Theory of Spectroscopy on Strongly Correlated Electron Systems

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Leibniz Institute for Solid State and Materials Research Dresden

TECHNISCHE UNIVERSITÄT DRESDEN

Ament, van Veenendaal, Devereaux, Hill & JvdB
Rev. Mod. Phys. 83, 705 (2011)

7th MaNEP Winterschool
Saas Fee 09.01.2017

Lectures: outline

1. Intro to Spectroscopy, Correlations & RIXS

2. Theory for RIXS scattering amplitude

3. Magnetic RIXS: Theory and Experiment

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Introduce cuprates

Bimagnon RIXS
Cu K-edge

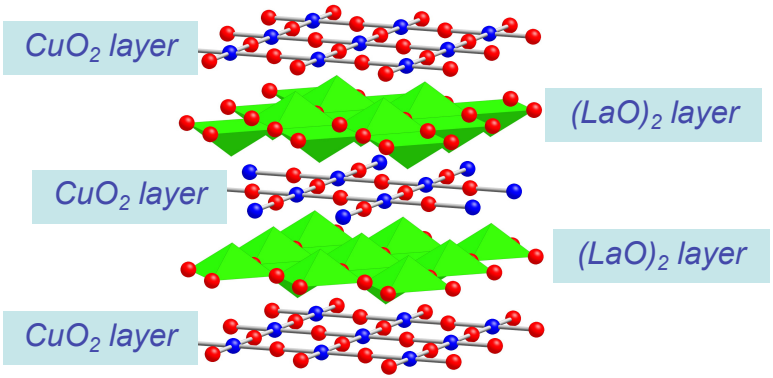
Single Magnon RIXS
Cu L-edge

Magnetic RIXS on iridates

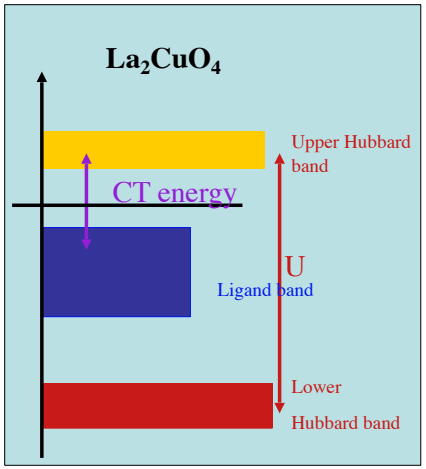
Paramagnons in doped cuprates

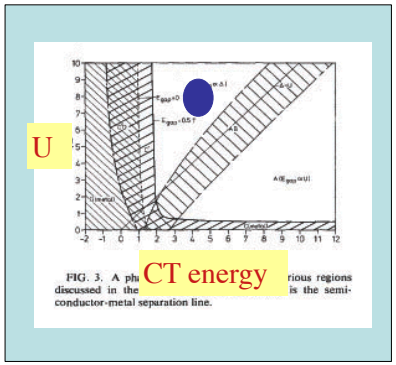
High Tc superconducting cuprates

La_2CuO_4 crystal structure



Correlation Effects: Zaanen-Sawatzky-Allen

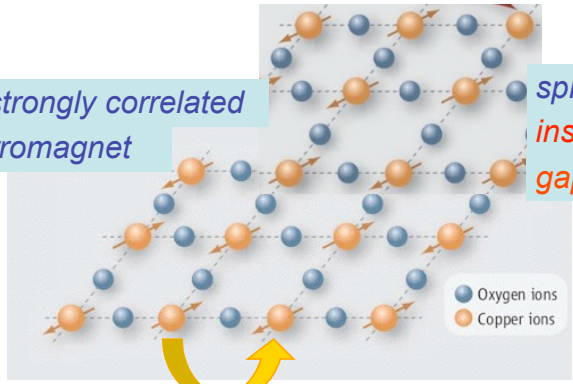




Zaanen, Sawatzky, Allen
PRL 55, 418 (1985)

Charge Transfer Insulator

La_2CuO_4 magnetic structure

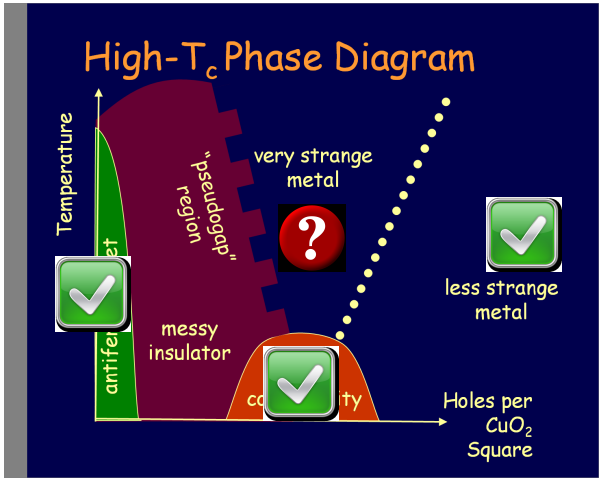


strongly correlated antiferromagnet

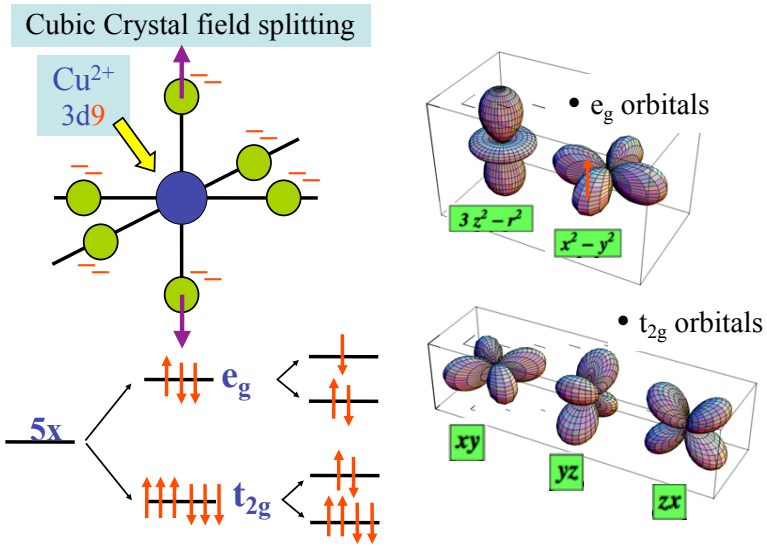
spin 1/2 insulator gap ~2 eV

U ~ 6 eV

Cu-O phase diagram



Atomic Model: Local d-d orbital splitting: Cu^{2+}



Ultra-short Core-hole Life-time expansion

short life-time τ of the high energy core-hole

large core-hole broadening $\Gamma=\hbar/\tau$

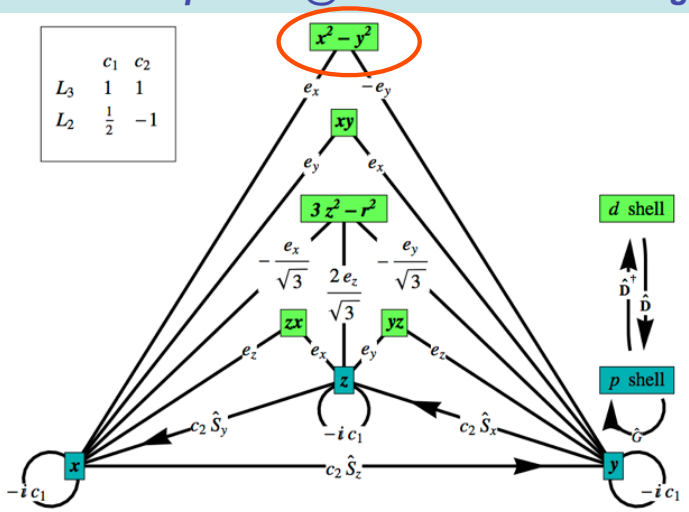
RIXS amplitude $\mathcal{F}_{fg} = \langle f|\mathcal{D}'^{\dagger} G(z_{\mathbf{k}})\mathcal{D}|g\rangle$

$z_{\mathbf{k}} = E_g + \hbar\omega_{\mathbf{k}} + i\Gamma$ *large*

$$G(z_{\mathbf{k}}) = \frac{1}{z_{\mathbf{k}} - H} = \sum_n \frac{|n\rangle\langle n|}{z_{\mathbf{k}} - E_n}$$ *≈ constant*

