The Electronic Ground State of Sr_2IrO_4: a Core Level Resonant Inelastic X-ray Scattering Study

Stefano Agrestini

MAX-PLANCK-GESELLSCHAFT

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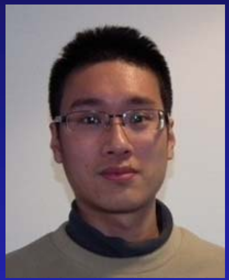
Collaborators

MPI for Chemical Physics of Solids (Dresden)



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MPI for Solid State Reasearch, Stuttgart

Hidenori Takagi

Tomohiro Takayama

The J_{eff}=1/2 Mott ground state

•Large Crystal Field (10*Dq*):

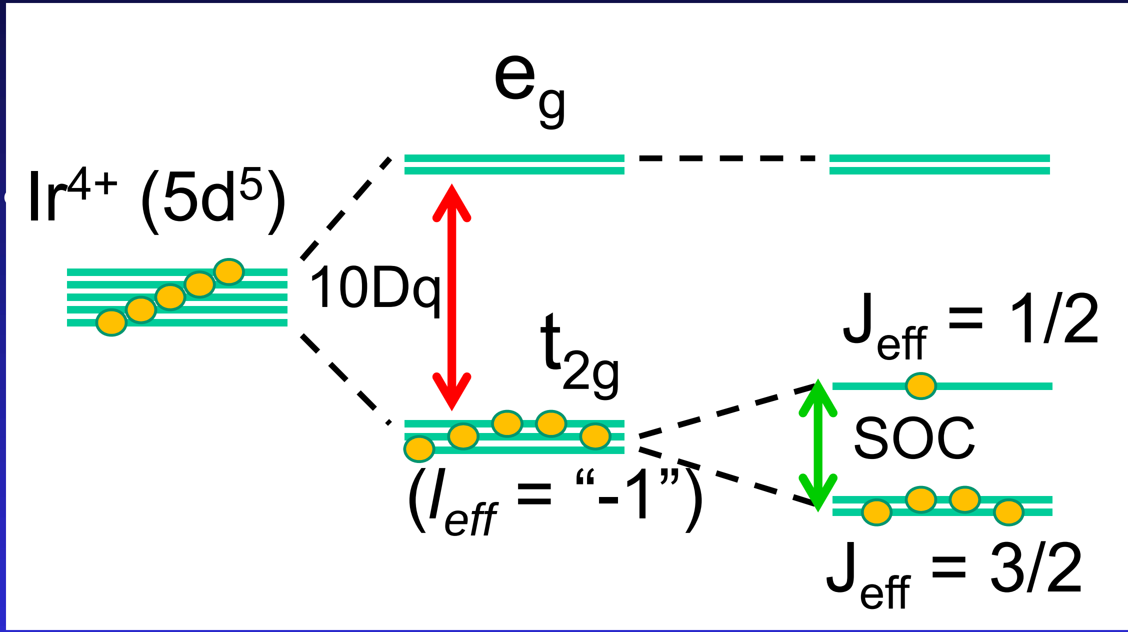
→ t_{2g}^5 configuration with l = 1

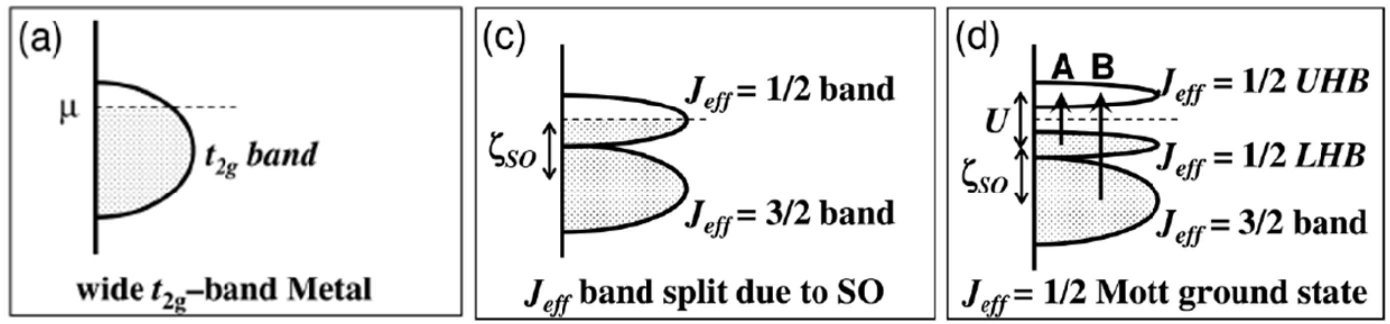
•Strong SOC:

→ t_{2g} splits into J_{eff} = 3/2 and 1/2

•Narrow J_{eff} = 1/2 band :

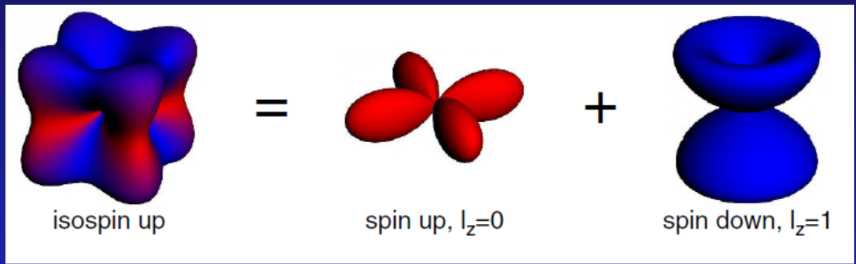
→ a moderate *U* can induce a Mott gap splitting





B.J. Kim et al, PRL (2008)

The J_{eff}=1/2 Mott ground state



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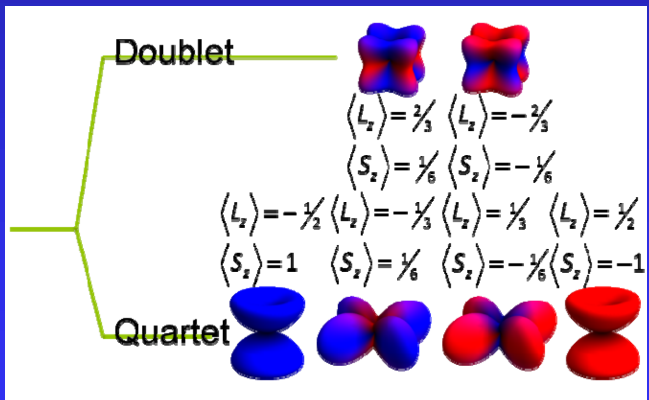

Ionic model in cubic local symmetry
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xy, xz, yz)

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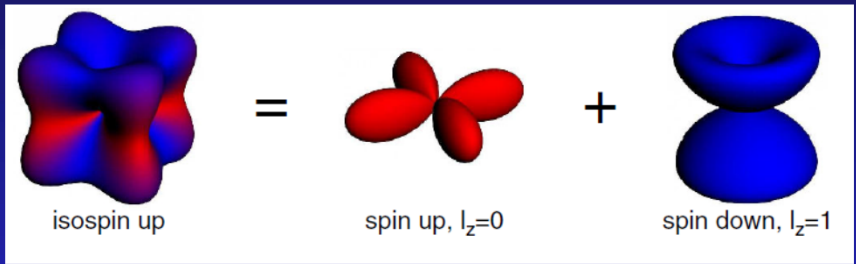

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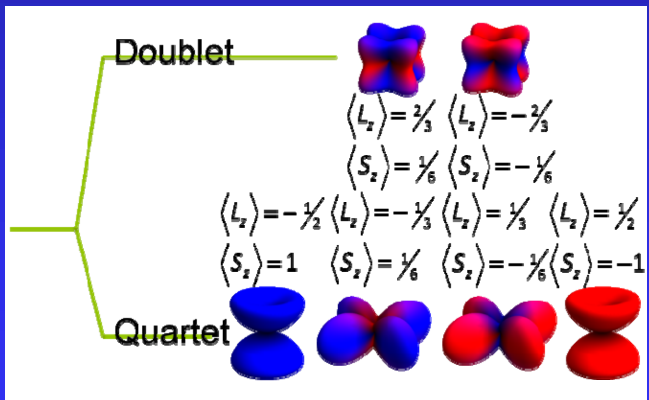

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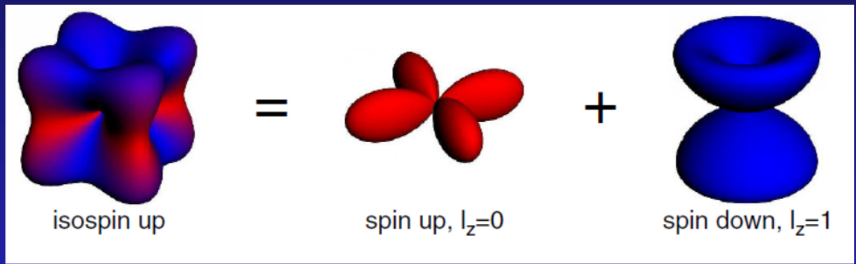

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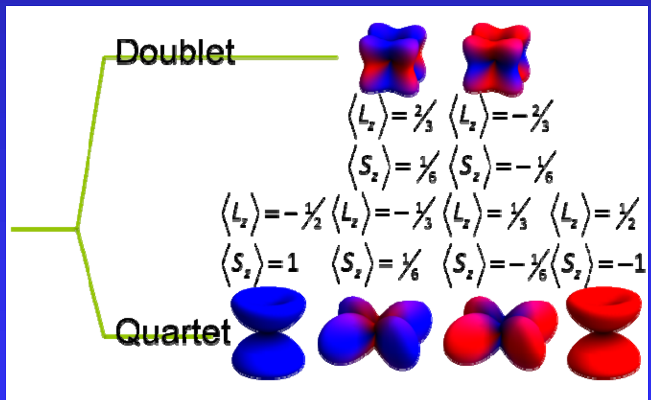

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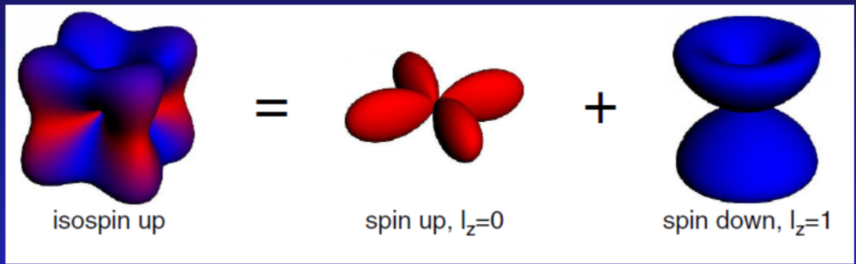

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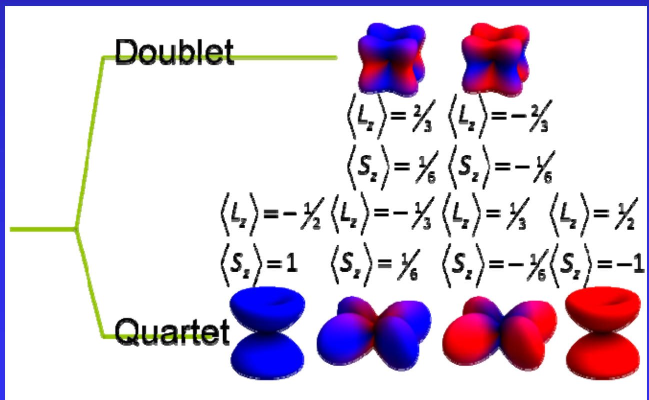

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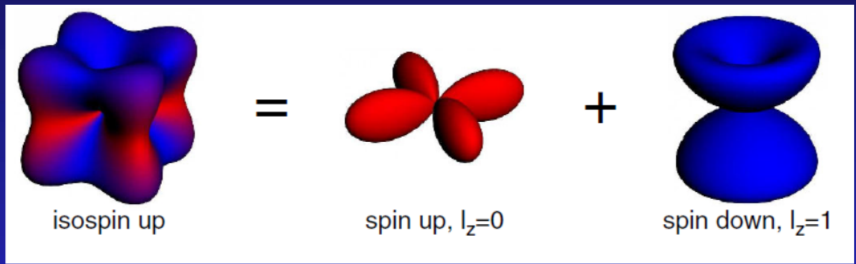
The J_{eff}=1/2 Mott ground state



