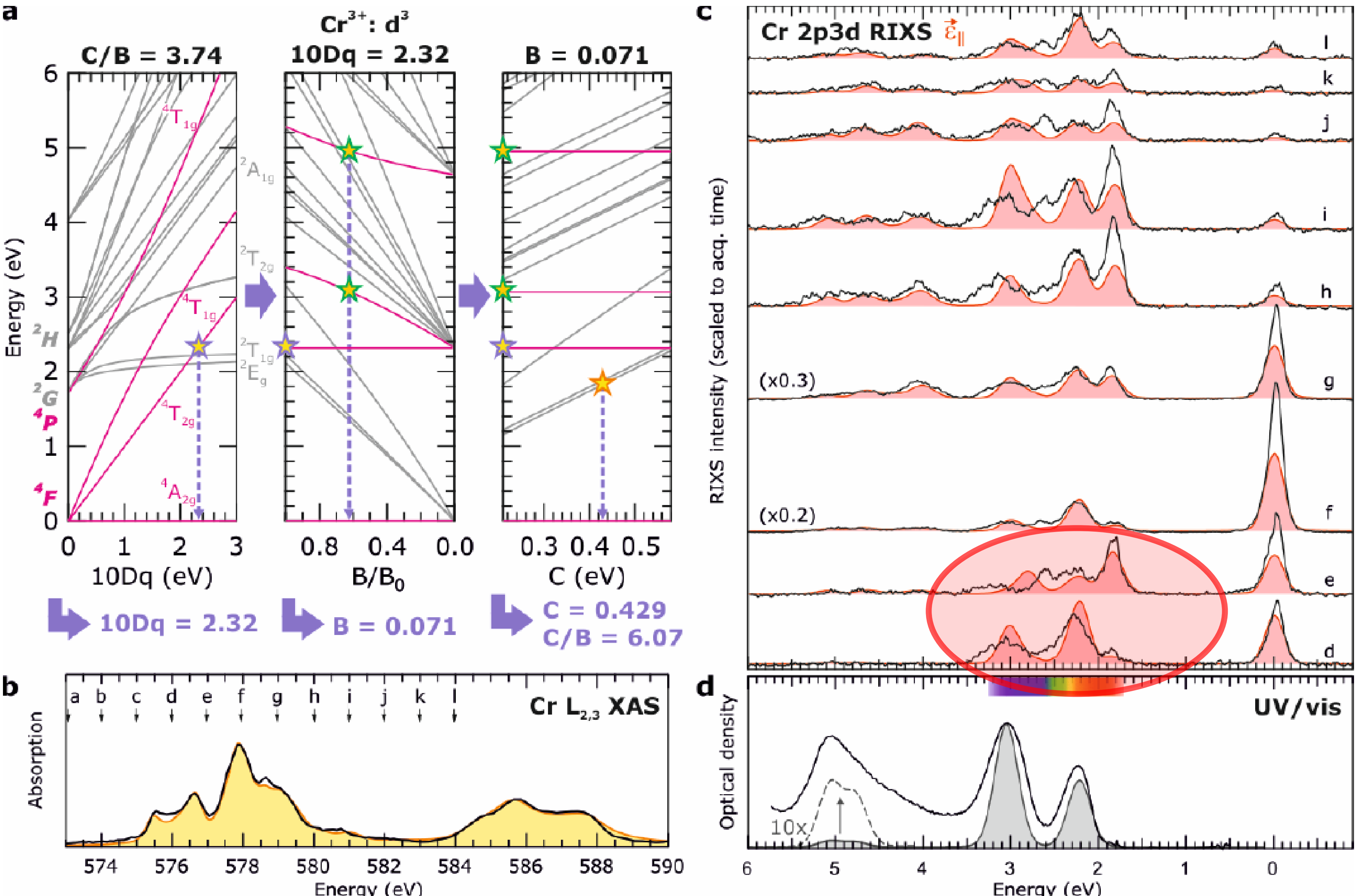
(in, sample, out)

2p3d RIXS of isolated chromium ions (ruby)