RIXS response governed by (dipole) transition operators

Direct *RIXS amplitude @ transition metal L-edge*



	c_1	c_2
L_3	1	1
L_2	$\frac{1}{2}$	-1

Ament, Ghiringhelli, Moretti, Braicovich & JvdB, PRL 103, 117003 (2009)

Marra, Wohlfeld & JvdB, PRL 109, 117401 (2012)

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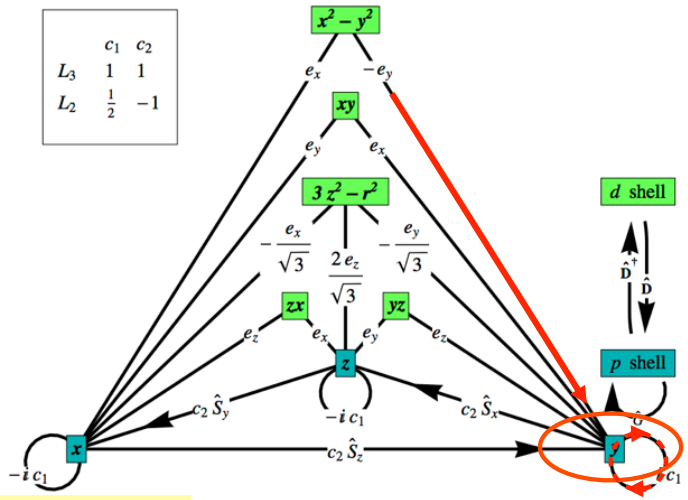

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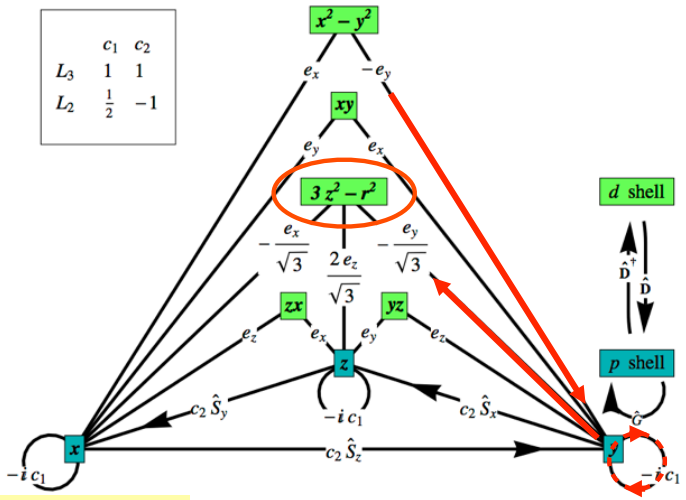

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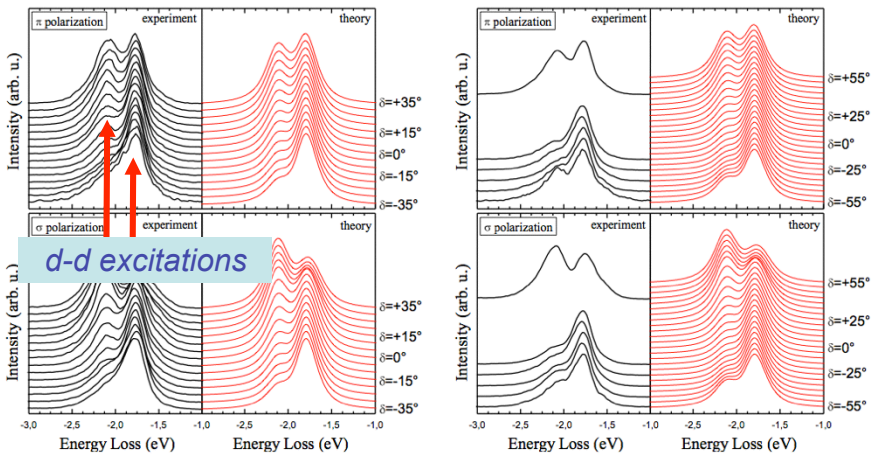
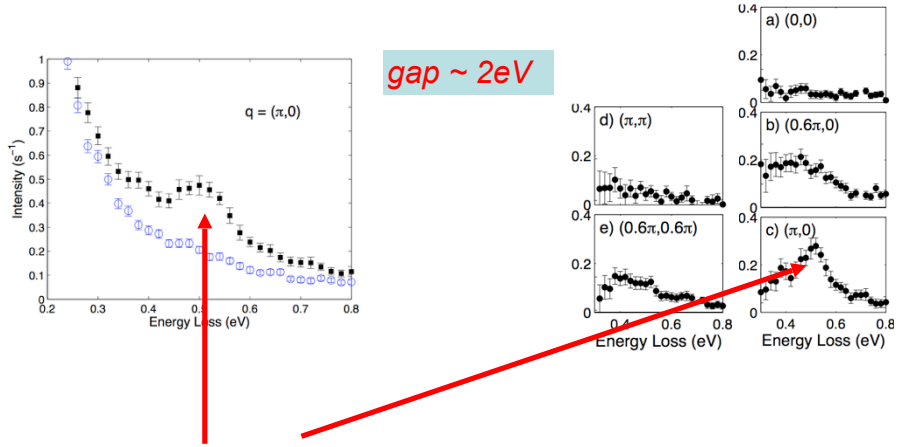
Orbital excitations by direct RIXS on La_2CuO_4



Moretti, Bisogni, Aruta, Balestrino, Berger, Brookes, Luca, Castro, Grioni, Guarise, Medaglia, Miletto, Minola, Perna, Radovic, Salluzzo, Schmitt, Zhou, Braicovich & Ghiringhelli, NJP 13, 043026 (2011)

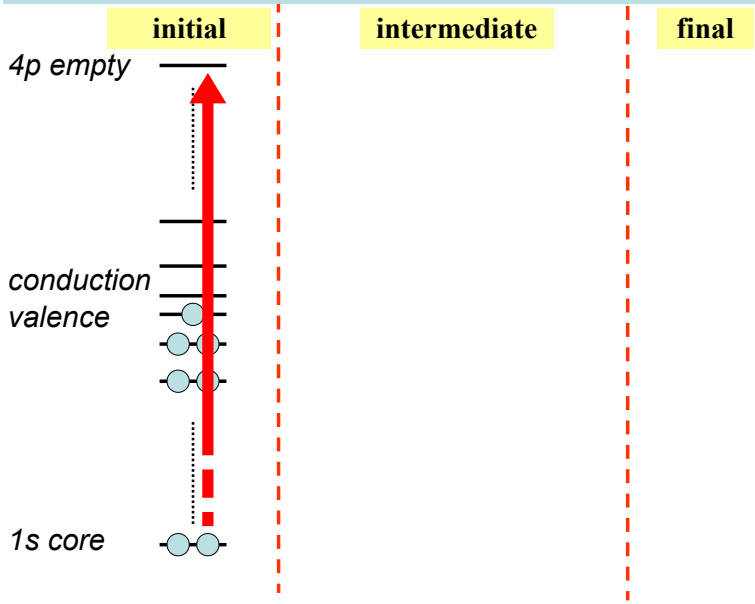
K-edge RIXS on La_2CuO_4



500 meV peak! Phonons?
d-d excitation?
Magnons?