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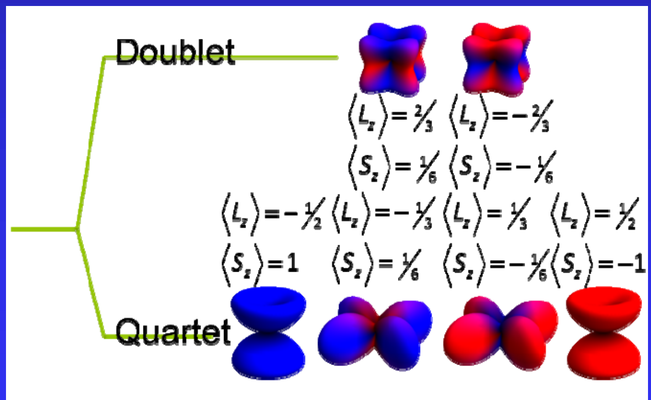

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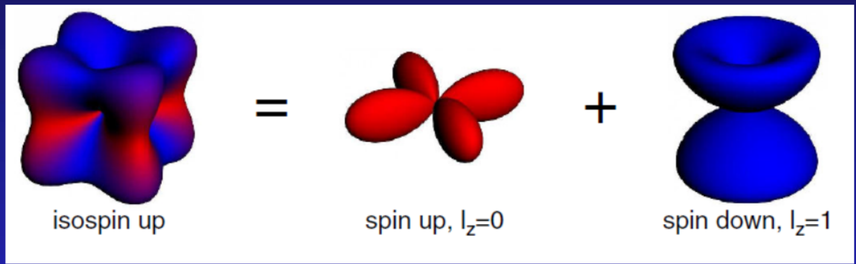

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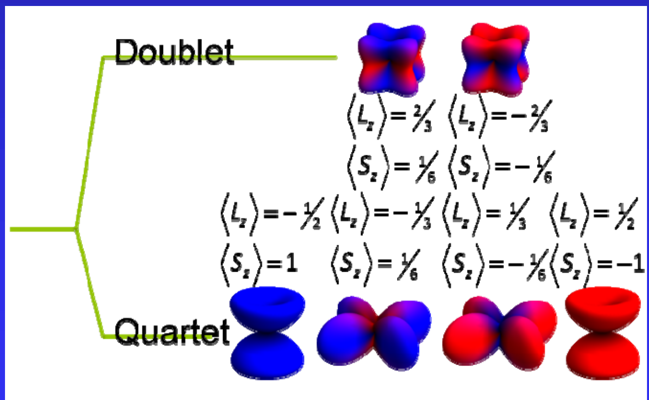

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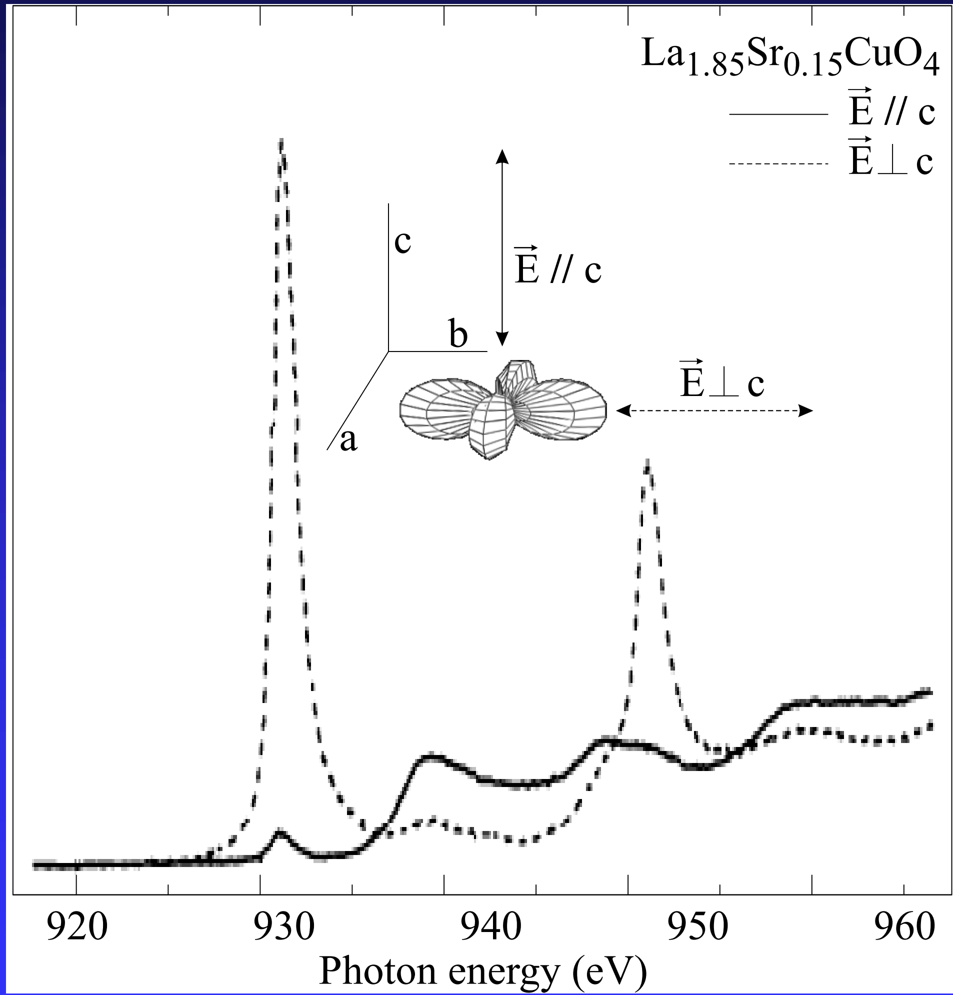
Find this out with polarization dependent spectroscopy

XAS LD (Linear Dichroism)

C. T. Chen *et al.* Phys. Rev. Lett. **68**, 2543 (1992)



Linear Dichroism is sensitive to orbital occupation

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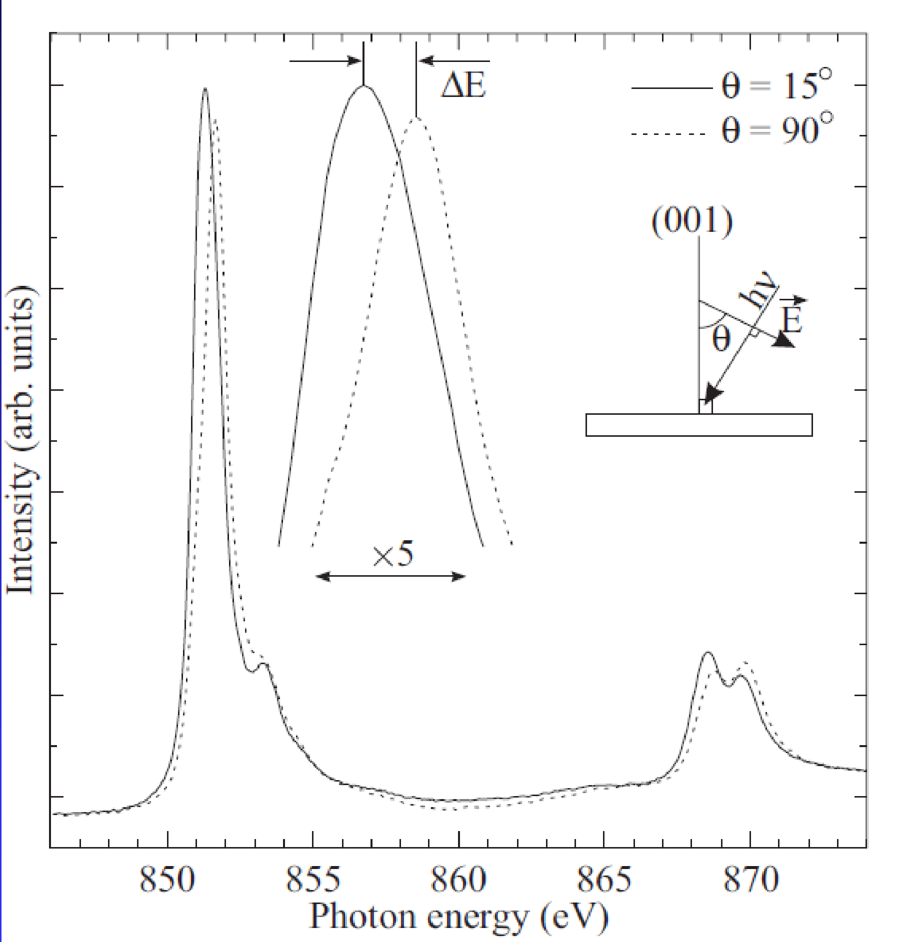
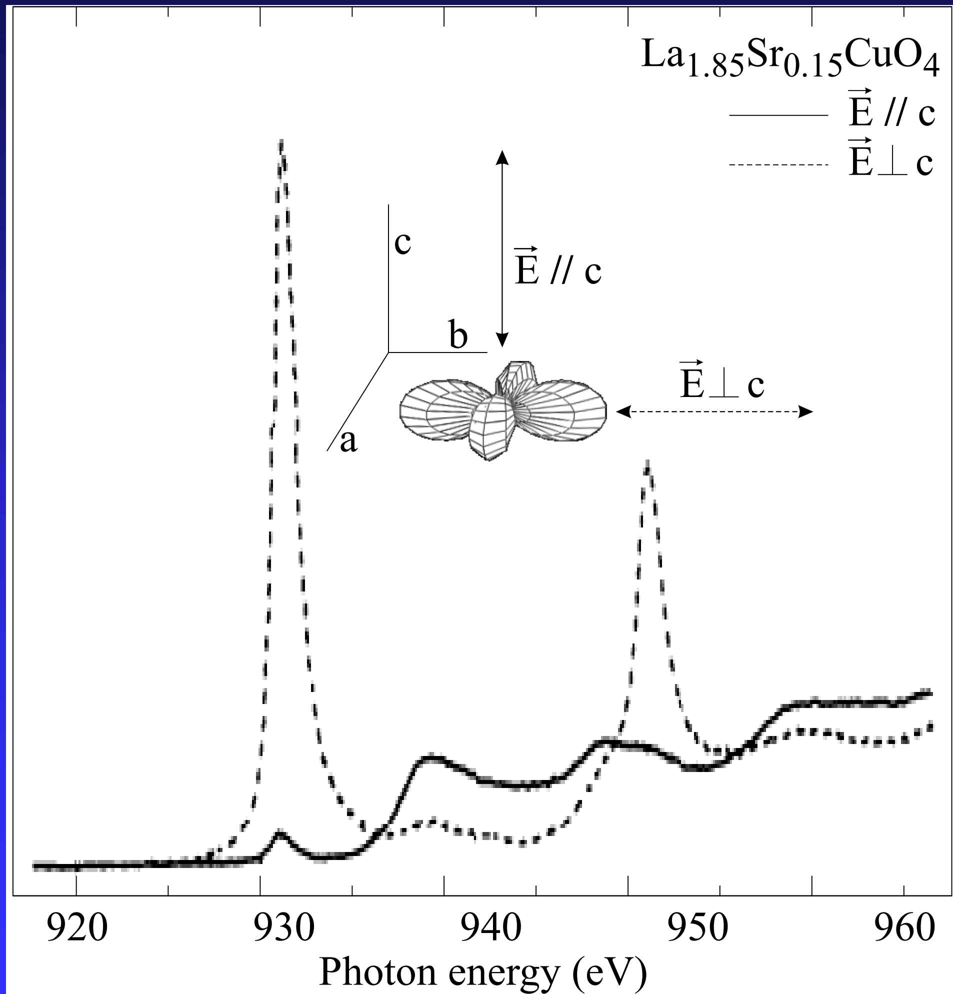
M. Haverkort *et al.* PRB **69**, 020408R (2004)



Sensitive to crystal field splitting

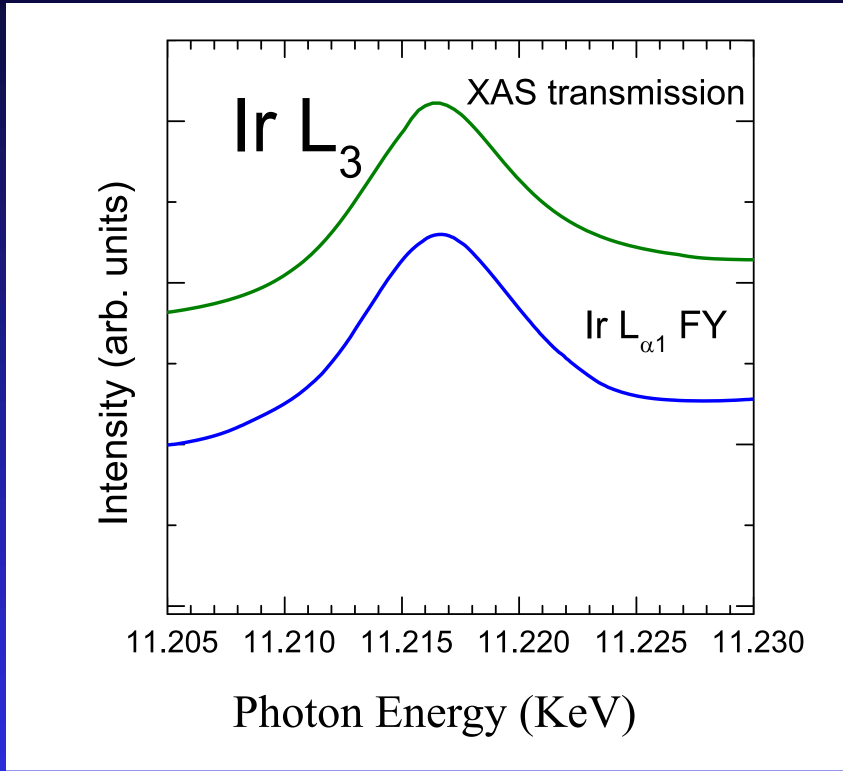
C. T. Chen *et al.* Phys. Rev. Lett. **68**, 2543 (1992)



