Some history on modeling RIXS

Multiplet challenge

Dispersions in open shell systems

RIXS-MCD & RIXS Angular Distribution in Fe$_3$O$_4$
1990: 2p XAS of MnO

- Spectrum did NOT improve in last 30 years
- Neutral (self-screened) excitation
- Local $3d^5 > 2p^53d^6$ simulation is sufficient

[Phys. Rev. B. 42, 5459 (1990)]
(Local) low energy states of NiO

\[ \alpha 3d^8 + \beta 3d^9L \]

Cu\(^{3+}\)  Ni\(^{2+}\)
1998: 2p3d RIXS of NiO

• Self-screened excitation and decay

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

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)
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$_2$O$_3$
Multiplet challenge: 2p3d RIXS on Cr$^{3+}$

**a)**
- C/B = 3.74
- 10Dq = 2.32
- B = 0.071

**c)**
- Cr 2p3d RIXS
- RIXS intensity (scaled to acq. time)

**b)**
- Cr L$_{2,3}$ XAS

**d)**
- Optical density

[J. Phys. Chem. A. 122, 4399 (2018)]
Dispersions in open shell systems

q-dep. RIXS

Dispersions in open shell systems

(first 400 meV)

- Cu$^{2+}$ 3d$^9$ $^2$E magnon, phonons
- Co$^{3+}$ 3d$^6$ $^1$A$_1$ phonons low-lying dd-excitation
- Fe$^{2+}$ 3d$^6$ $^5$T$_2$ 4 (multi) magnons phonons 14 dd-excitation (spin-orbit, distortions)
2p3d RIXS of Fe$_3$O$_4$

[Image: A graph showing energy loss versus incident photon energy with various peaks and labels for spin-orbit + exchange.]

[Huang et al. Nature Comm. 8, 15929 (2017)]
Magnetic contrast at spin-Flip excitations: An advanced X-ray spectroscopy tool to study magnetic-ordering

By: Hebatalla Elnaggar
Magnetite (Fe$_3$O$_4$)

$\text{Fe}_3\text{O}_4$

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

Metal to insulator transition @ 120K
Disentangling the A and B sites

2p3d RIXS-MCD

\[ J_{\text{eff}} = 90 \text{ meV} \]
RIXS magnetic angular distribution

Fe$^{3+}$

Spin flip
Elastic

$V(0)$
$H(90)$
Quantifying the orbital moment

Orbital moment is zero (no induced moment)

Fe$^{3+}$

(a) Exp

(b) Calc

Elastic line (0 meV)  Spin flip (90 meV)
Orbital moment is smaller than 0.25

Trigonal distortion: $\sigma = 67(\pm 10)$ meV

(saturation and self-absorption improve agreement)
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