J. Hill et al., PRL 100, 097001 (2008)

Indirect *Resonant Inelastic scattering mechanism*



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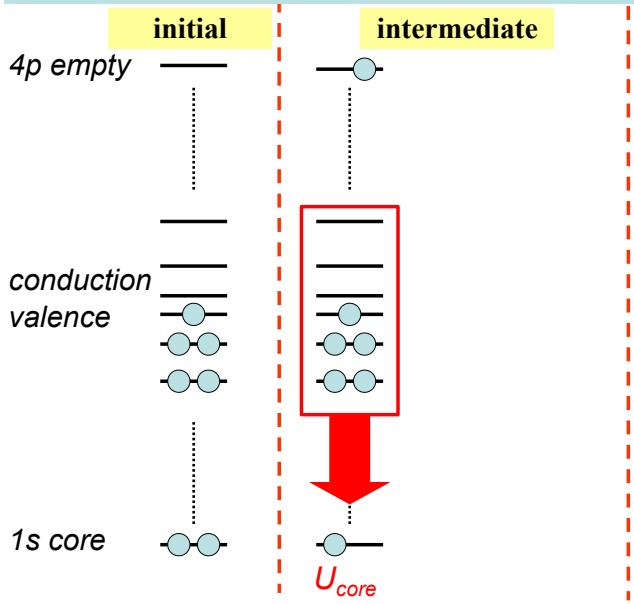

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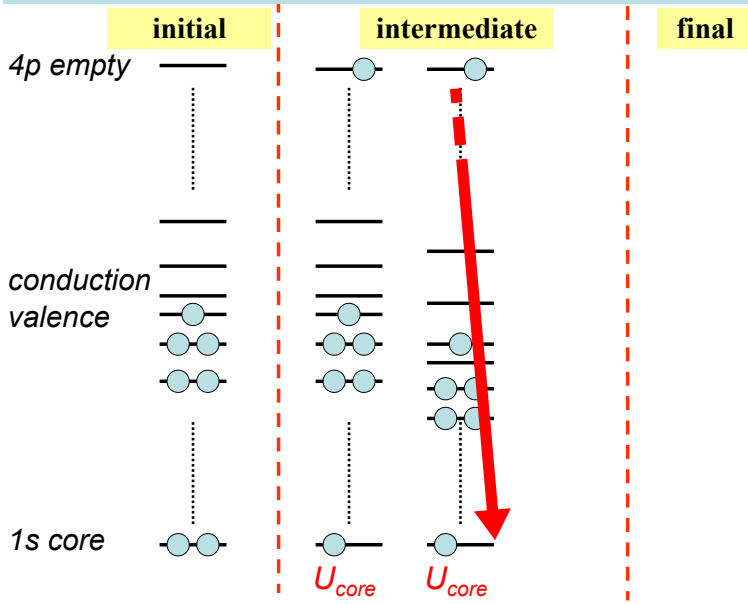

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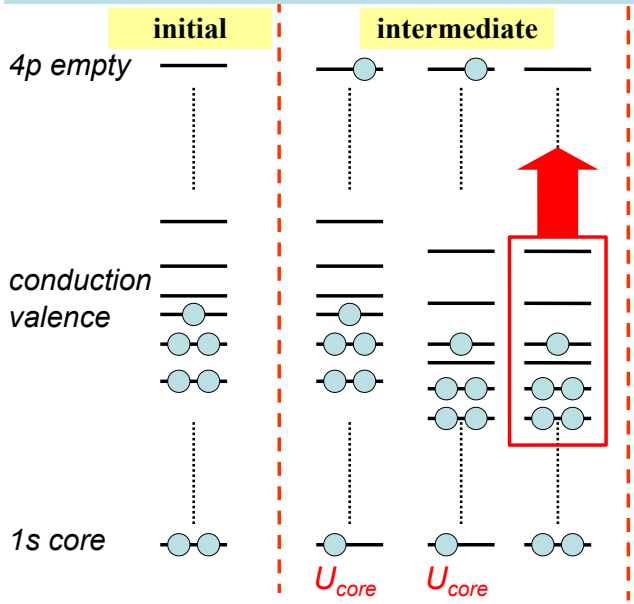

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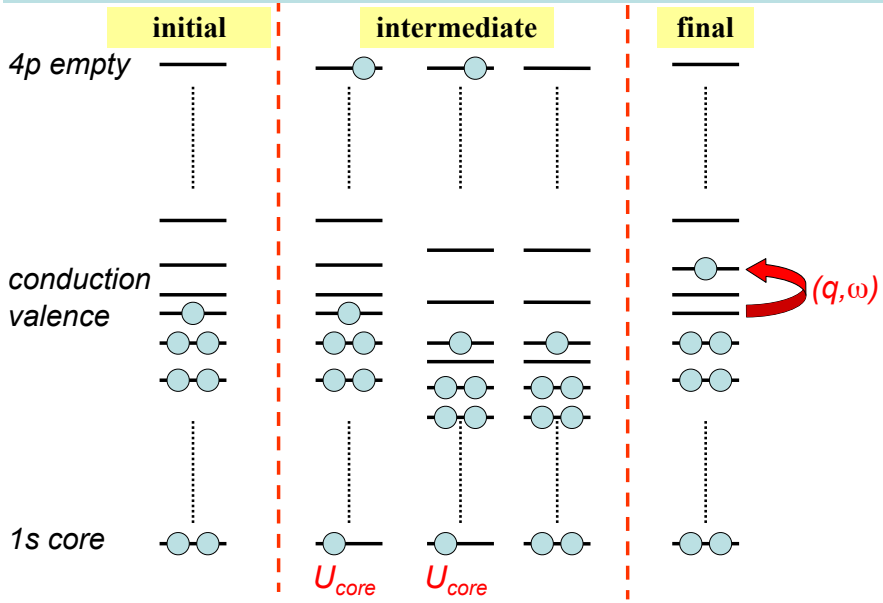

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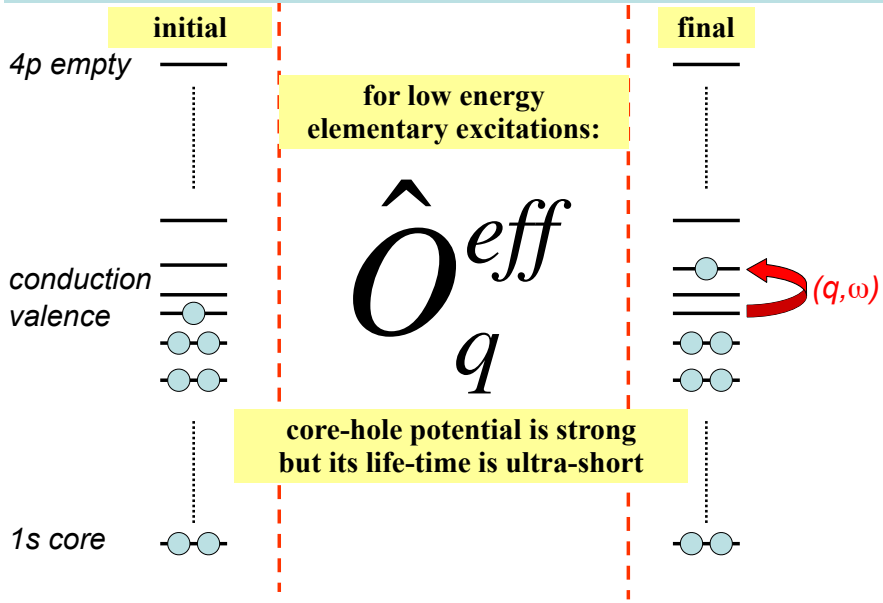


Theory: Kramers-Heisenberg

Fermi's Golden Rule

Scattering amplitude: A_{fi}

Scattering intensity: $I_{scatter}(q,\omega) = \sum_f |A_{fi}|^2 \delta(\omega - \omega_{fi})$

RIXS amplitude: $A_{fi} = \sum_n \frac{\langle f|\overline{D}|n\rangle\langle n|\overline{D}|i\rangle}{\omega_{in} - E_n - i\Gamma}$ → Core-hole life-time

H. Kramers and W. Heisenberg, Z. Phys. 31, 681 (1925)

Theory: Ultra-short Corehole Life-time Expansion

core-hole life time is ultra-short: ~ femto sec

core-hole broadening Γ is large: ~1-2 eV K-edge; ~0.5 eV L-edge

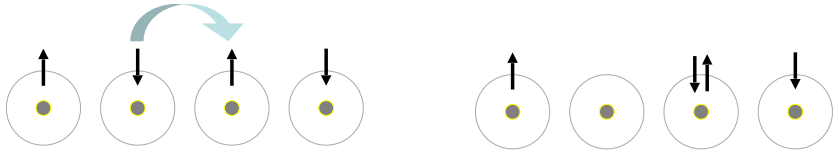
suggests expansion parameter:

$$\frac{1}{\omega_{in} - \omega_{res} - i\Gamma} = \frac{1}{\Delta} \quad \text{with} \quad \Delta = \omega_{in} - \omega_{res} - i\Gamma$$

JvdB & Veenendaal, J. Phys. Chem. Solids 66 2145 (2005)

Spin-photon coupling for Cu K-edge RIXS

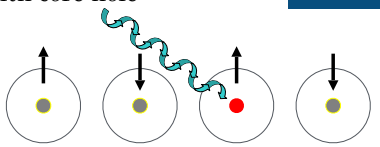
How does intermediate state core-hole couple to spins?
Go back to Hubbard model