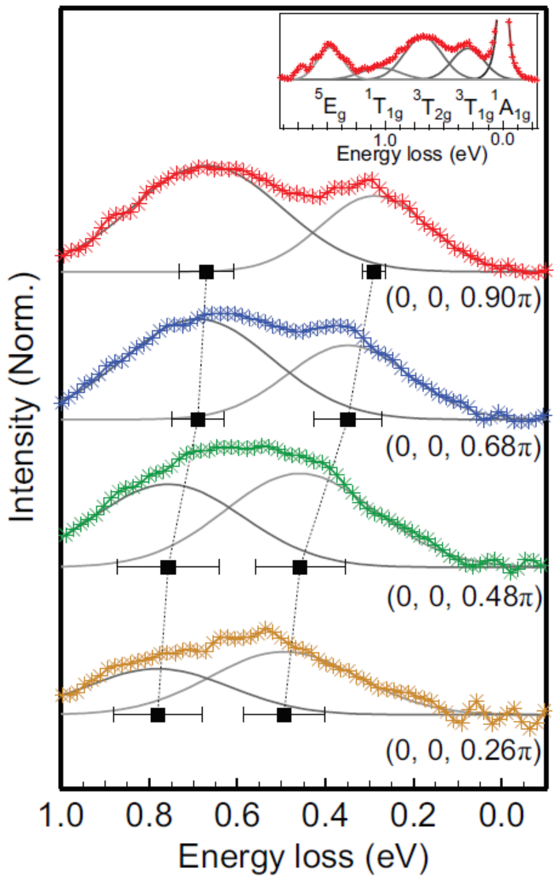
Cr^{3+}
in Al_2O_3

Multiplet challenge: 2p3d RIXS on Cr^{3+}

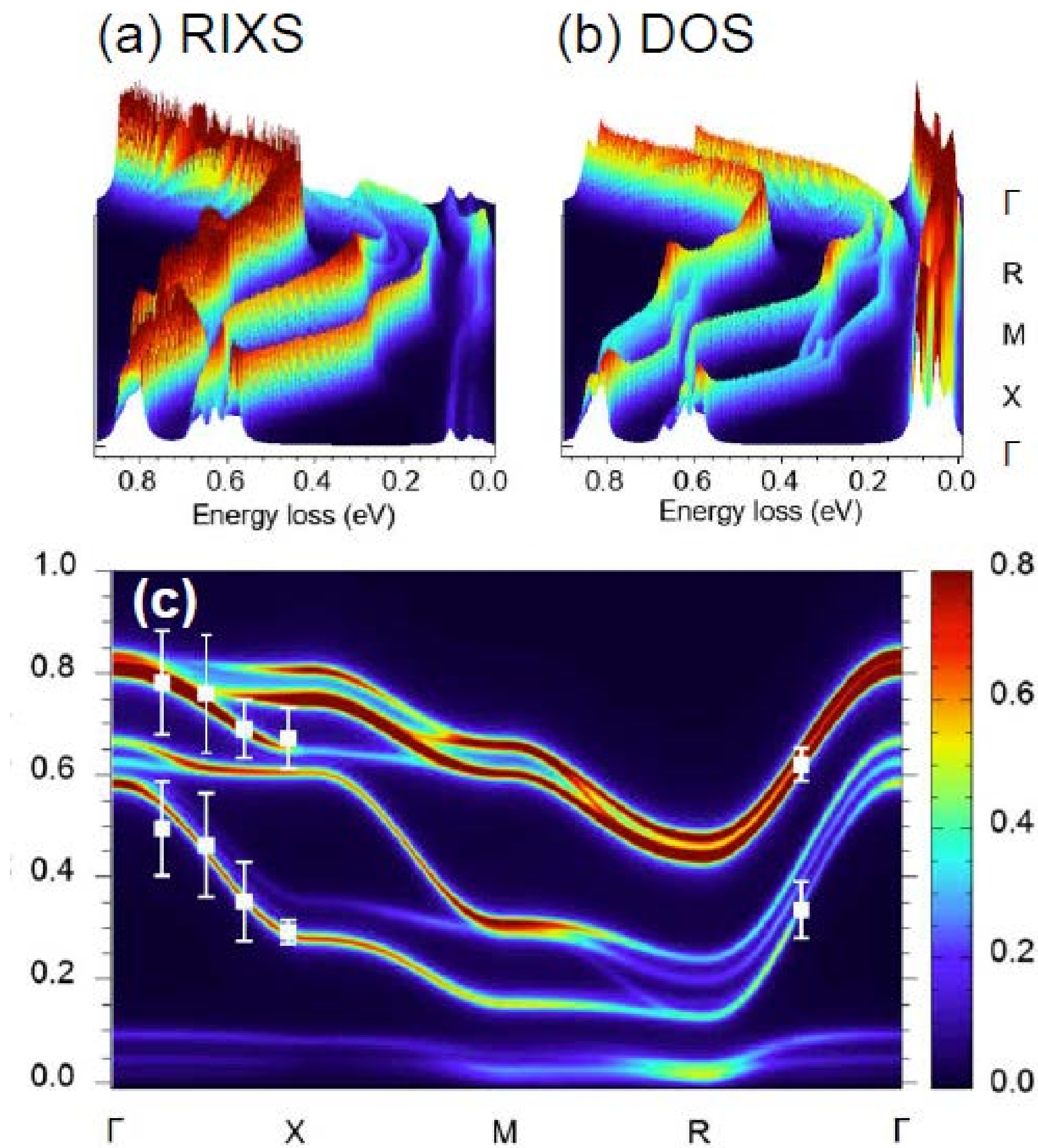


[J. Phys. Chem. A. 122, 4399 (2018)]