Linear Dichroism is sensitive to orbital occupation

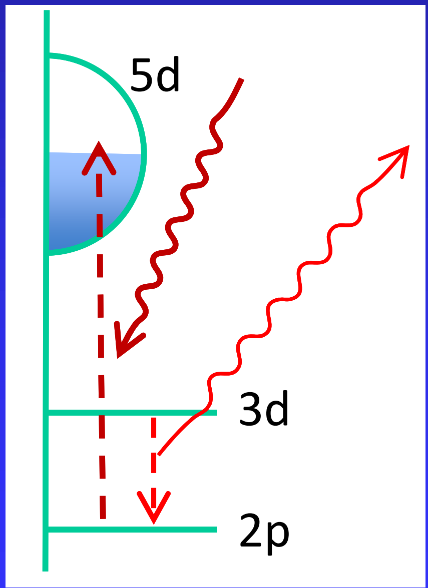
XAS on Ir $L_{2,3}$

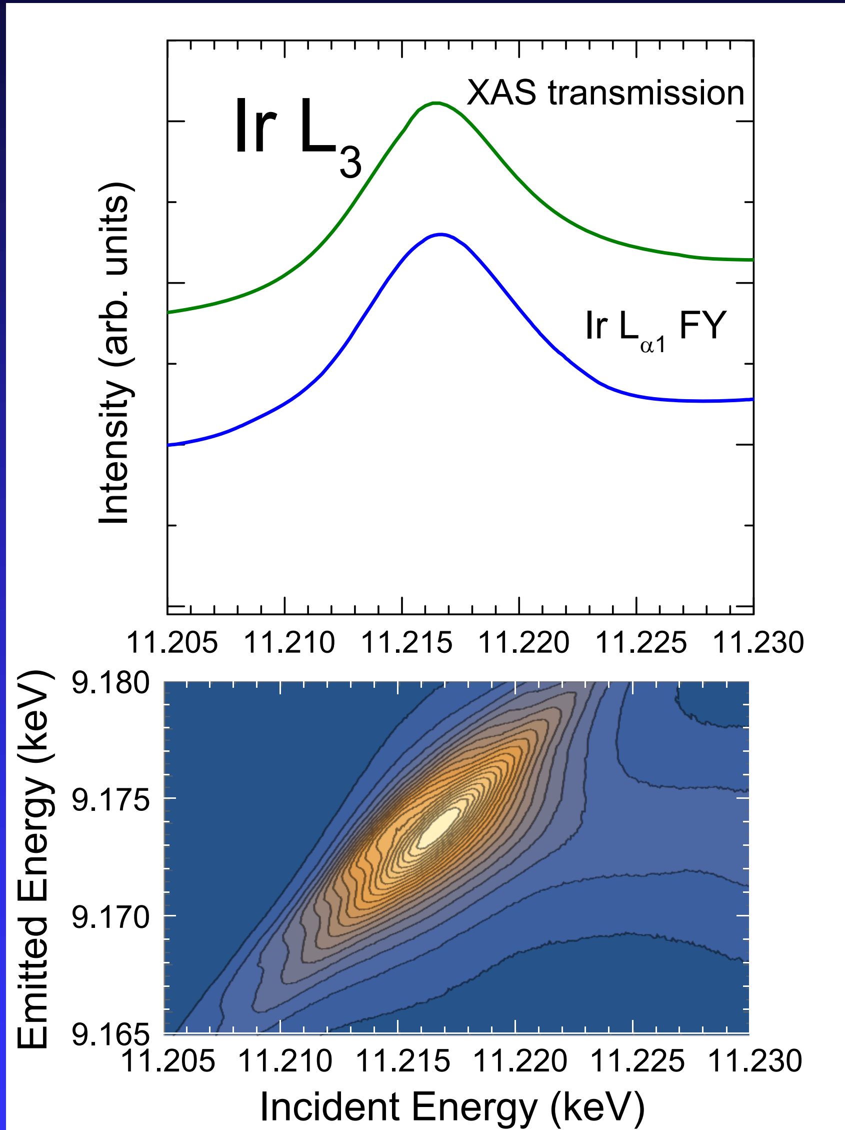
Unfortunately the broadening of the lineshape due to the *2p3/2* core hole lifetime makes the spectrum rather featureless



RIXS on Ir L_3

- Scanning the incoming photon energy
- Analysing the emitted photon energy





(Ir-L_3-M_5: 2p→5d, 3d→2p)

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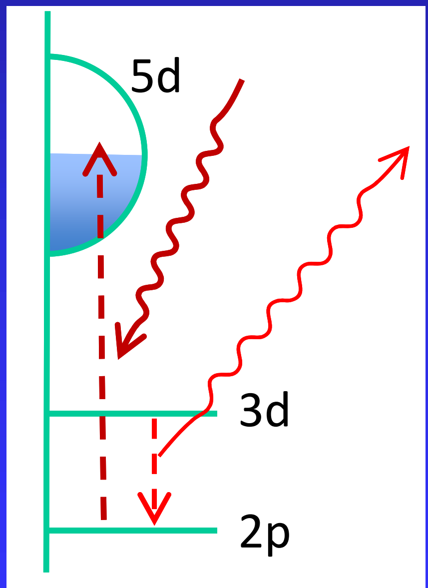

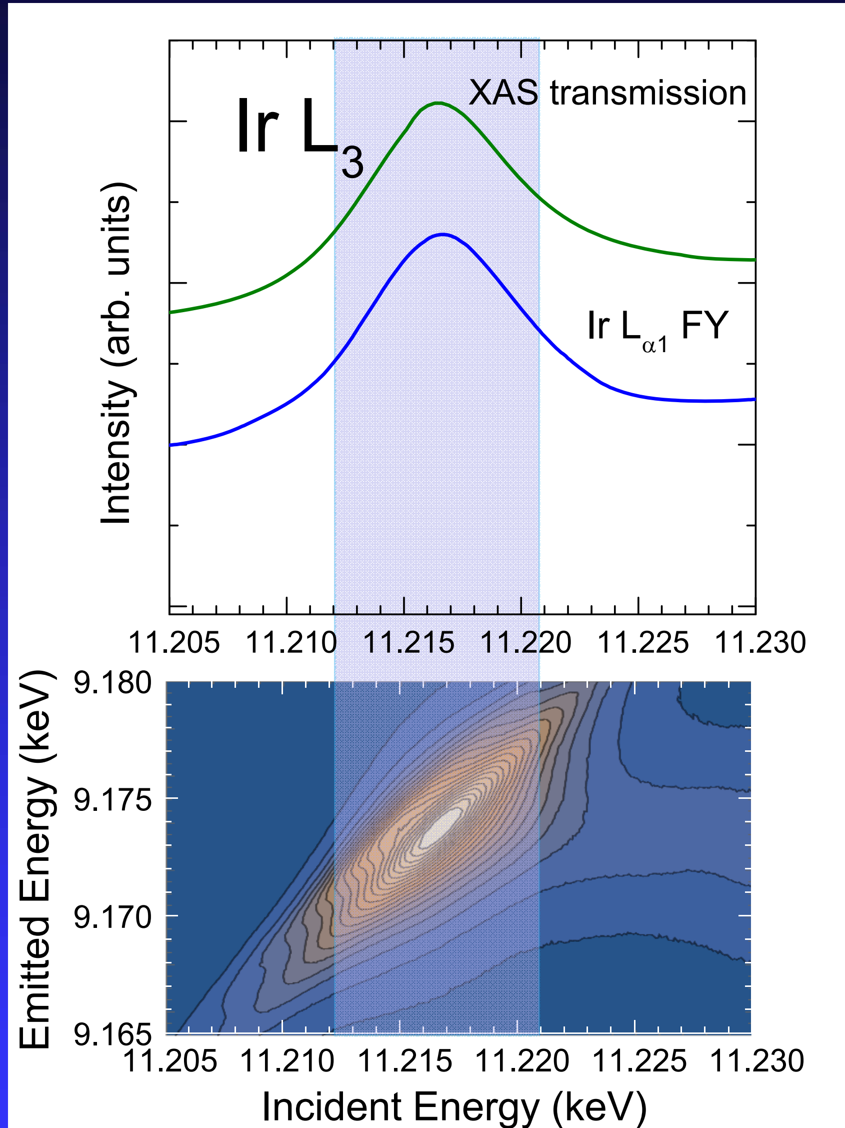

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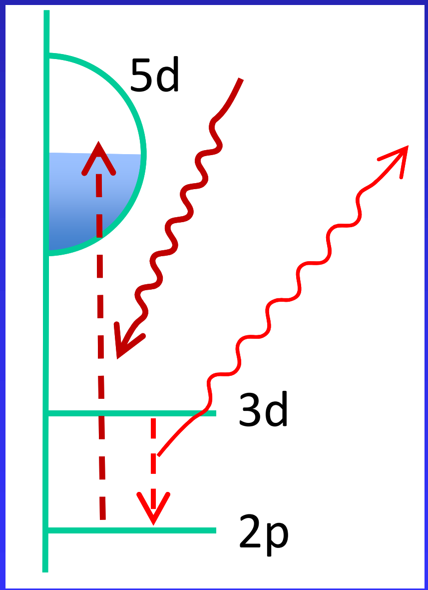

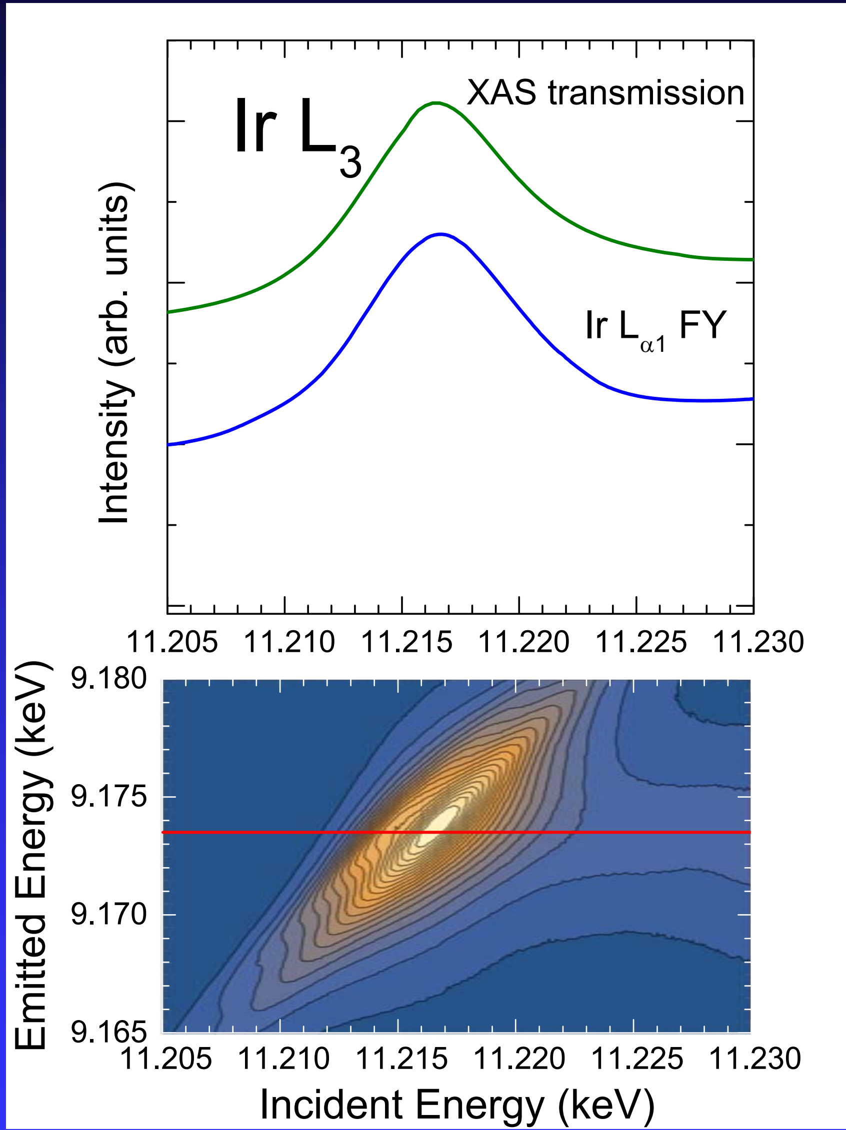

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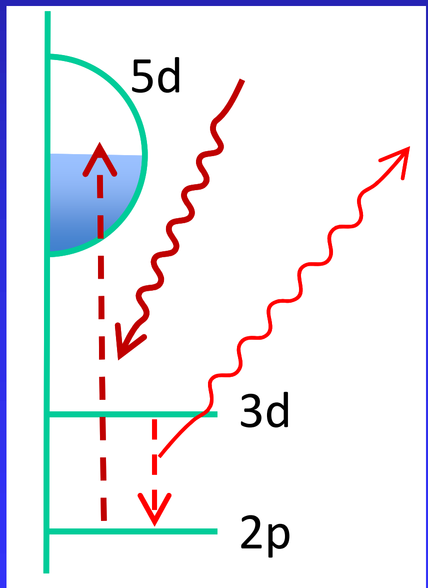