Hopping amplitude t Coulomb repulsion U

$$\Delta E = U \Rightarrow J = \frac{4t^2}{U}$$

with core hole:



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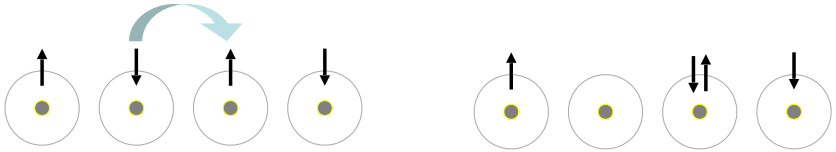

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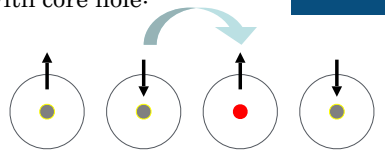
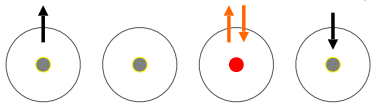


intermediate state at $U - U_c$



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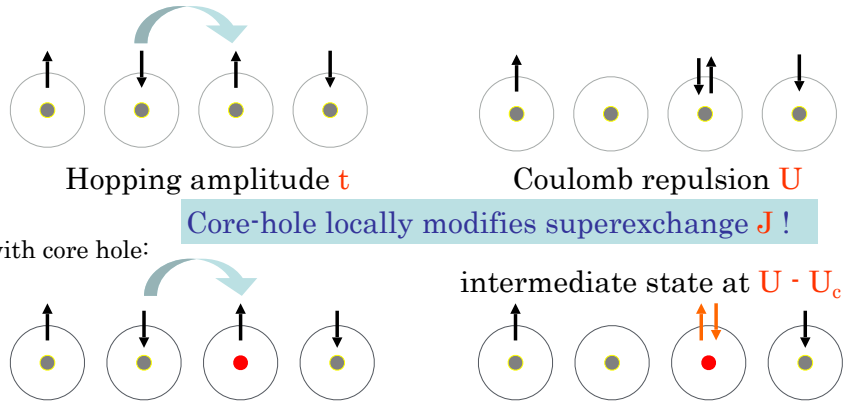


Core-hole locally modifies superexchange J !

K-edge RIXS on La_2CuO_4: two-magnon scattering

Intermediate state modification of J leads in first order to

$$A_{fi} = \frac{\omega_{\rm res}}{\Delta^2}\eta \langle f| \sum_{k,q} J_{k+q}\mathbf{S}_k \cdot \mathbf{S}_{-k+q} |i\rangle$$

2-magnon excitations are probed:

$$\propto \eta J \langle f| \sum_{\mathbf{k}} f(\mathbf{k}) \alpha^\dagger_{\mathbf{k}} \alpha^\dagger_{-\mathbf{k}-\mathbf{q}} |0\rangle$$

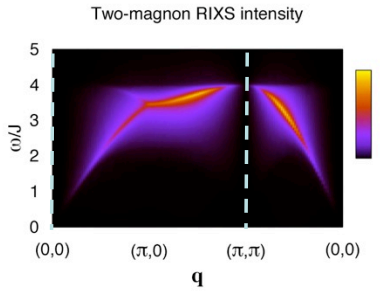
JvdB, EPL 80, 47003 (2007) F. Forte, L. Ament & JvdB, PRB 77, 134428 (2008)

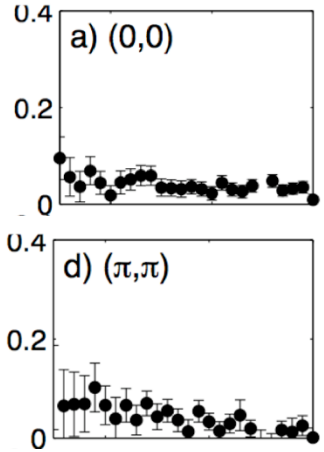
+ magnon-magnon interactions:

T. Nagao, J. Igarashi, PRB 75, 214414 (2007)

Two-magnon scattering: experiment v. theory I

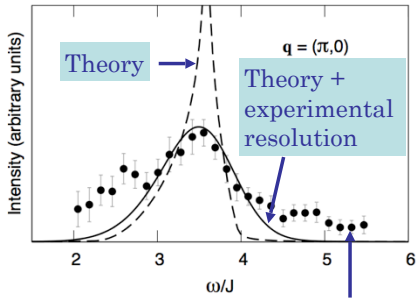
Selection rule: vanishing intensity at q = (0,0) and q = (π,π)





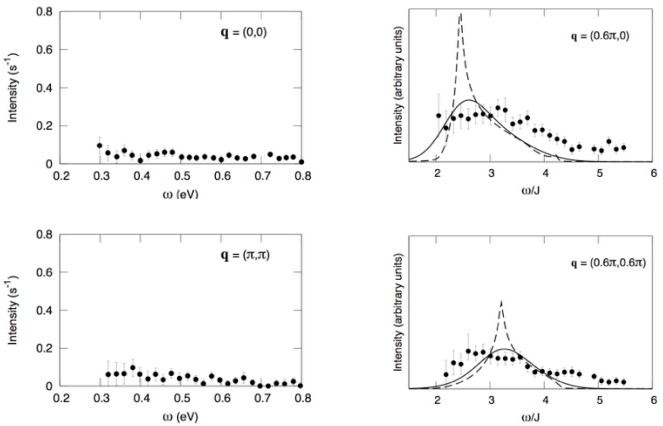
Two-magnon scattering: experiment v. theory II

F. Forte, L. Ament & JvdB, PRB 77, 134428 (2008)



J. Hill et al., PRL 100, 097001 (2008)

Two-magnon scattering: experiment v. theory III

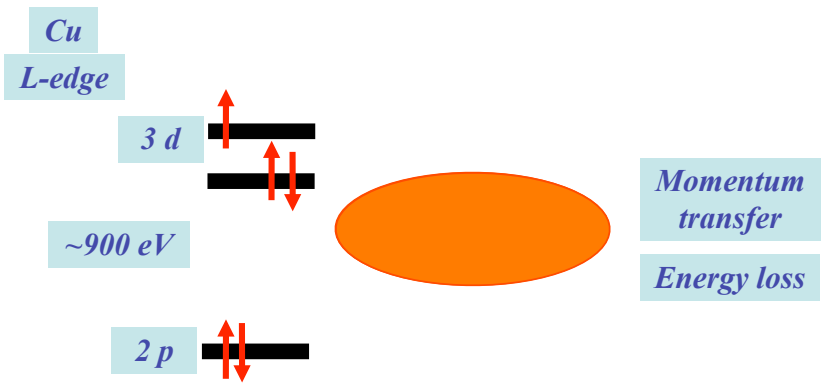


F. Forte, L. Ament & JvdB, PRB 77, 134428 (2008)

Direct Magnetic RIXS

Cu L-edge

Direct RIXS @ TM L-edges



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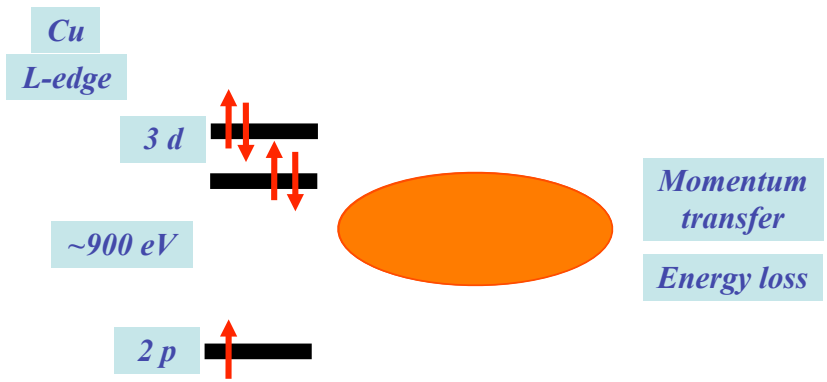