Dispersions in open shell systems



q-dep. RIXS



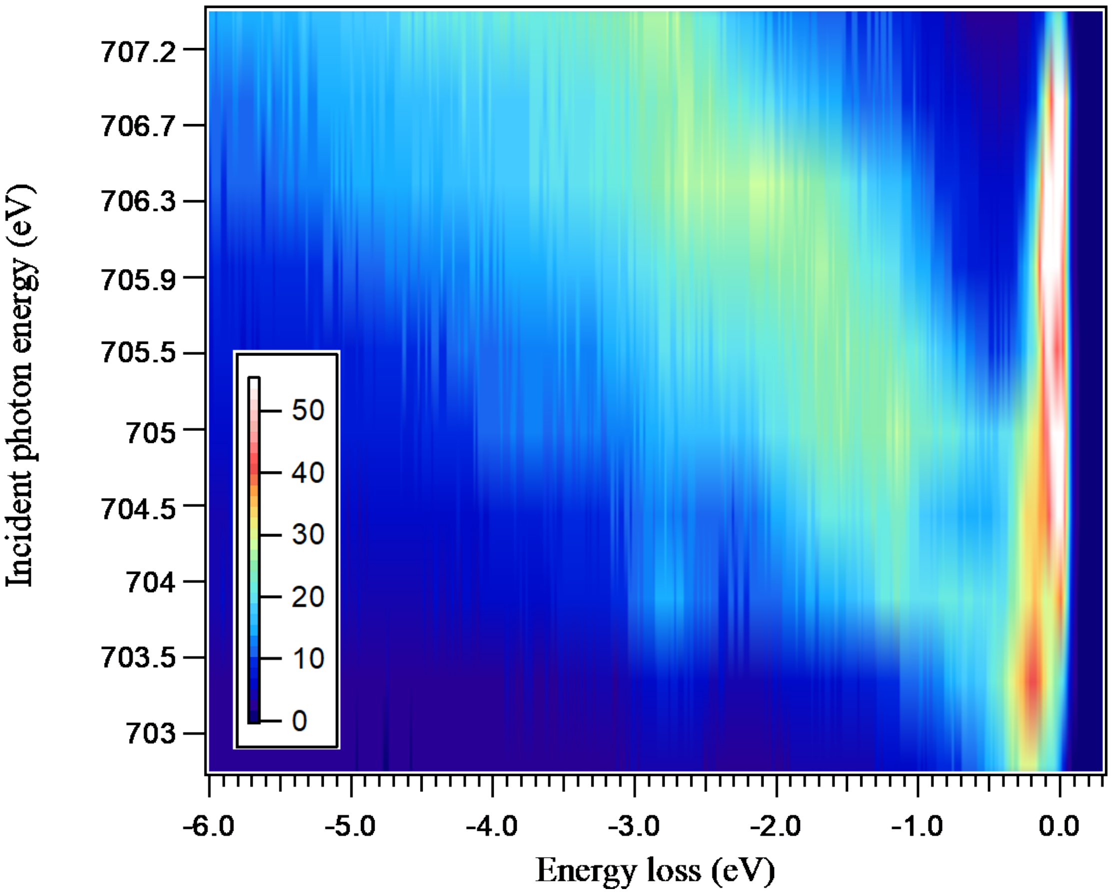
[Wang et al. Phys. Rev. B. 98, 035149 (2018)]