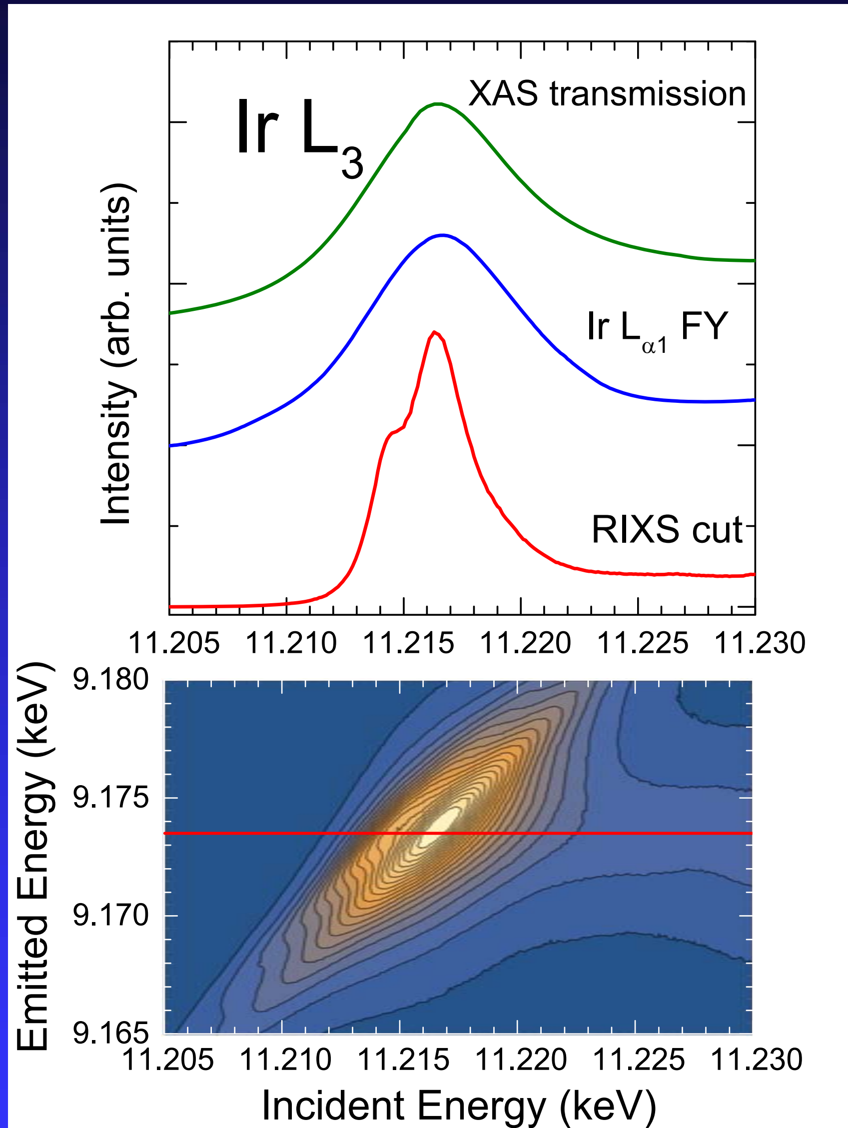

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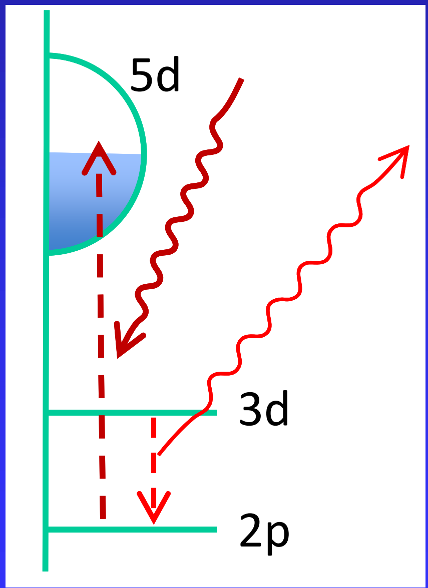

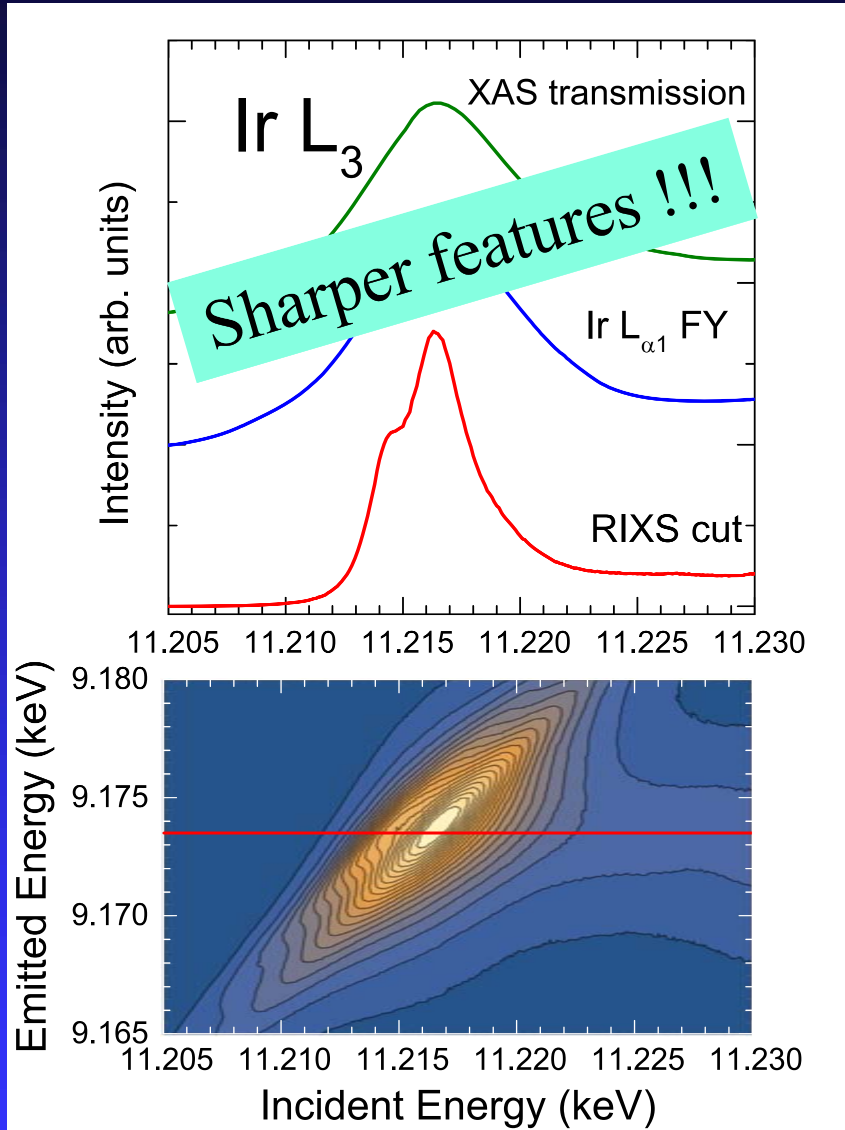

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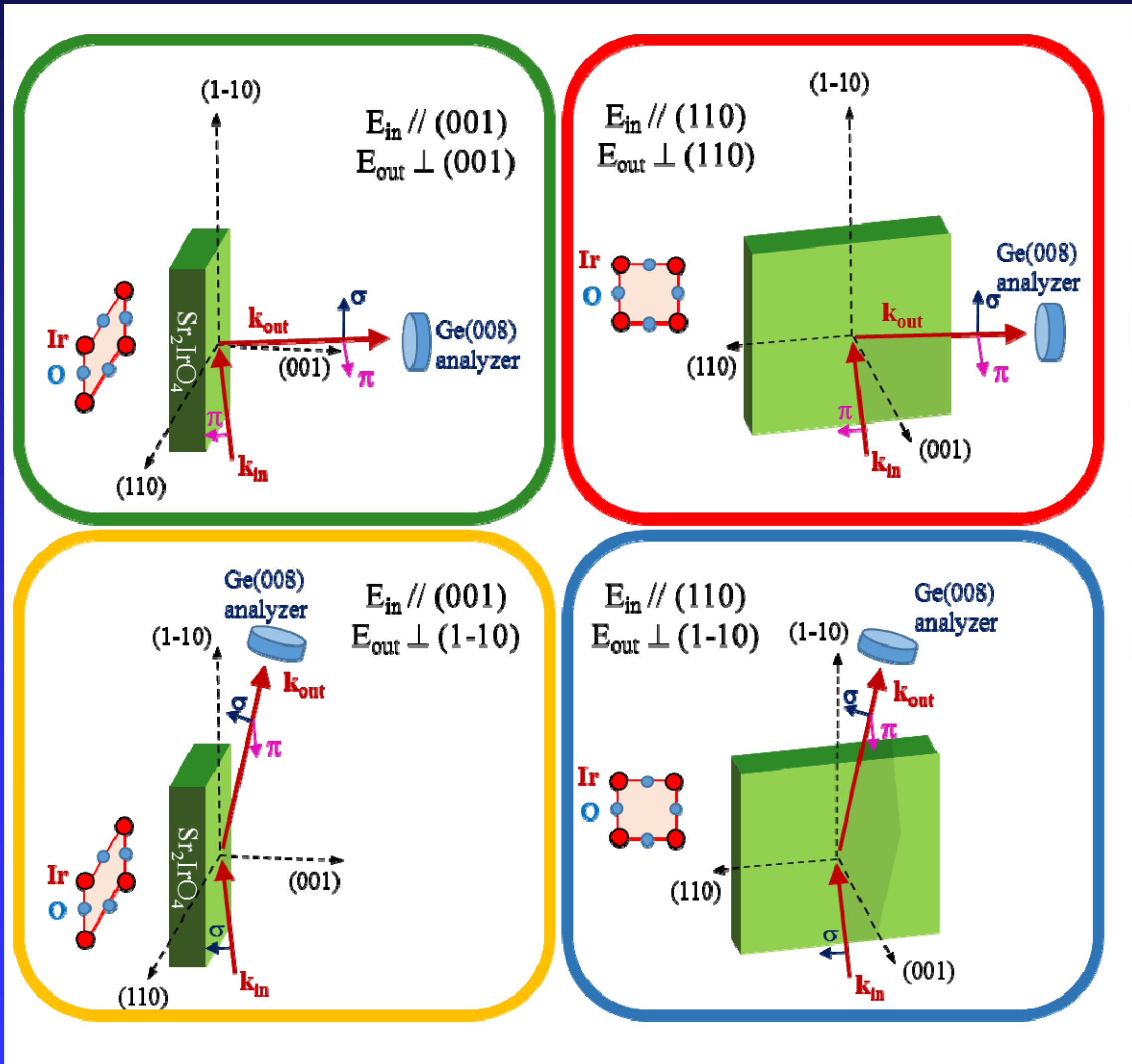
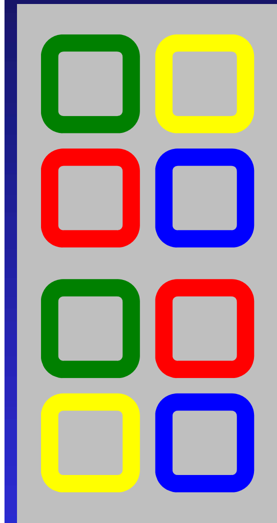
Polarization dependent RIXS