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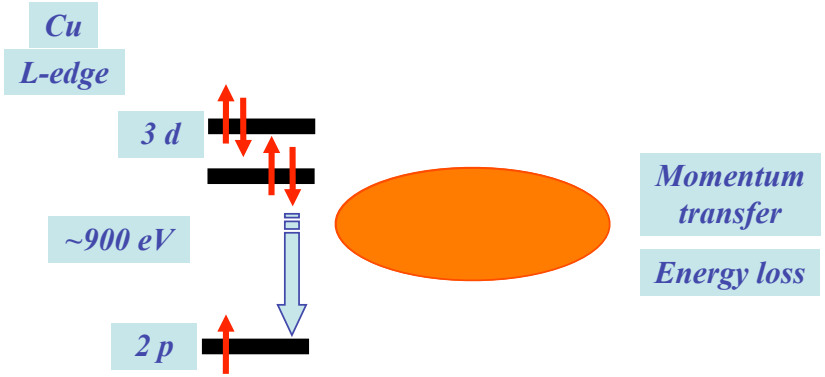

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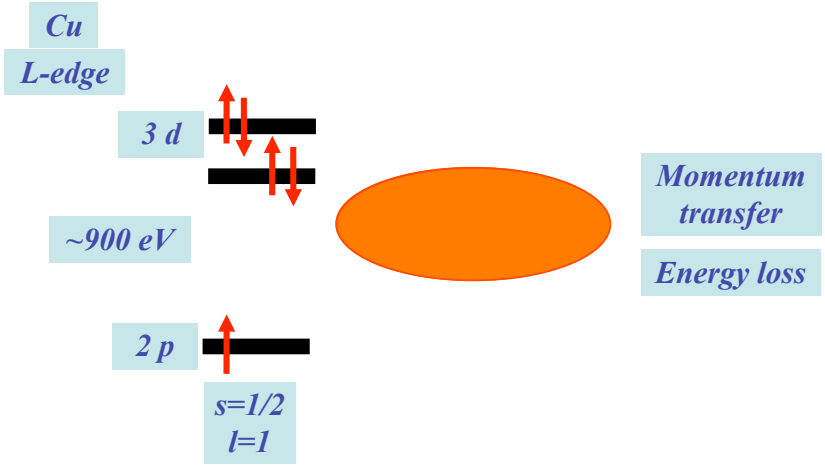

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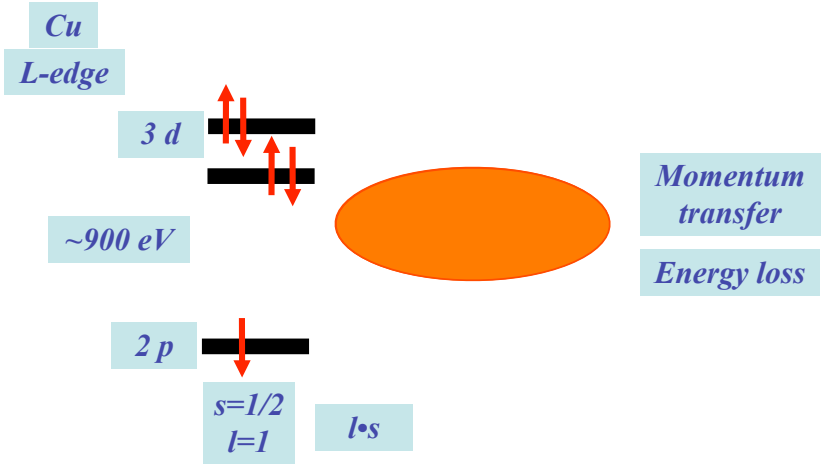

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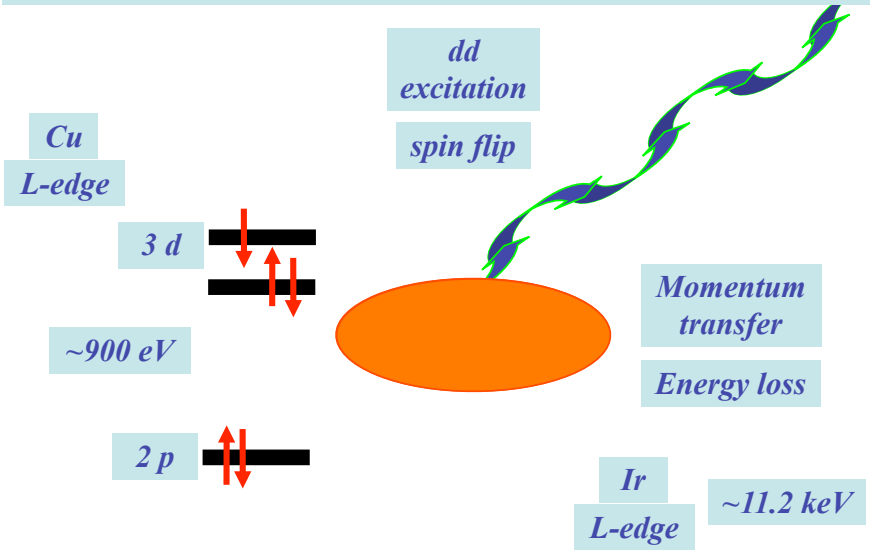
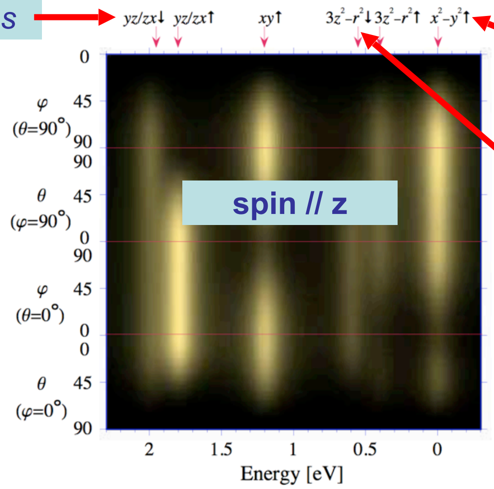


Atomic RIXS on Cu L-edge



final states

ground state

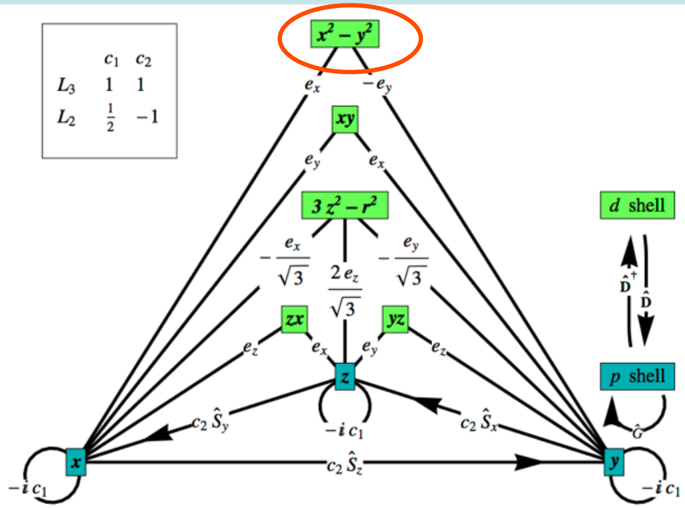
orbital+spin flip

spin // z

no spin flip **without** *orbital flip*

Michel van Veenendaal
PRL 96, 117404 (2006)

RIXS *spin-flip* amplitude @ transition metal L-edge



	c_1	c_2
L_3	1	1
L_2	$\frac{1}{2}$	-1

Ament, Ghiringhelli, Moretti,
Braicovich & JvdB,
PRL 103, 117003 (2009)

Marra, Wohlfeld & JvdB,
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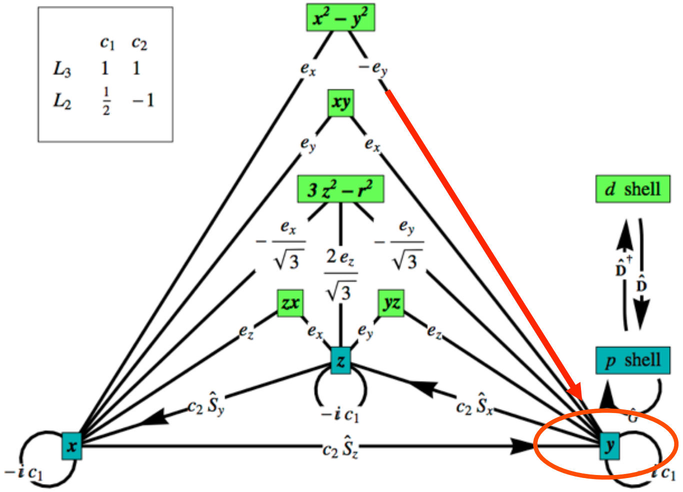