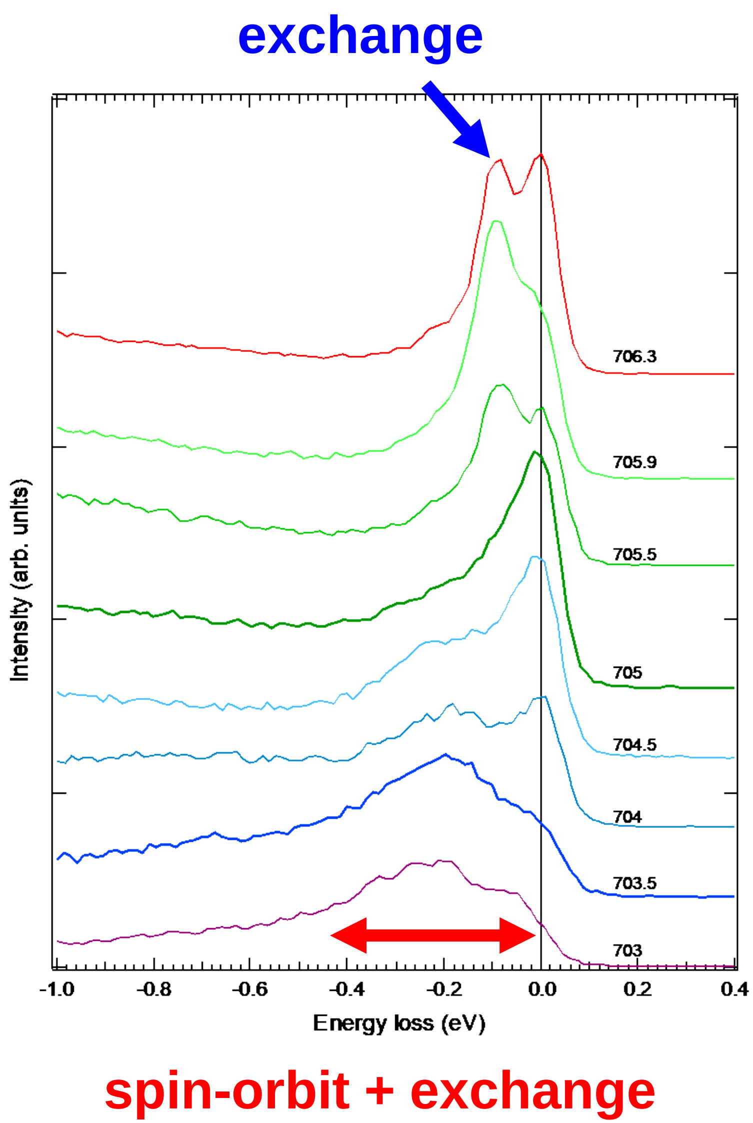
Dispersions in open shell systems

(first 400 meV)

- Cu^{2+} $3d^9$ ^2E — magnon, phonons
- Co^{3+} $3d^6$ $^1A_1$ — phonons, low-lying dd-excitations
- Fe^{2+} $3d^6$ $^5T_2$ — 4 (multi) magnons, phonons, 14 dd-excitations (spin-orbit, distortions)

2p3d RIXS of Fe_3O_4





[Huang et al. Nature Comm. 8, 15929 (2017)]

Utrecht University

Department of Chemistry
Debye Institute for Nanomaterials Sciences

Magnetic contrast at spin-Flip excitations:

An advanced X-ray spectroscopy tool to study magnetic-ordering

By: Hebatalla Elnaggar

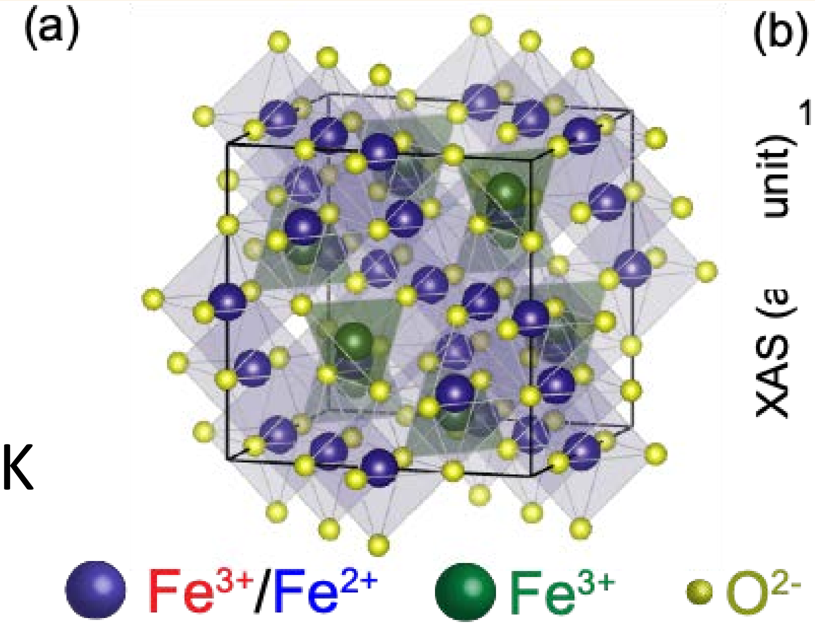
Utrecht University

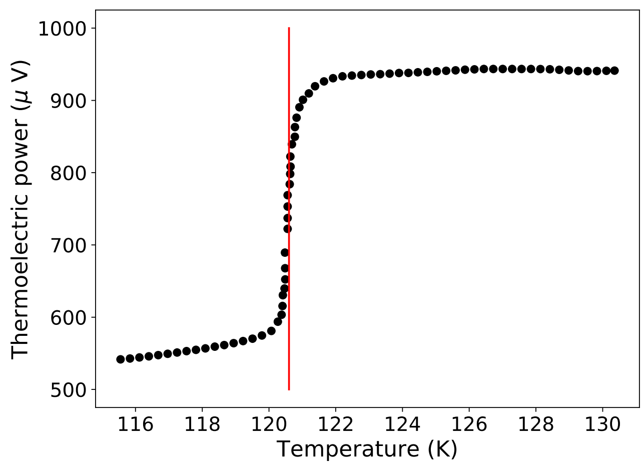
Magnetite (Fe_3O_4)