Polarization in: ~c direction

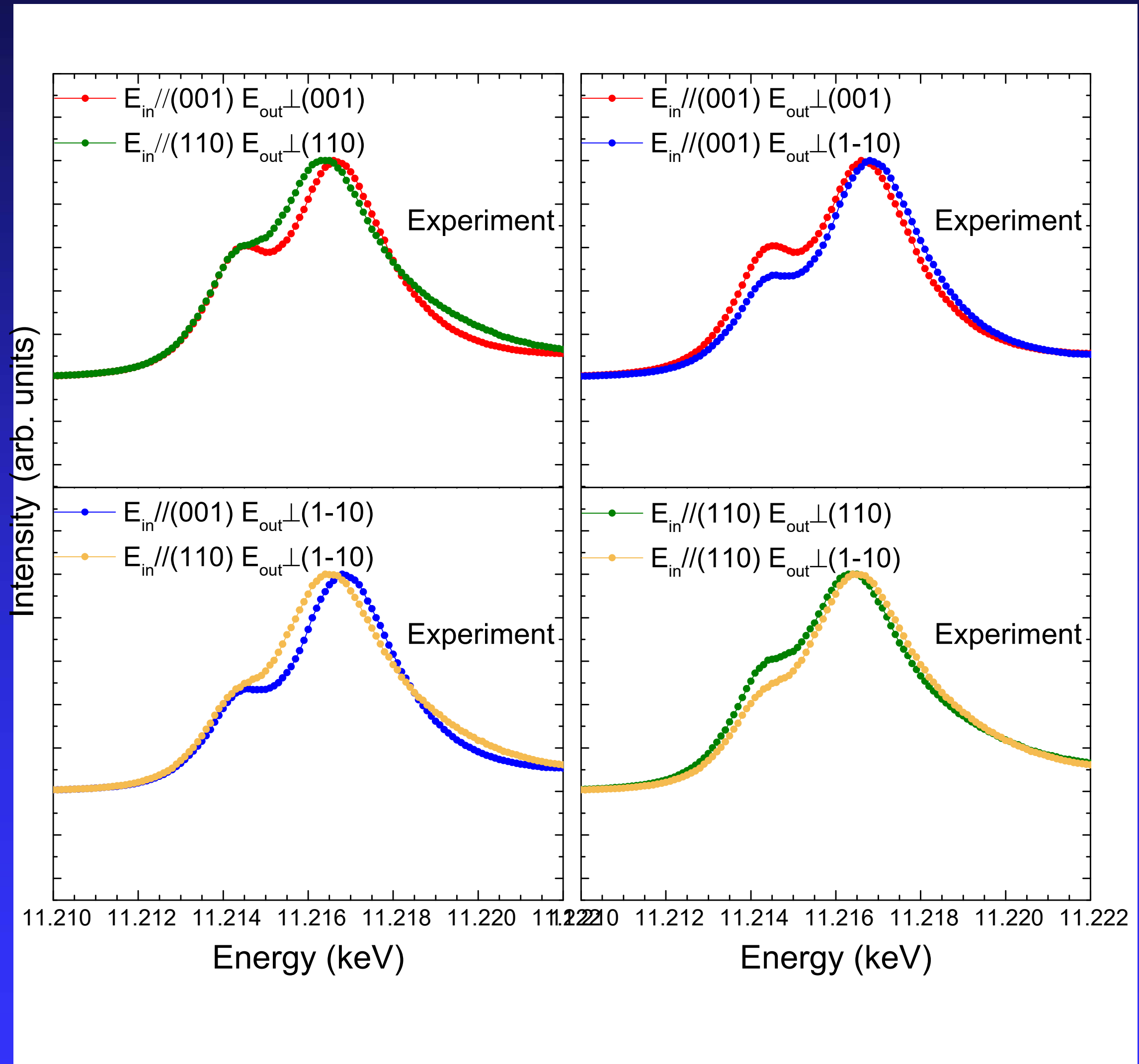
Polarization in: ~(a+b) direction

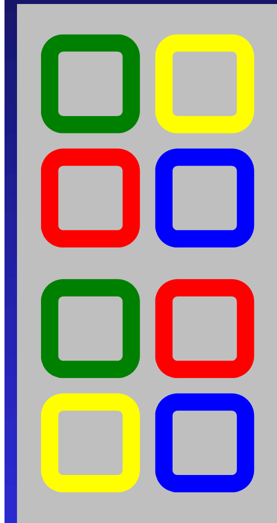
‘Cross’ polarized scattering

‘Parallel’ polarized scattering

Polarization dependent RIXS

Ir-L_3M_5 RIXS





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S. Agrestini et al., Phys. Rev. B **95**, 205123 (2017)

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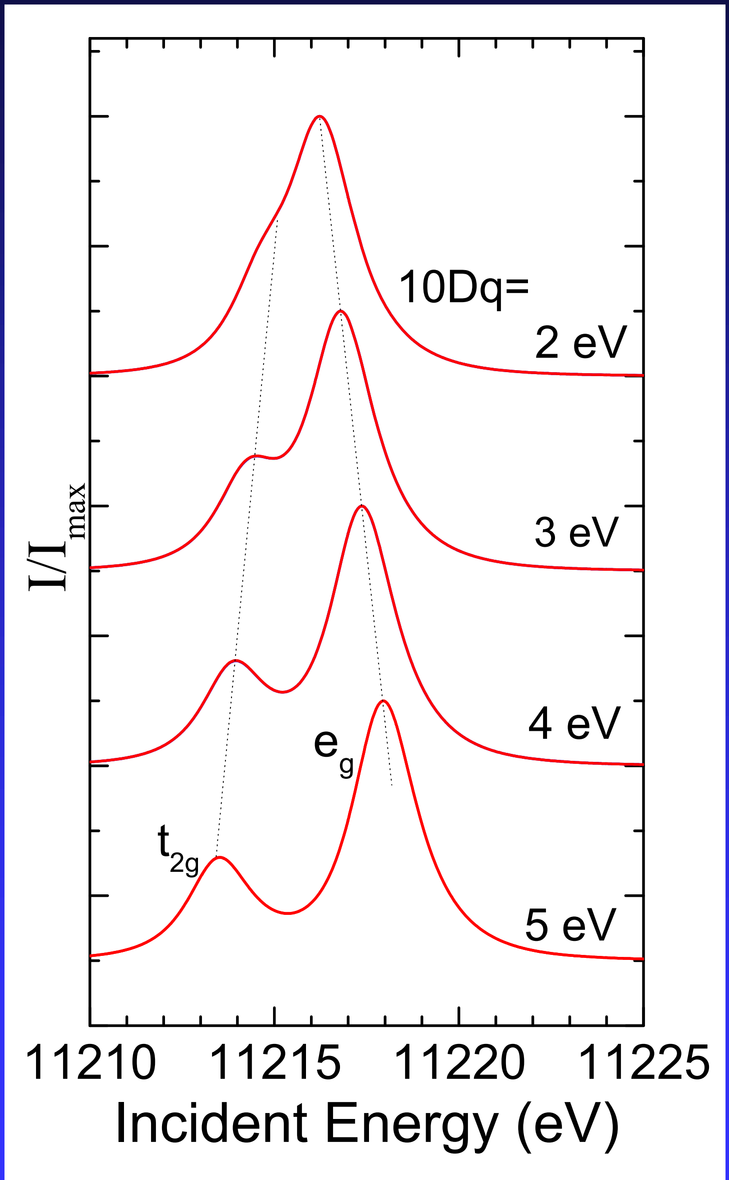
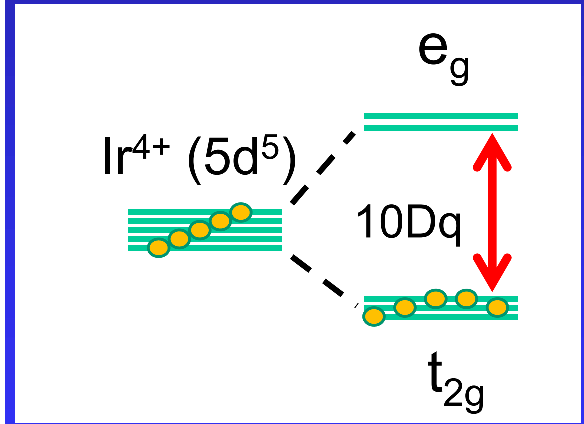


Ir-L_3M_5 RIXS

Ionic model

Cubic symmetry

Slater Integrals (SI)=0% atomic HF



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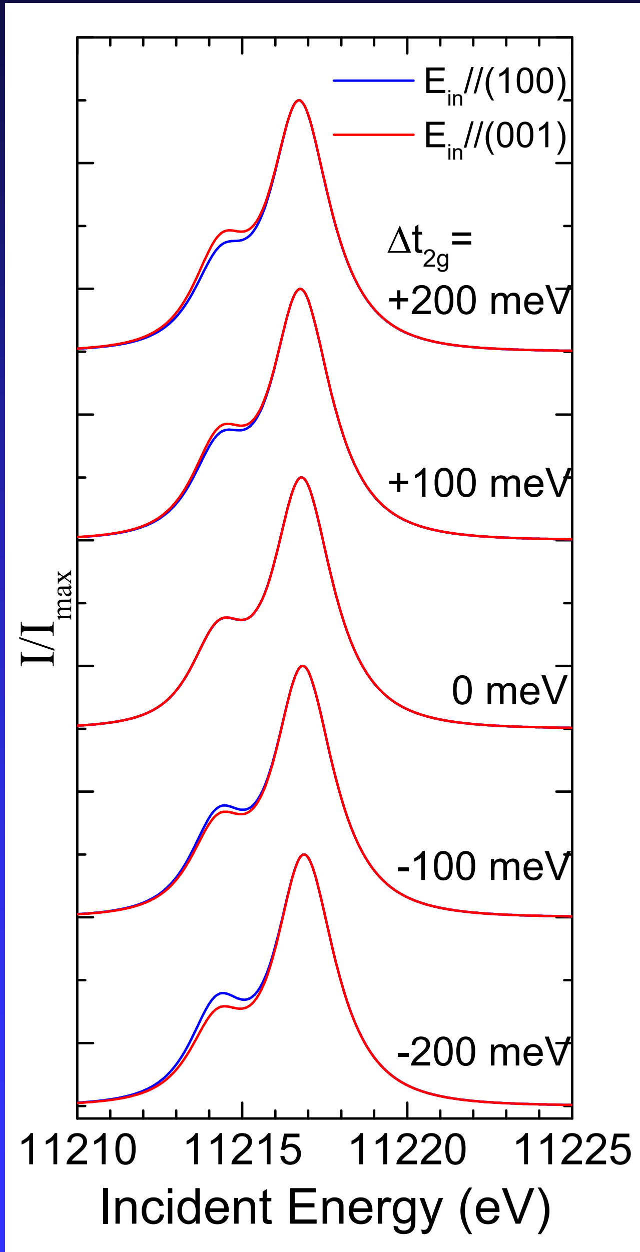

Ir-L_3M_5 RIXS

Ionic model
Tetragonal distortion
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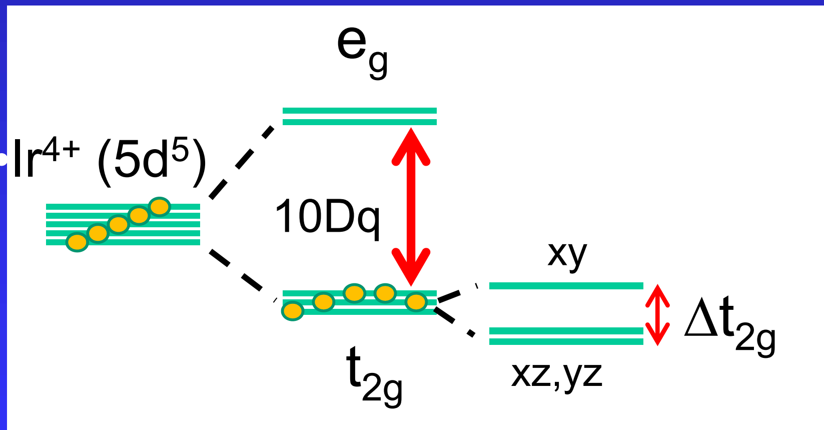