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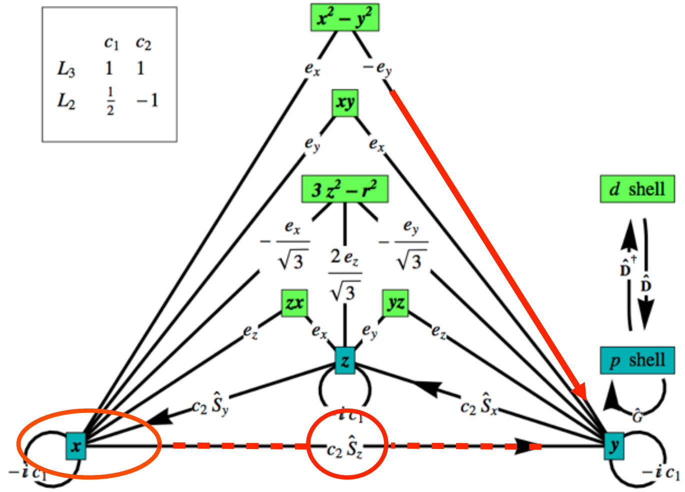

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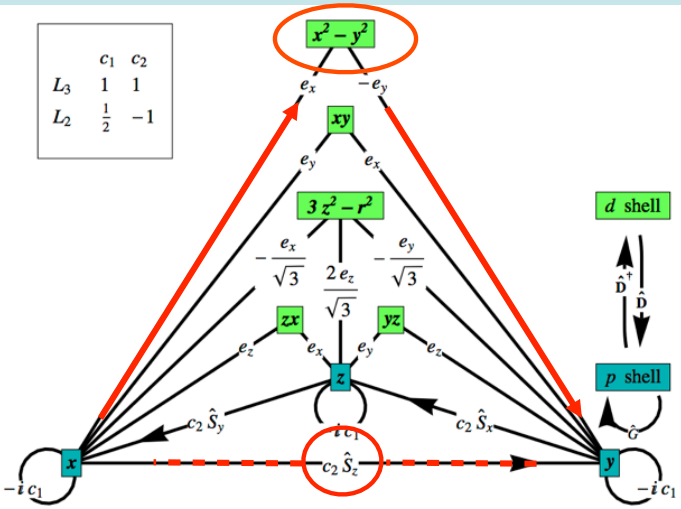


Ament, Ghiringhelli, Moretti, Braicovich & JvdB, PRL 103, 117003 (2009)

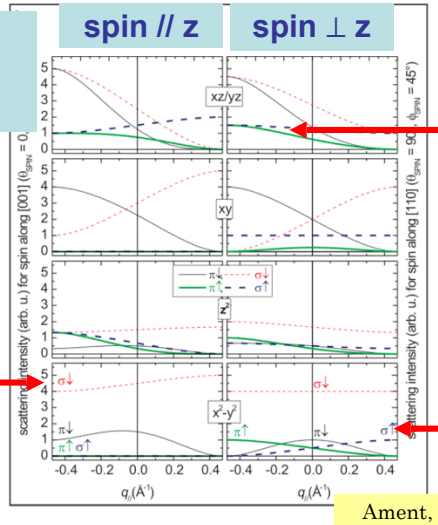
x^2-y^2 spin *NOT* // z: *pure spin flip*

Marra, Wohlfeld & JvdB, PRL 109, 117401 (2012)

Magnetic RIXS on Cu L-edge

⇓ hole in x^2-y^2 orbital

spin // z | **spin ⊥ z**



orbital+spin flip

ground state

spin flip **only**

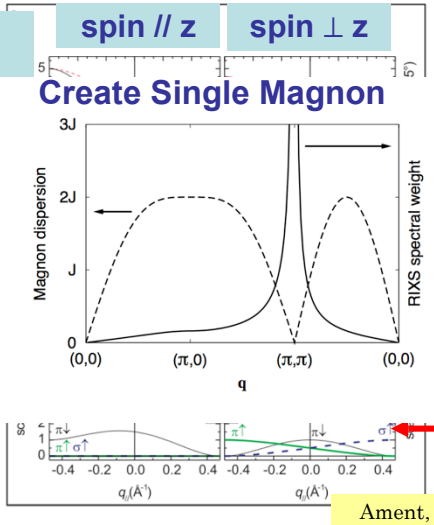
Ament, Ghiringhelli, Moretti, Braicovich & JvdB, PRL 103, 117003 (2009)

Magnetic RIXS on Cu L-edge

⇓ hole in x^2-y^2 orbital

spin // z | **spin ⊥ z**

Create Single Magnon



orbital+spin flip

ground state

spin flip **only**

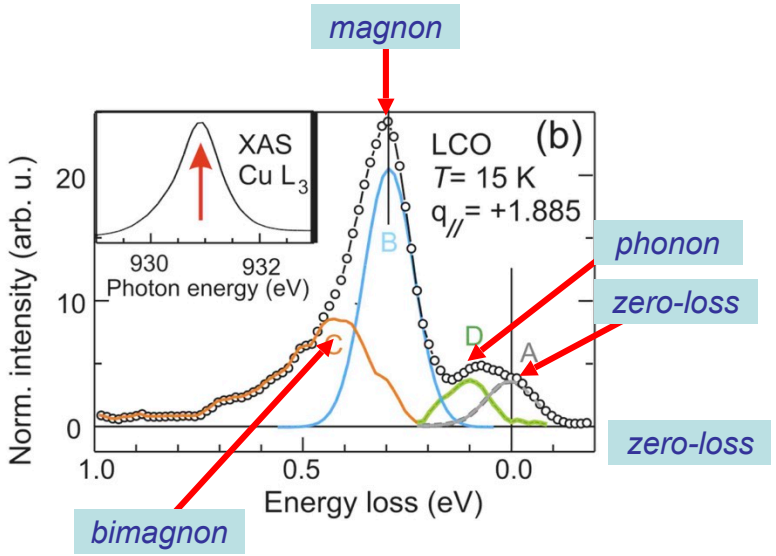
Ament, Ghiringhelli, Moretti, Braicovich & JvdB, PRL 103, 117003 (2009)

Magnetic RIXS on La_2CuO_4 @ Cu L-edge

In special cases direct spin-flip scattering is allowed at Cu L-edge

CuO's are such special cases...

High resolution Cu L-edge RIXS spectrum



Magnetic L-edge RIXS on La_2CuO_4

In special cases direct spin-flip scattering is allowed

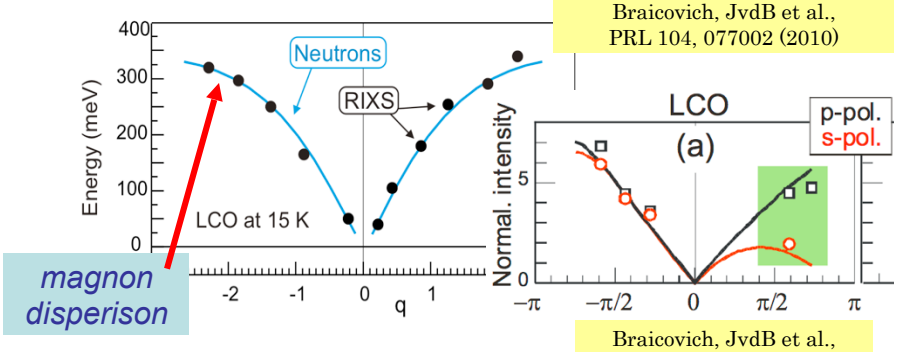
Magnetic L-edge RIXS on La_2CuO_4

In general: *Single spin-flip amplitude related to circular magnetic dichroism in absorption*

CuO's are such cases!