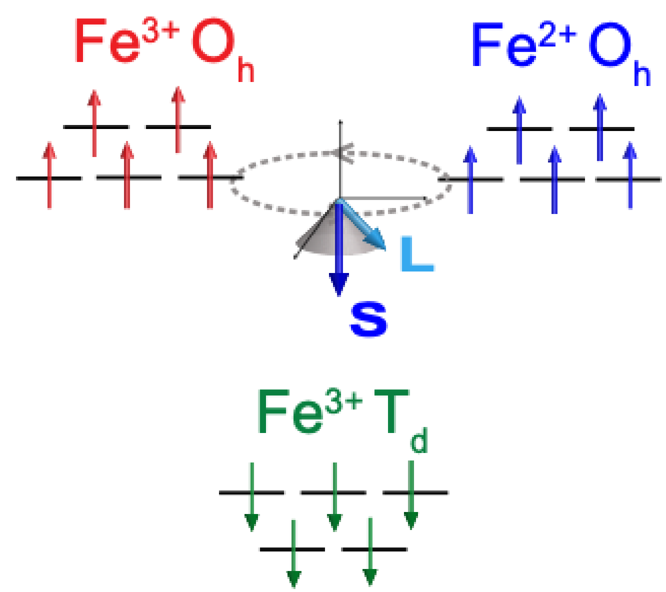
Fe_3O_4

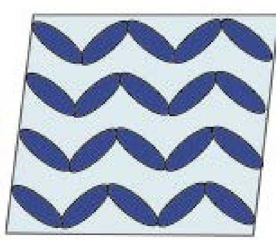
$Fe^{3+}{}_A[Fe^{3+},Fe^{2+}]_B O^{2-}{}_4$