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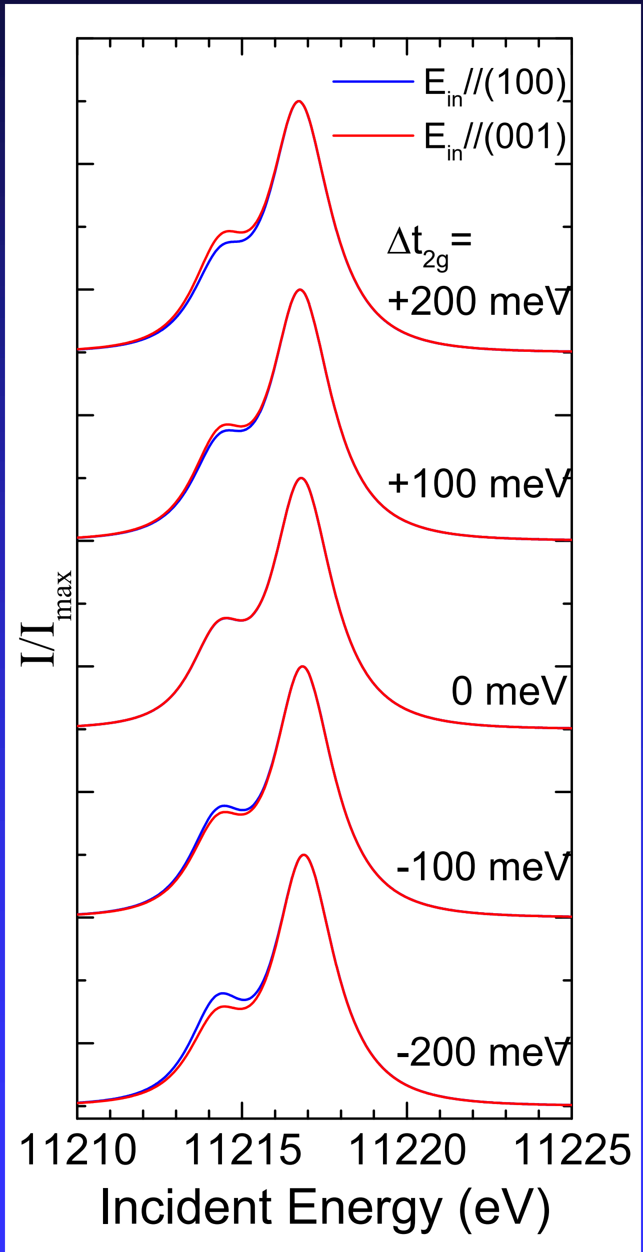

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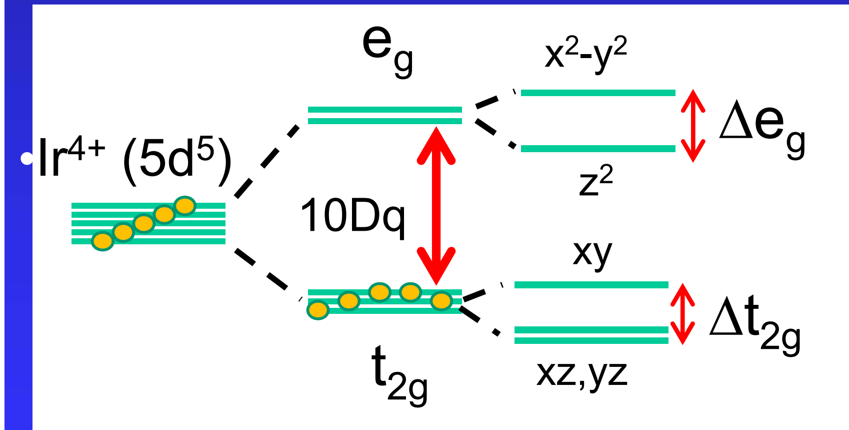
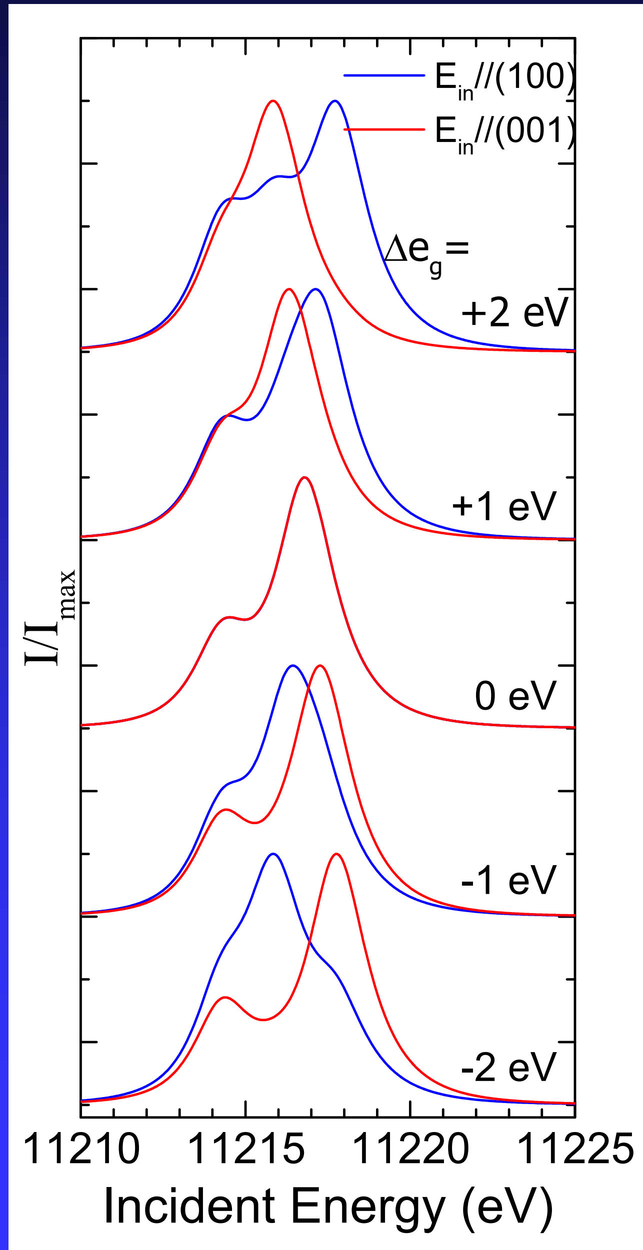
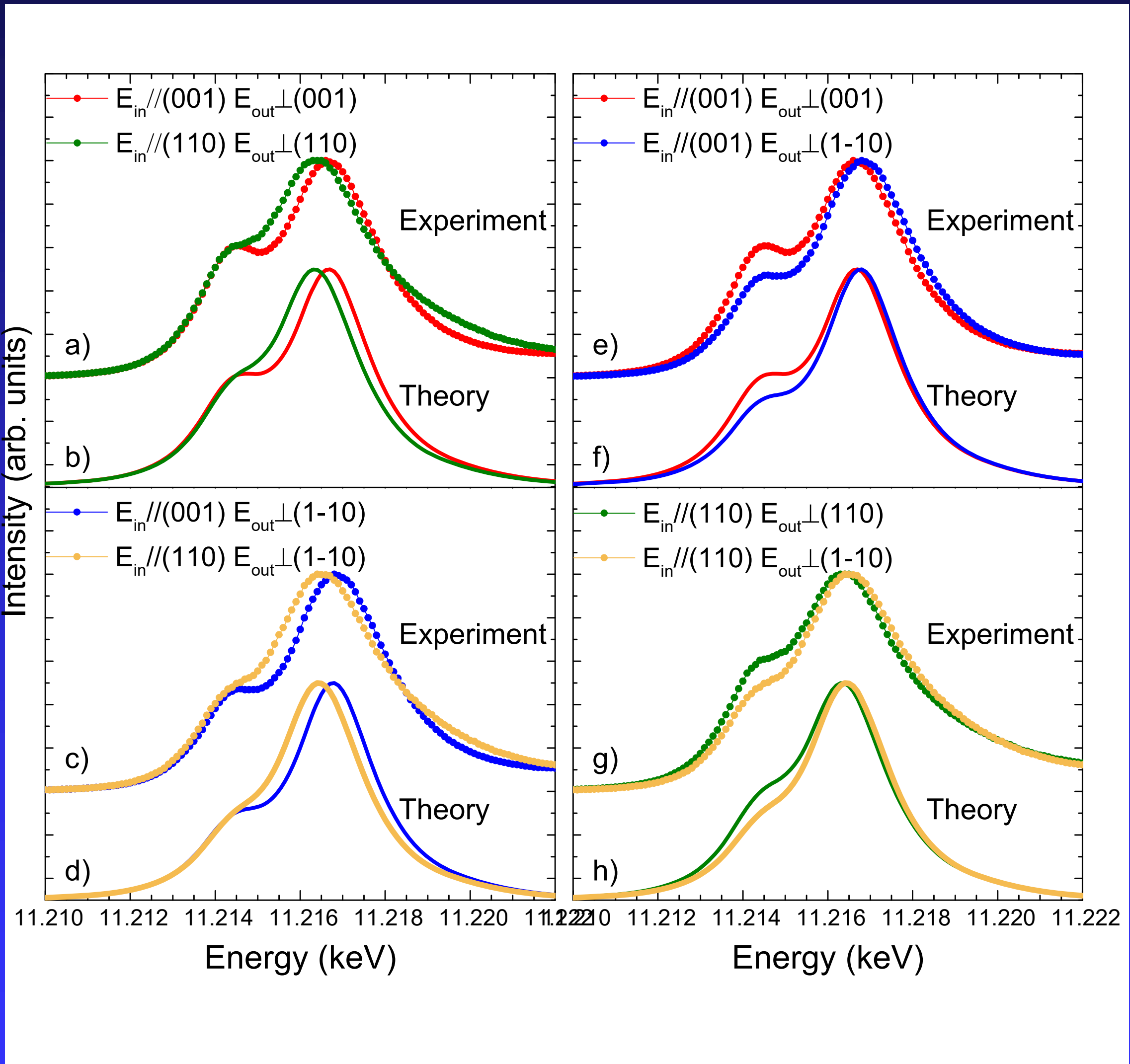




RIXS: fit with ionic model



S. Agrestini et al., Phys. Rev. B **95**, 205123 (2017)

Forced ionic fit

SOC = 0.4 eV

10Dq = 2.7 eV

Δt_{2g} = -150 meV

Δe_g = -0.5 eV

Slater Integrals = 20% Hartree-Fock

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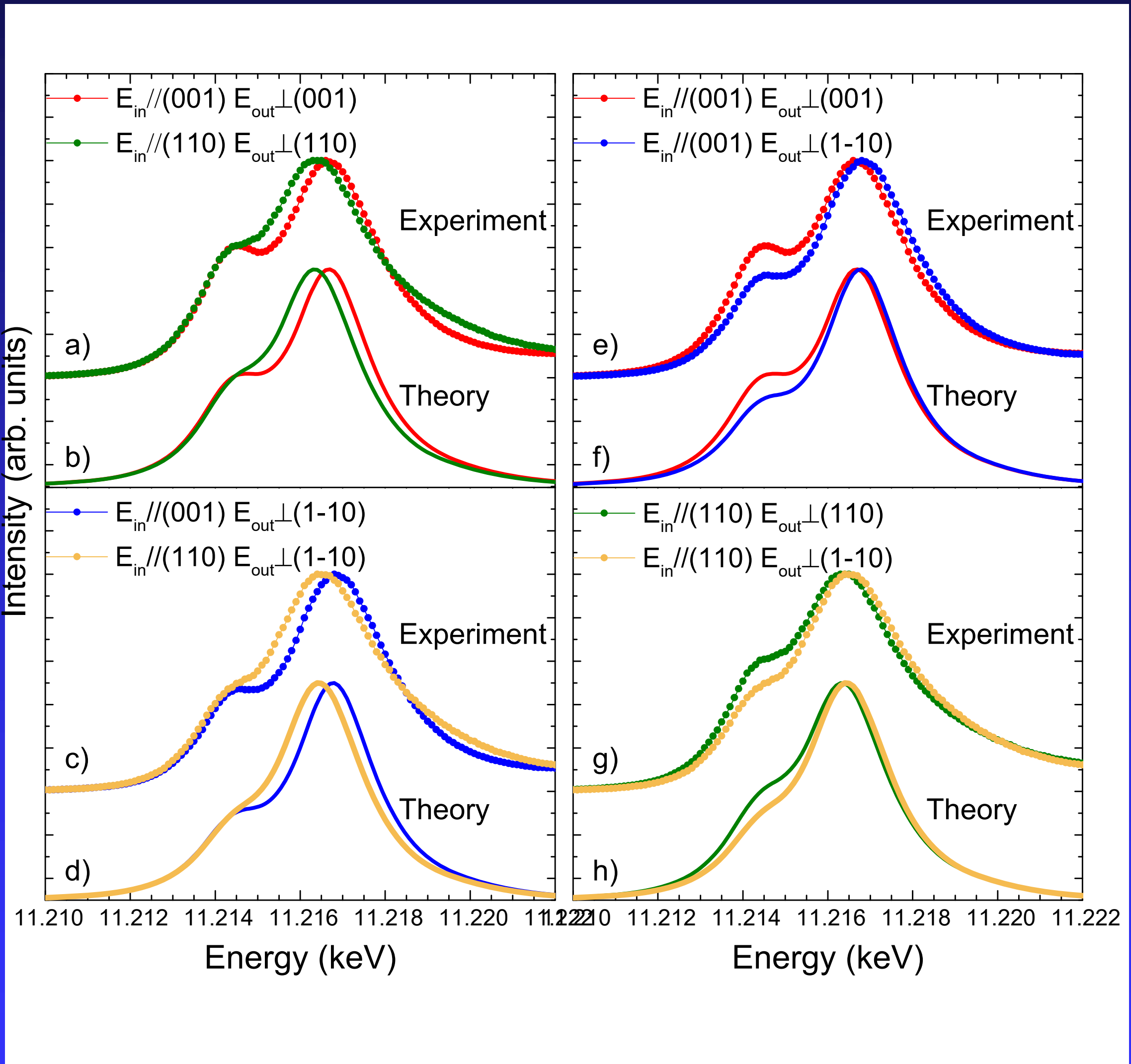