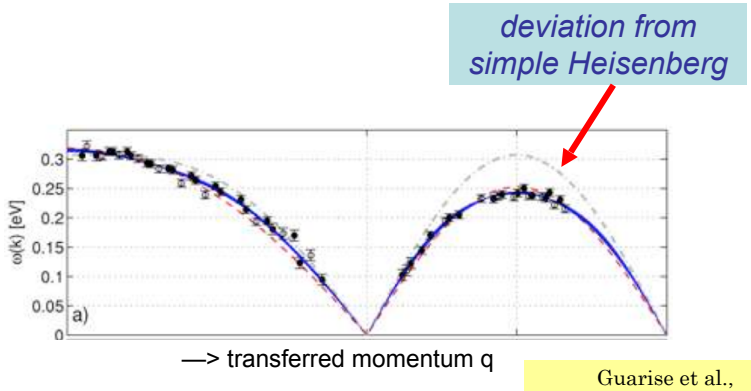
Haverkort
PRL 105, 167404 (2010)

Braicovich, JvdB et al.,
PRL 104, 077002 (2010)



Braicovich, JvdB et al.,
PRB 81, 174533 (2010)

RIXS magnon dispersion of $Sr_2CuO_2Cl_2$



Guarise et al.,
PRL 105, 157006 (2010)

Magnetic RIXS vs. Inelastic Neutron Scattering

	RIXS	Neutrons
amount of material needed	small	large
magnon energy accessible	high (>10^2 meV)	best low (<10^2 meV)
materials	Cu, Fe ...	non-absorbers

Direct Magnetic RIXS

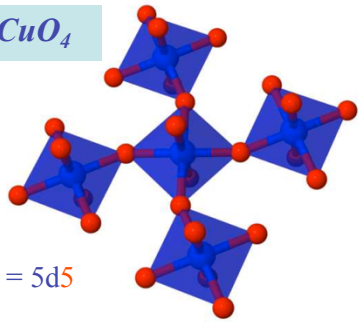
Ir L-edge

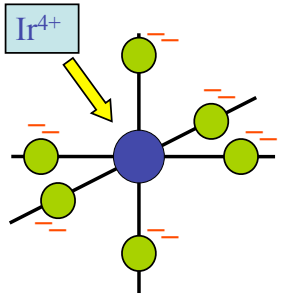
What about magnons in iridates?

Sr_2IrO_4 : equivalent of cuprate La_2CuO_4

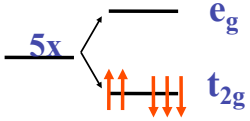
Jackeli & Khaliullin, PRL 102,017205 (2009)

B.J. Kim, Ohsumi, Komesu, Sakai, Morita, Takagi, Arima, Science 323, 1329 (2009)





Ir (4+) = 5d5



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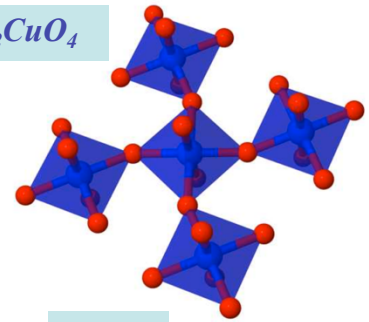
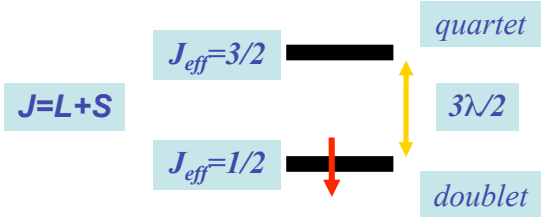
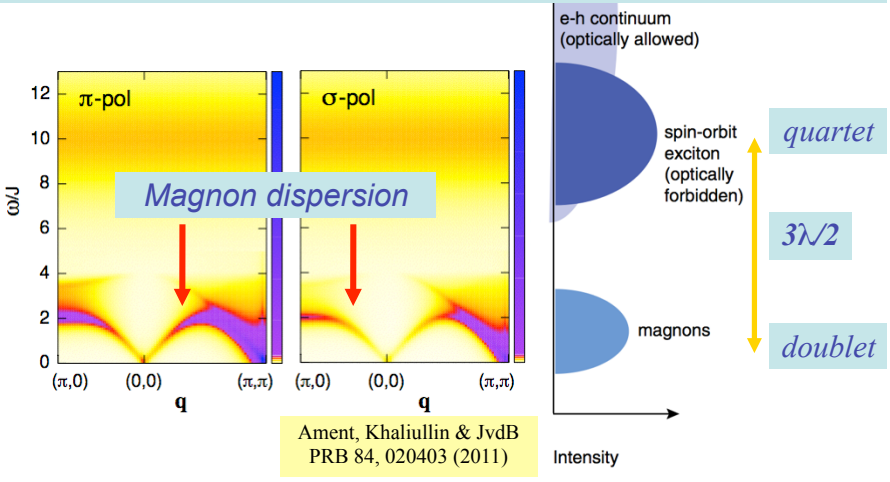


t_{2g}^5: single hole s=1/2 in 3-fold degenerate l=1 state

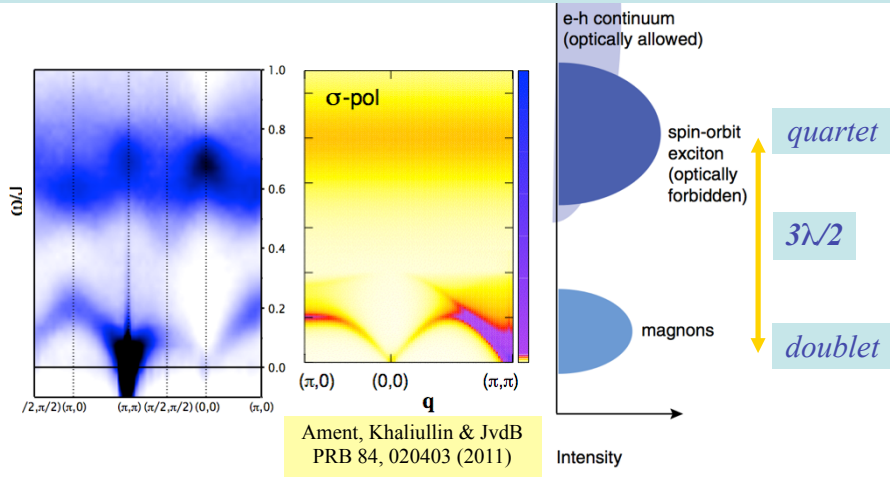


Direct RIXS on Sr_2IrO_4



Ament, Khaliullin & JvdB
PRB 84, 020403 (2011)

Direct RIXS on Sr_2IrO_4



Ament, Khaliullin & JvdB
PRB 84, 020403 (2011)

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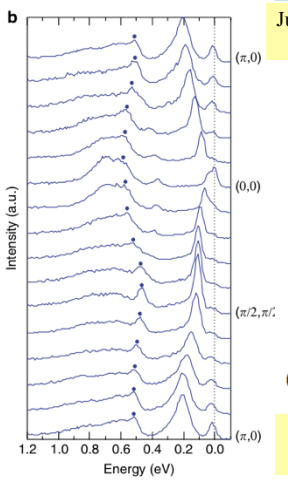
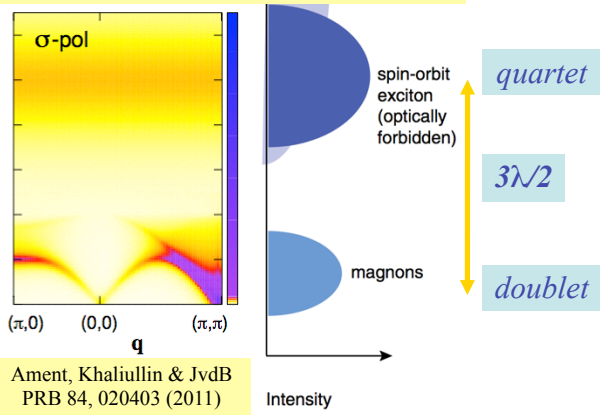
G. Khaliullin,[6] B. J. Kim[3,*] PRL 108, 177003 (2012)

Direct RIXS on Sr_2IrO_4



Jungho Kim, Daghofer, Said, Gog, JvdB, Khaliullin & B.J. Kim, Nat. Comm. 5, 4453 (2014)



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PRB 84, 020403 (2011)