Metal to insulator transition @ 120K

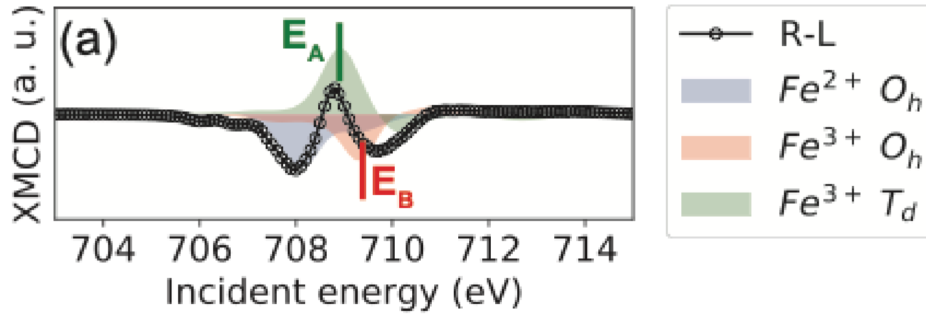




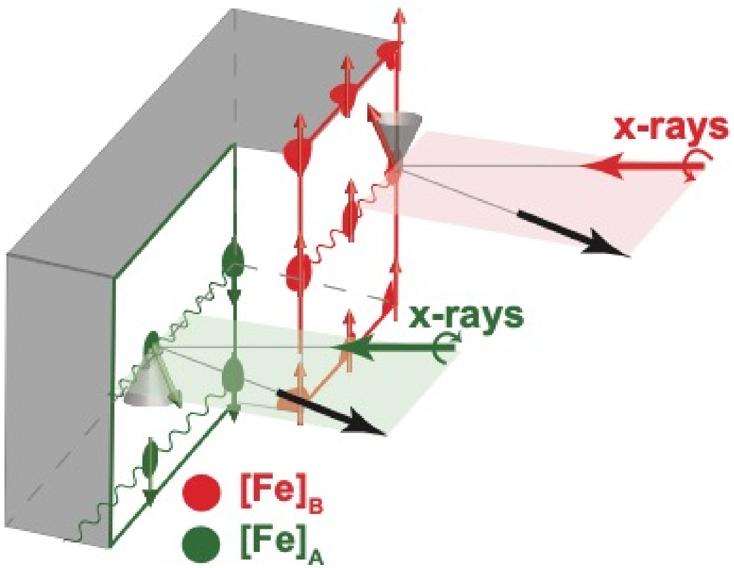


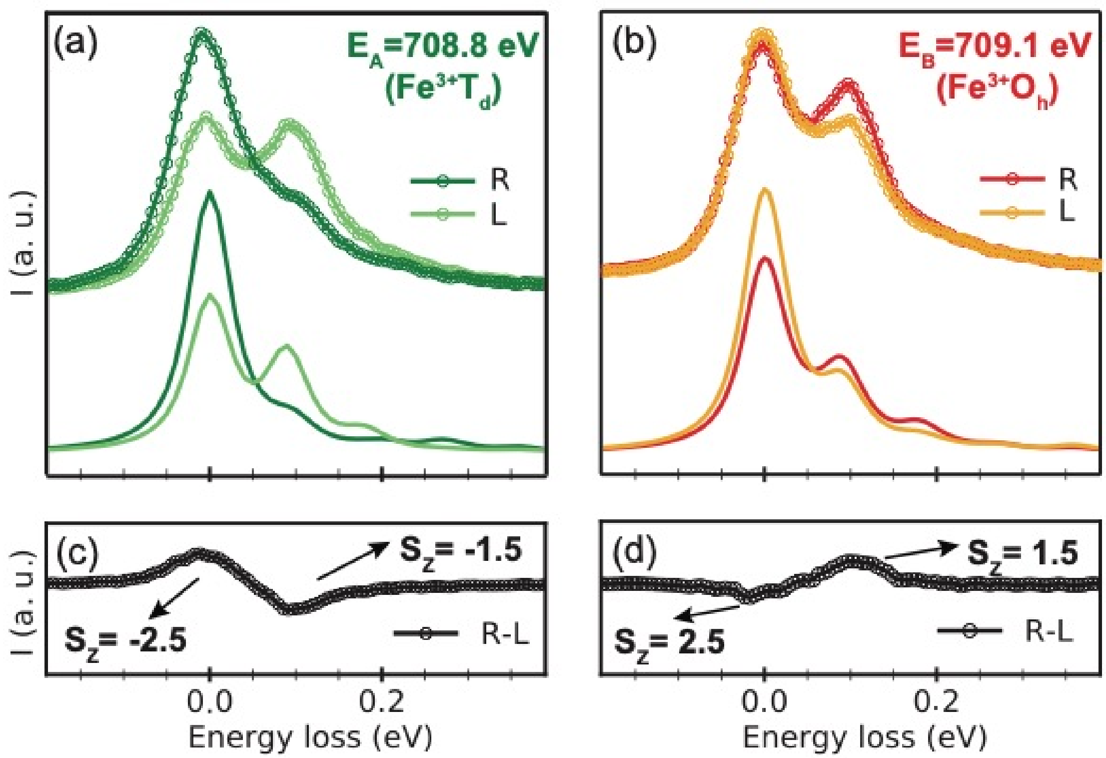


Disentangling the A and B sites



2p3d RIXS-MCD

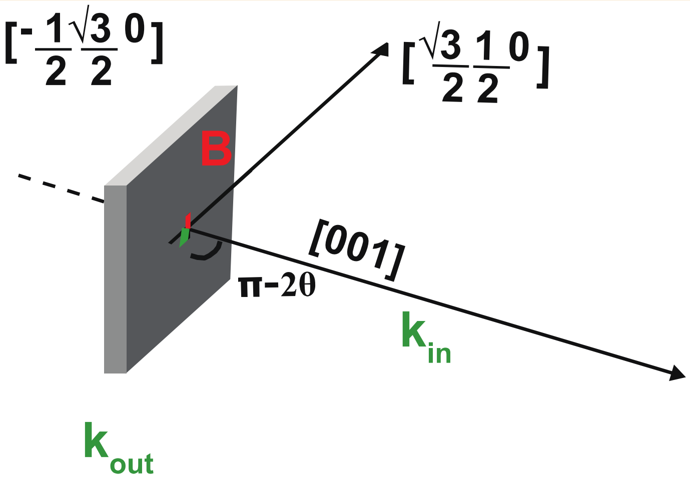


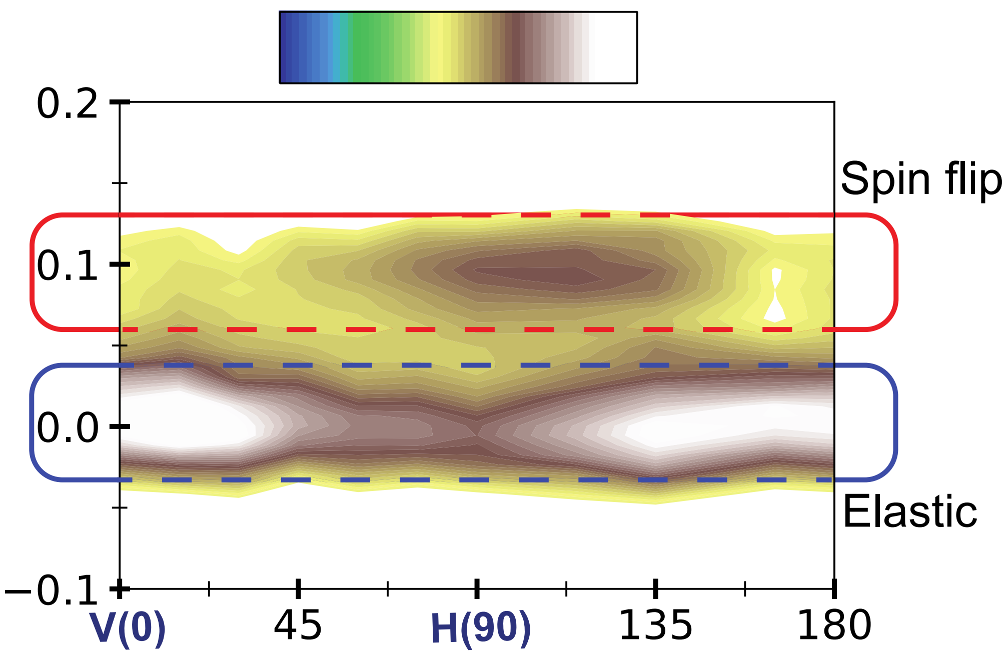