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Not perfect for J_{eff}=1/2
but not too bad

However Slater Integrals
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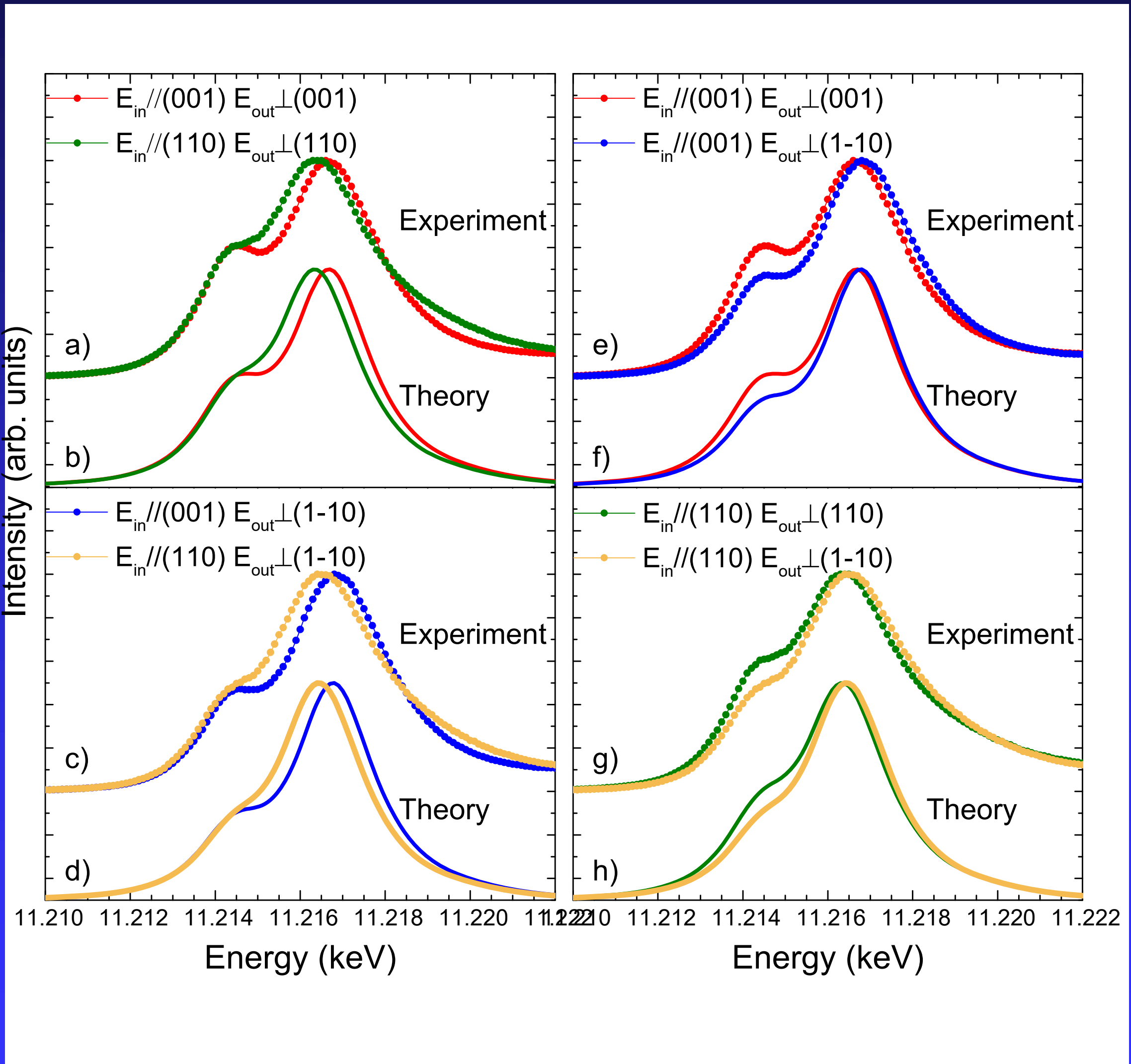

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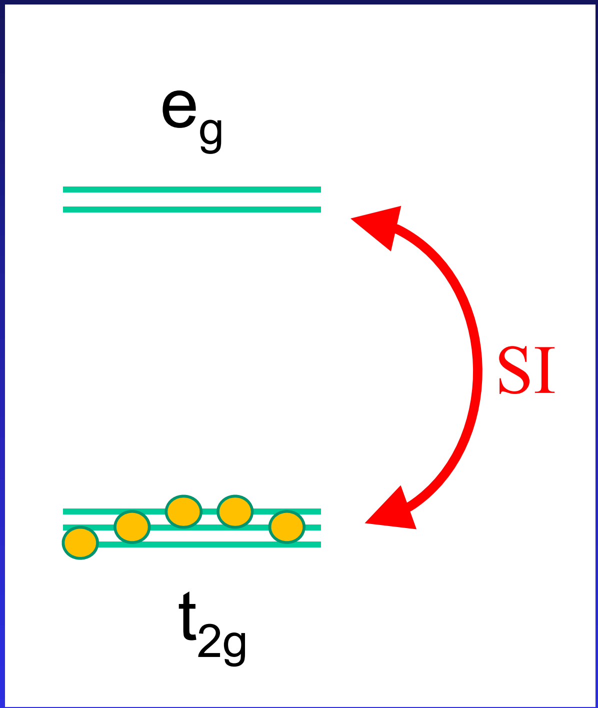

Slater Integrals mix
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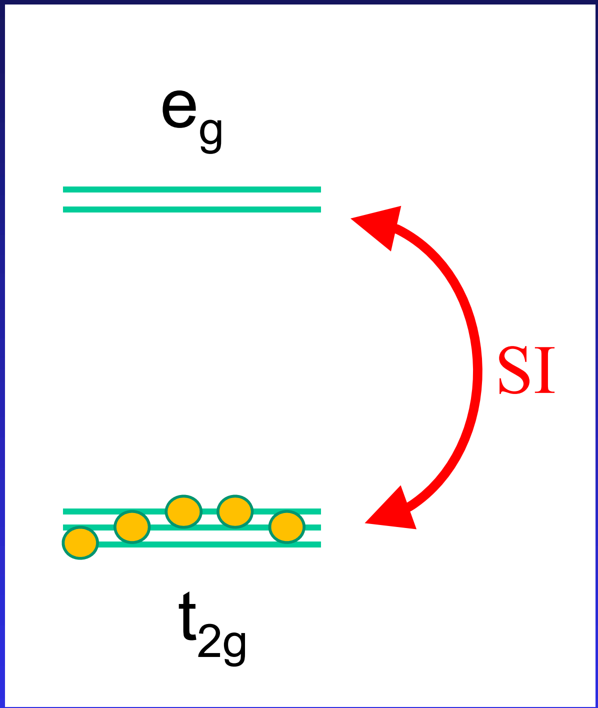

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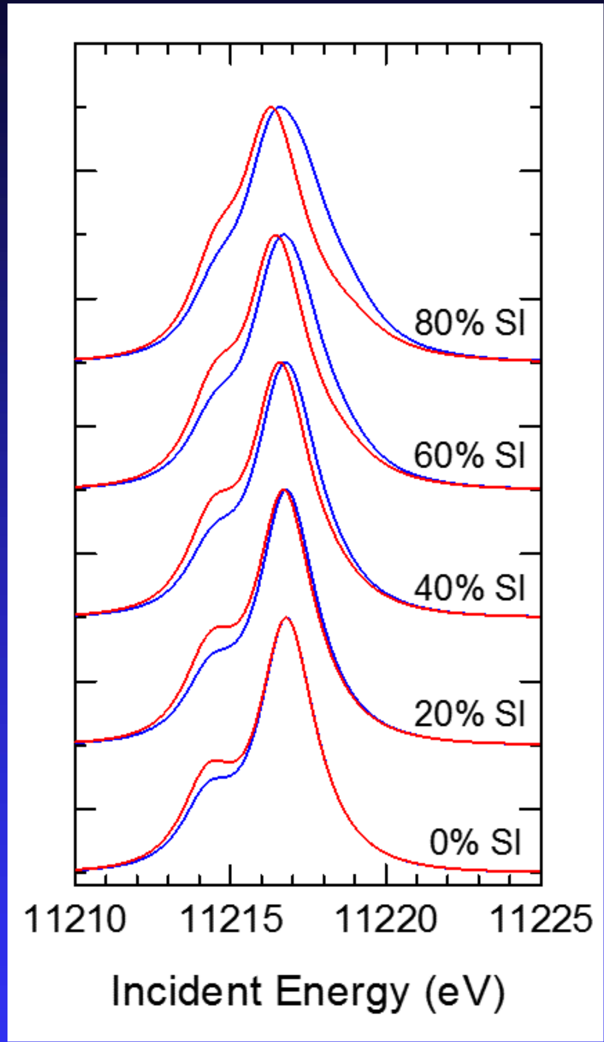
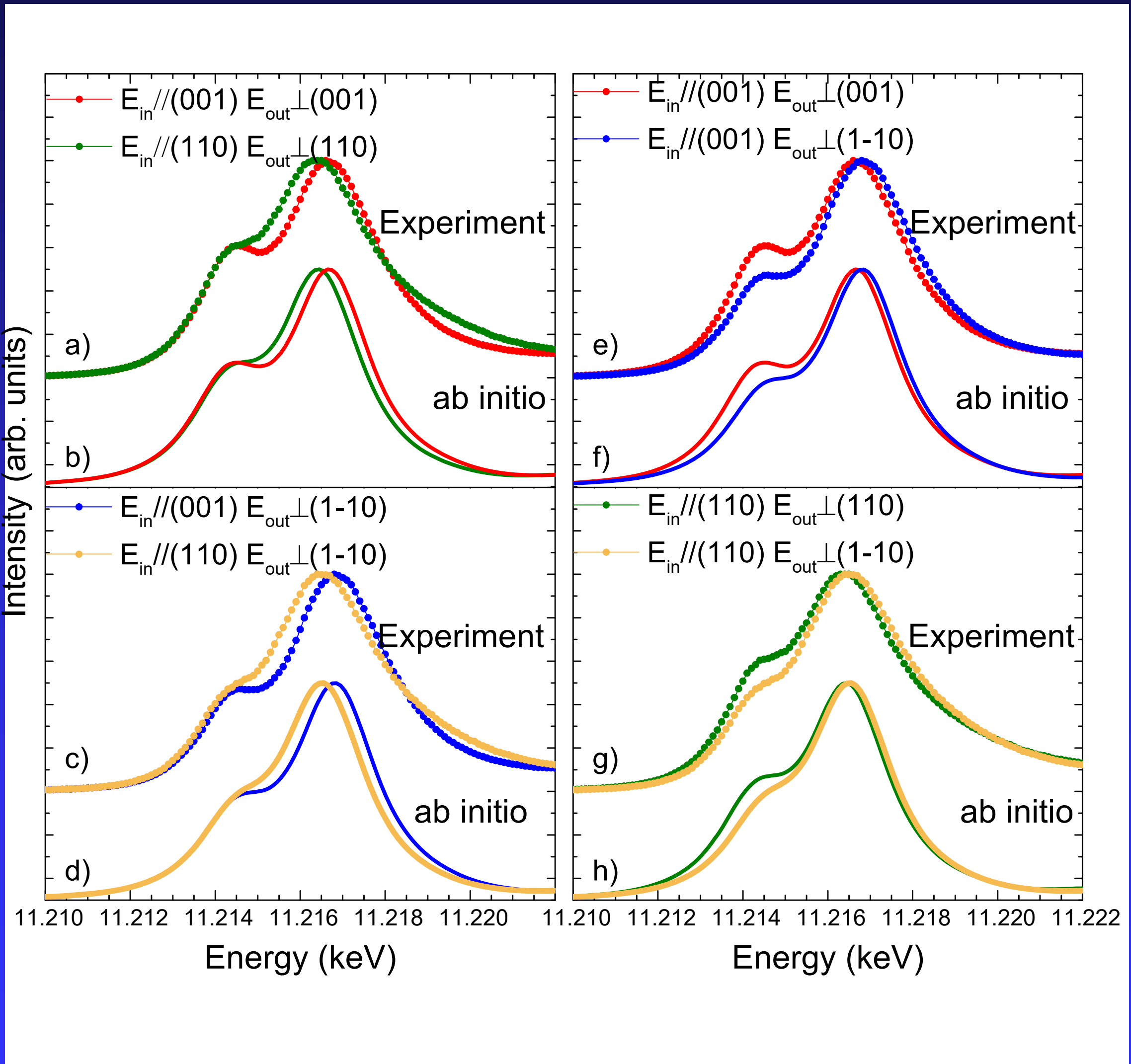