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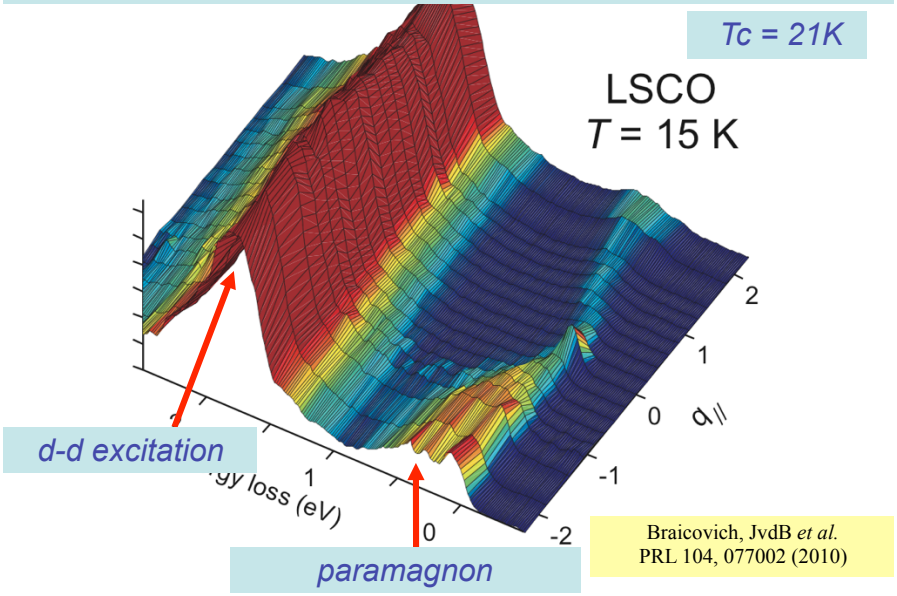
Magnetic RIXS paramagnons

doped quasi-2D Cu-oxides

Magnetic L-edge RIXS on 8% doped $La_{2-x}Sr_xCuO_4$



Braicovich, JvdB *et al.*
PRL 104, 077002 (2010)

nature physics
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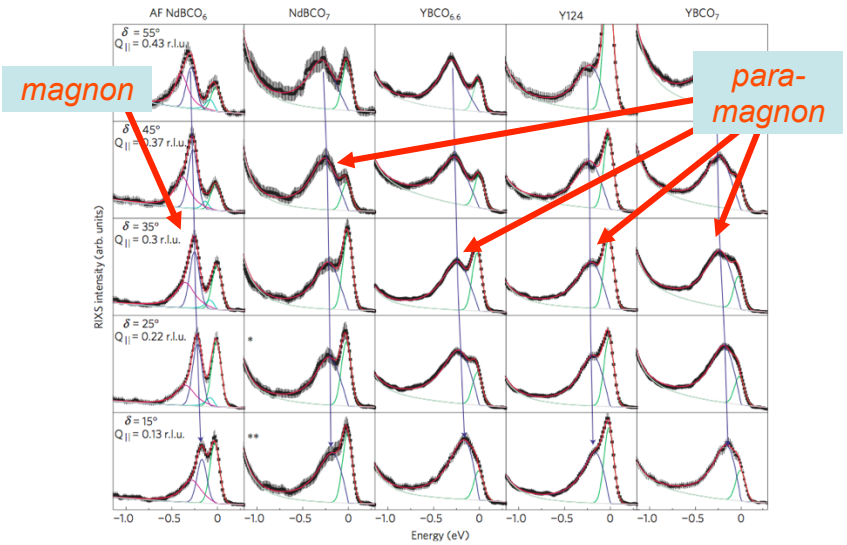
Intense paramagnon excitations in a large family of high-temperature superconductors

M. Le Tacon[1]*, G. Ghiringhelli[2], J. Chaloupka[1], M. Moretti Sala[2], V. Hinkov[1,3], M. W. Haverkort[1], M. Minola[2], M. Bakr[1], K. J. Zhou[4], S. Blanco-Canosa[1], C. Monney[4], Y. T. Song[1], G. L. Sun[1], C. T. Lin[1], G. M. De Luca[5], M. Salluzzo[5], G. Khaliullin[1], T. Schmitt[4], L. Braicovich[2] and B. Keimer[1]*



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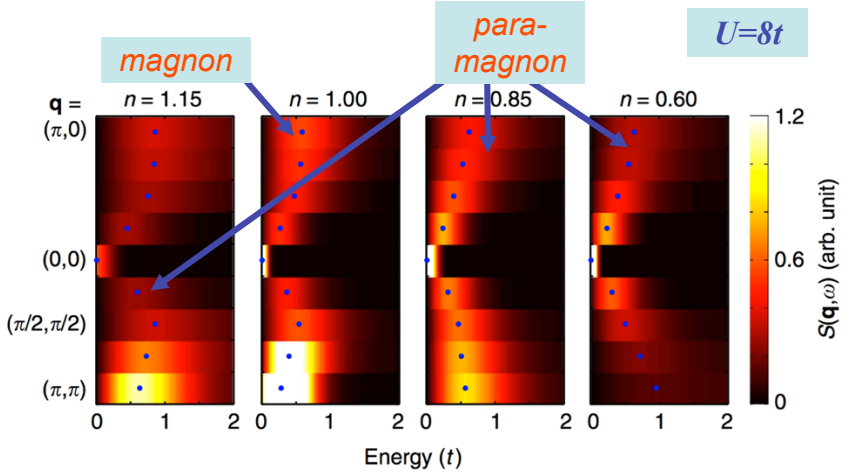
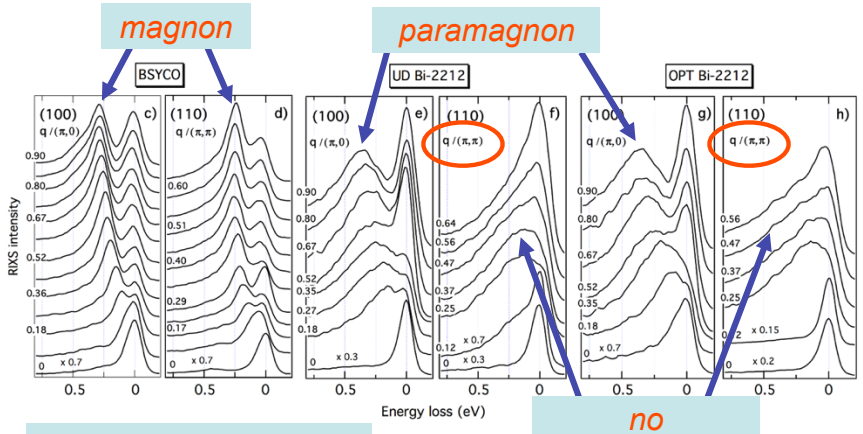
Dynamical structure factor Hubbard model, QMC



Jia, Nowadnick, Wohlfeld, Kung, Chen, Johnston, Tohyama, Moritz & Devereaux
Nat. Comm. 5, 3314 (2014)

RIXS on Bi-2212 cuprate



Related to presence of e-h continuum?

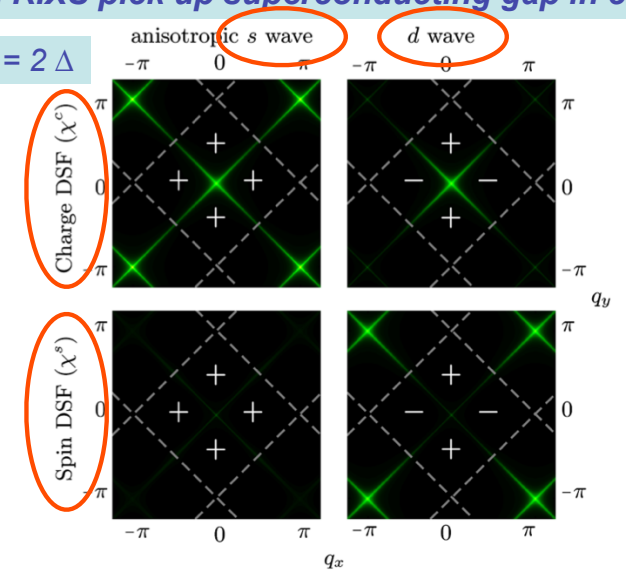
no paramagnon!

Benjamin, Klich & Demler PRL 112, 247002 (2014)

Guarise, Piazza, Berger, Giannini, Schmitt, Rønnow, Sawatzky, JvdB, Altenfeld, Eremin & Grioni, Nat. Comm. 5, 5760 (2014)

Can RIXS pick up superconducting gap in cuprates?

$E_{loss} = 2\,\Delta$



Marra, Sykora, Wohlfeld & JvdB, PRL 110, 117005 (2013)

Summary part 3

- RIXS sensitive to magnetic excitations of e.g.
  low D cuprates, iron pnictides and iridates
- Magnons, spinons and paramagnons are observed
- Dispersion of these modes can be determined
- Observed paramagnons are challenge for theory
- It can reasonably be assumed that the future of
  RIXS is even brighter than its past
- More and better experiments, instruments, theory

End of Lecture III