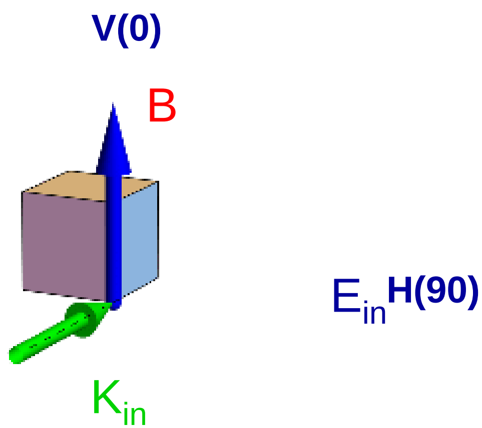
J_{eff} = 90 meV

RIXS magnetic angular distribution

$Fe^{3+}{}_A$



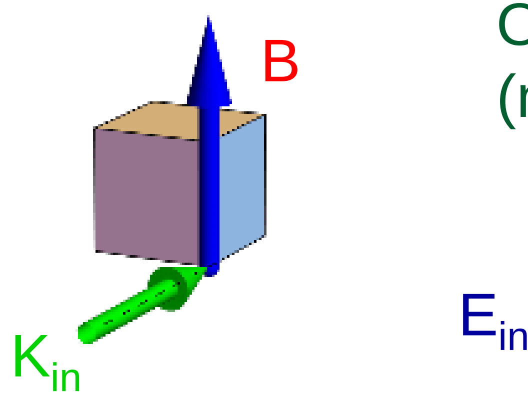


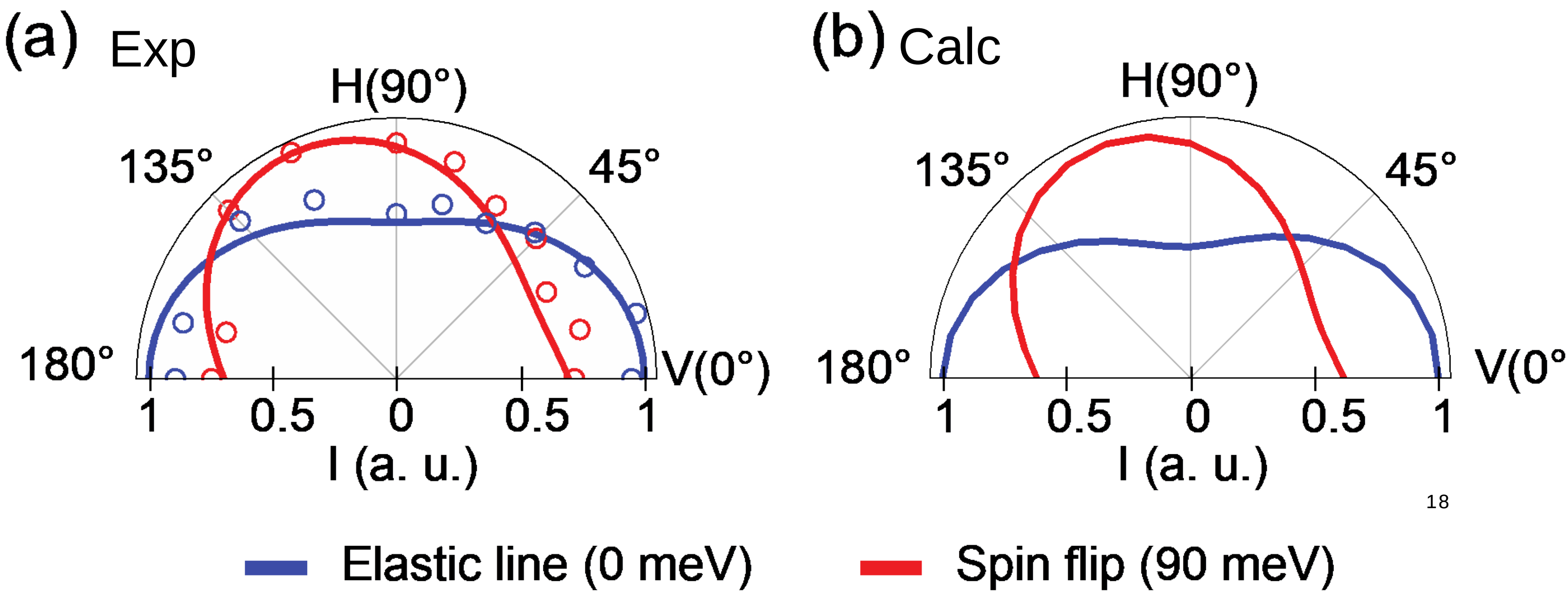


Quantifying the orbital moment

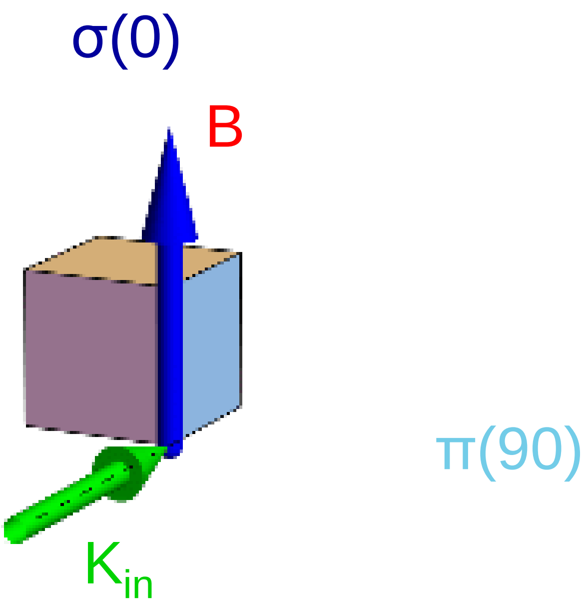
$Fe^{3+}{}_A$

Orbital moment is zero
(no induced moment)