Linear Dichroism between

'Cross' polarized scattering (horizontal)

'Parallel' polarized scattering (vertical)

RIXS: Ab Initio calculation including hybridization

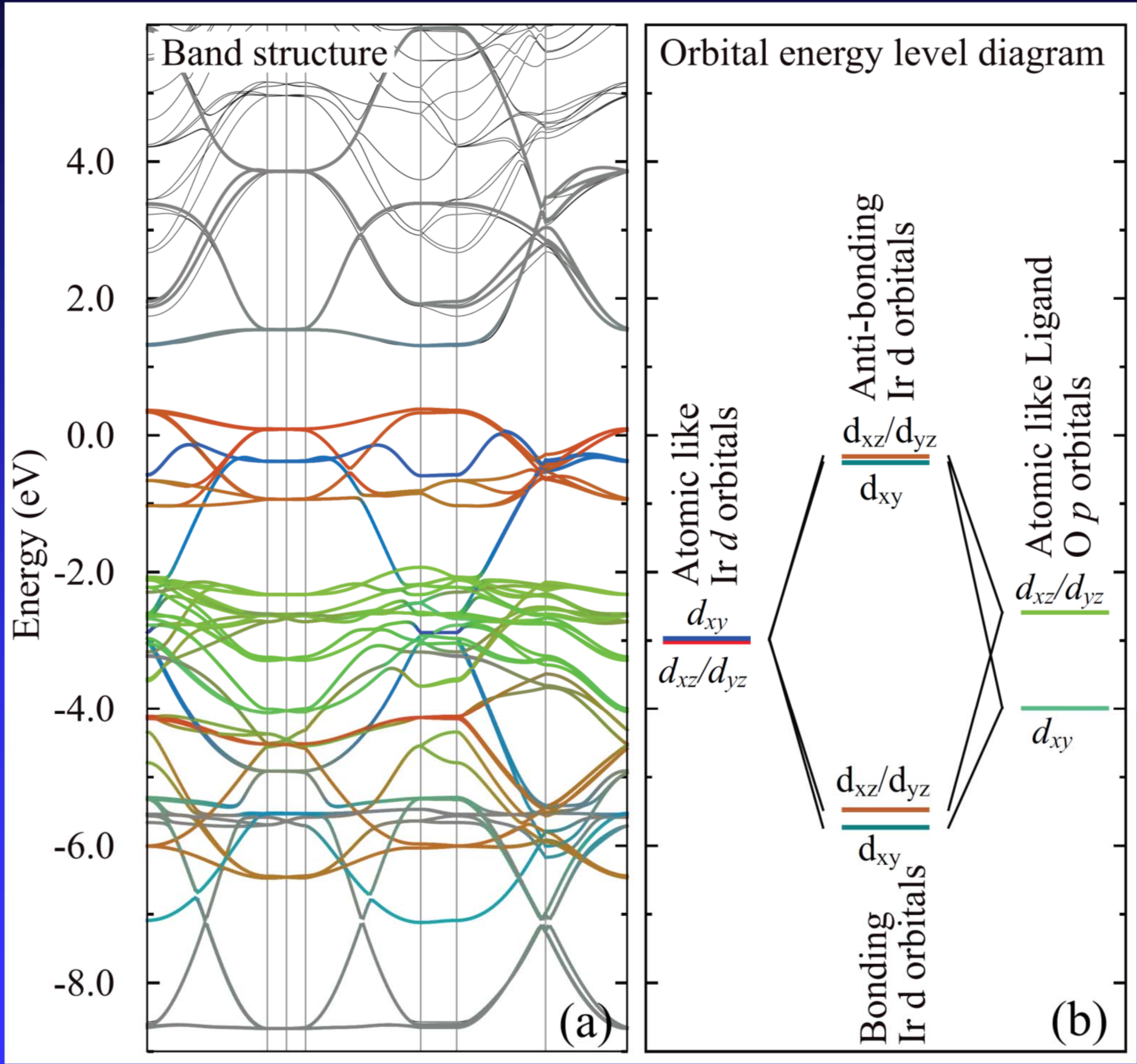


SOC = 0.4 eV
$10Dq_{eff}$ = 2.5 eV
Δt_{2g}= -160 meV
Δe_g= -0.5 eV
Slater Integrals = 75% Hartree-Fock

O-2p to Ir-5d
charge transfer energy ≈ -1.5 eV
Ve_g ~ 4.3-4.5 eV
Vt_{2g} ~ 2.5-2.9 eV
Vmix ~ 0.7 eV

S. Agrestini et al., Phys. Rev. B **95**, 205123 (2017)

Orbital energy level diagram



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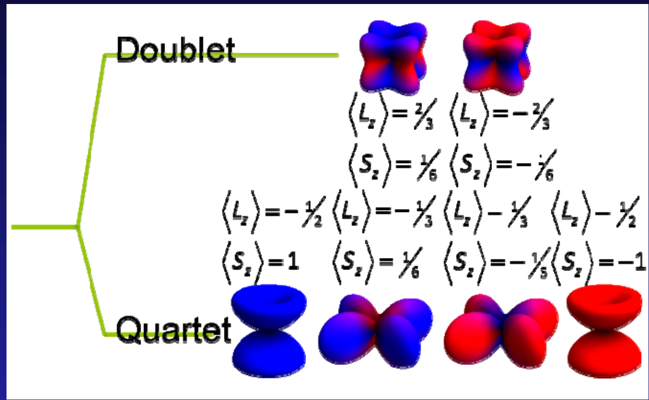
PRB **95**, 205123 (2017)

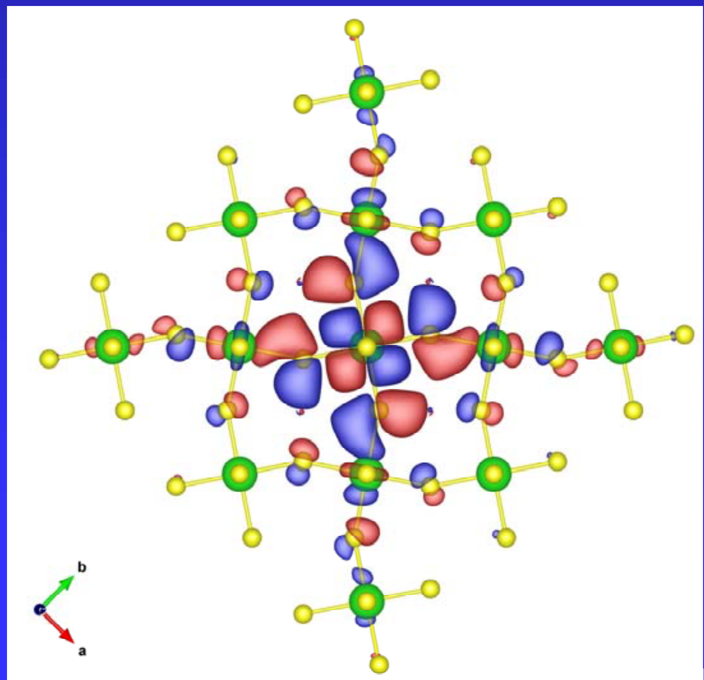
t_{2g} orbitals in Sr_2IrO_4

Cubic orbitals in Jeff=1/2 model

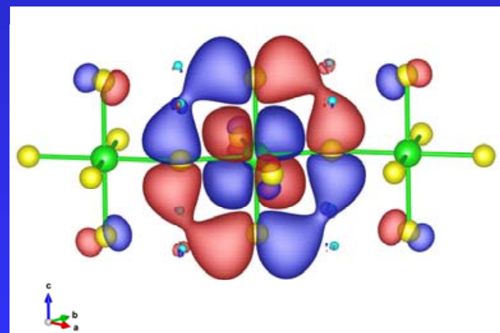


The t_{2g} orbitals in Sr_2IrO_4 are very different from the cubic orbitals considered in the J_{eff}=1/2 model .

d_{xy}



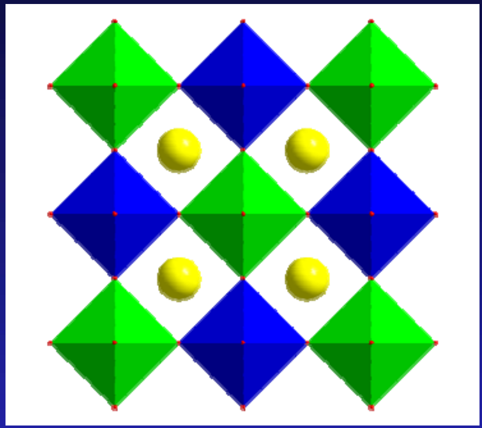
d_{xz} / d_{yz}

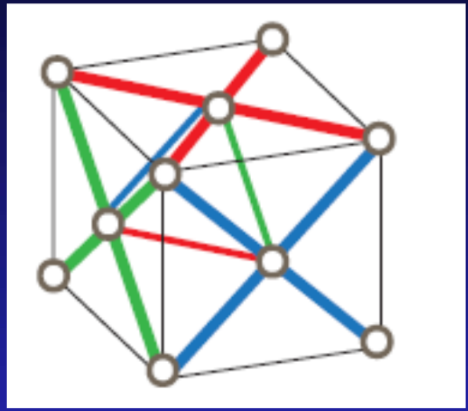


orbitals (xy) and (xz/yz) have very different shape, extension and interactions.

S. Agrestini et al., PRB **95**, 205123 (2017)

How to solve the covalency problem ?

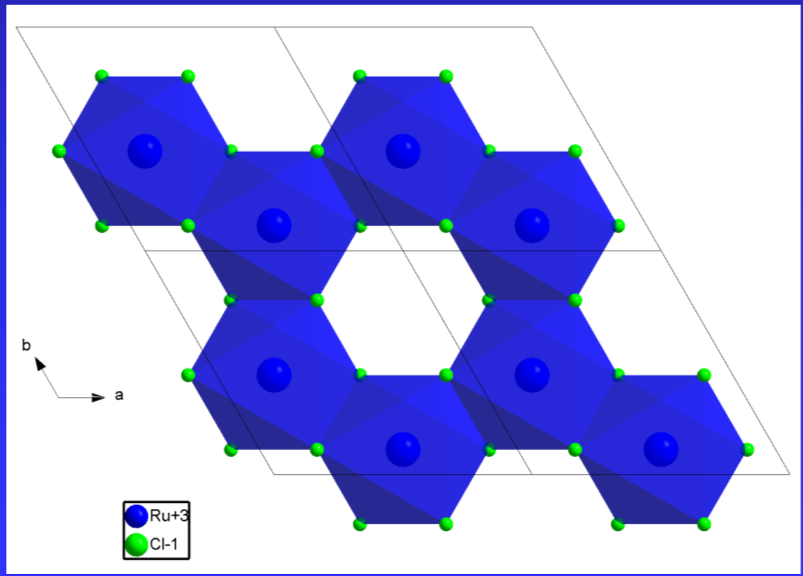




- Enhance distance between Ir atoms to create nearest neighbor models: double perovskites.

- Reduce covalency : less charged d^5 ions (Os^{3+}, Ru^{3+}) and/or fluorine compounds

- Tuning the Ir-O-Ir bonding angle (Roser Valenti, Takagi)



$RuCl_3$

Conclusions

- The 160 meV intra t_{2g} splitting compared to the SOC brings Sr_2IrO_4 “somewhat” away from the cubic symmetry
- We found that Ir4+ compounds are highly covalent
- The orbitals (xy) and (xz/yz) have very different shape, extension and interactions.
- Anisotropic long range interactions prevent a nearest neighbor Kitaev or other compass model to be formed.
- The reduction of covalency is a key ingredient in the design of Kitaev materials

I thank you very much
for
your kind attention