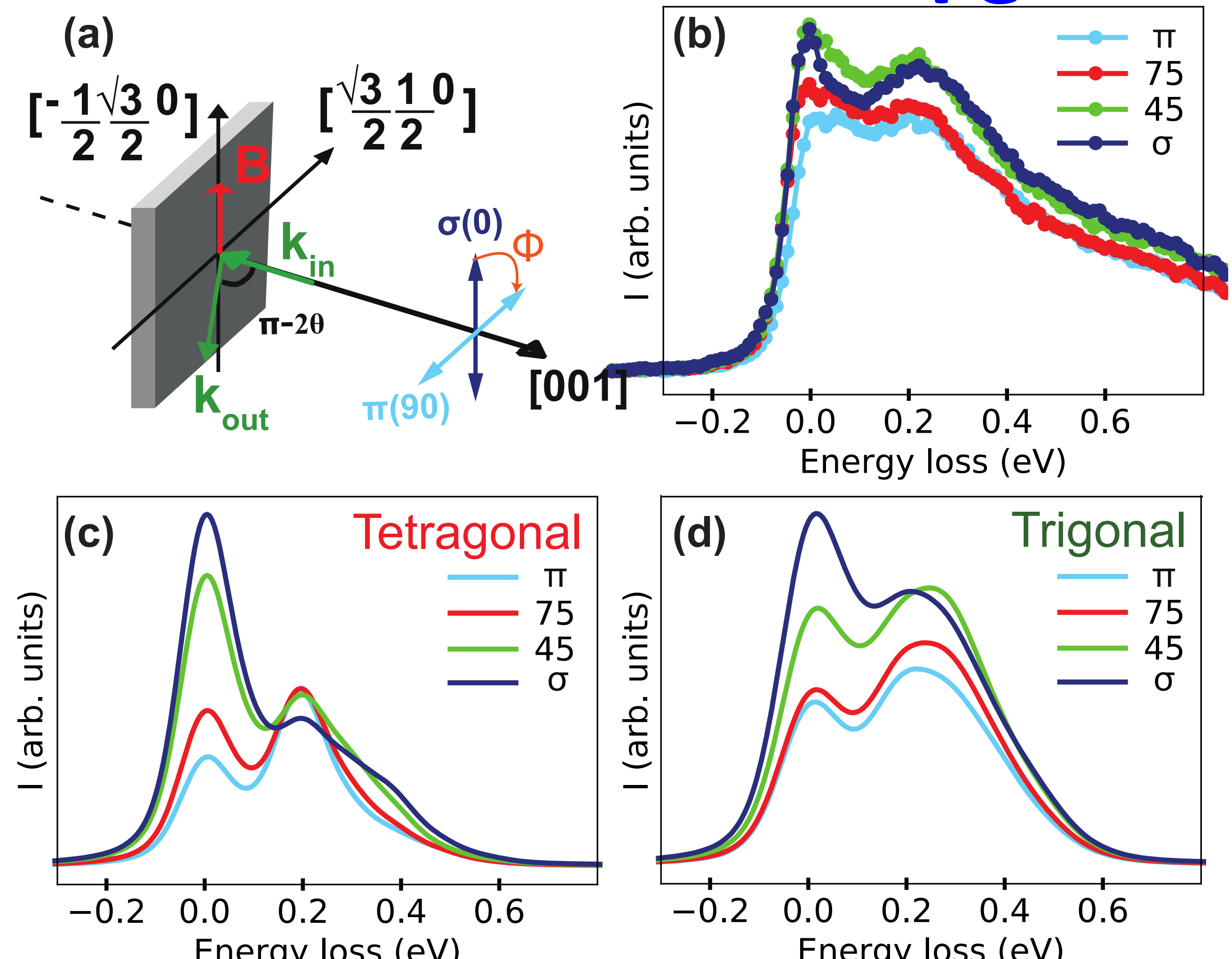




RIXS-MLD in optimized geometry



Fe^{2+}



Orbital moment is smaller than 0.25

Trigonal distortion:
σ = 67(±10) meV

(saturation and self-absorption improve agreement)

Acknowledgements

Utrecht University:

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