Ultrashort Lifetime Expansion
for
Resonant Inelastic X-ray Scattering

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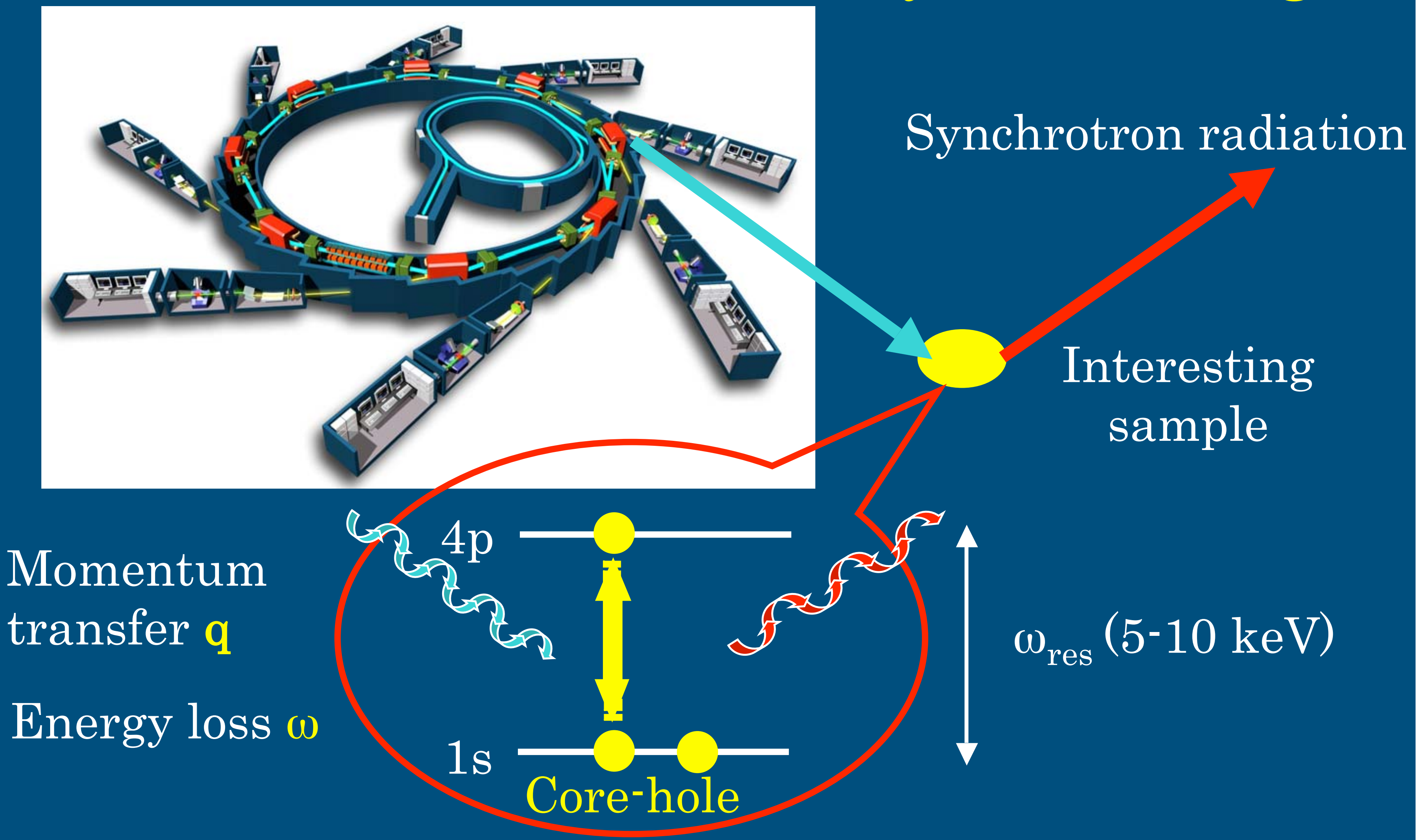
What is RIXS?
Resonant Inelastic X-ray Scattering



Why RIXS?

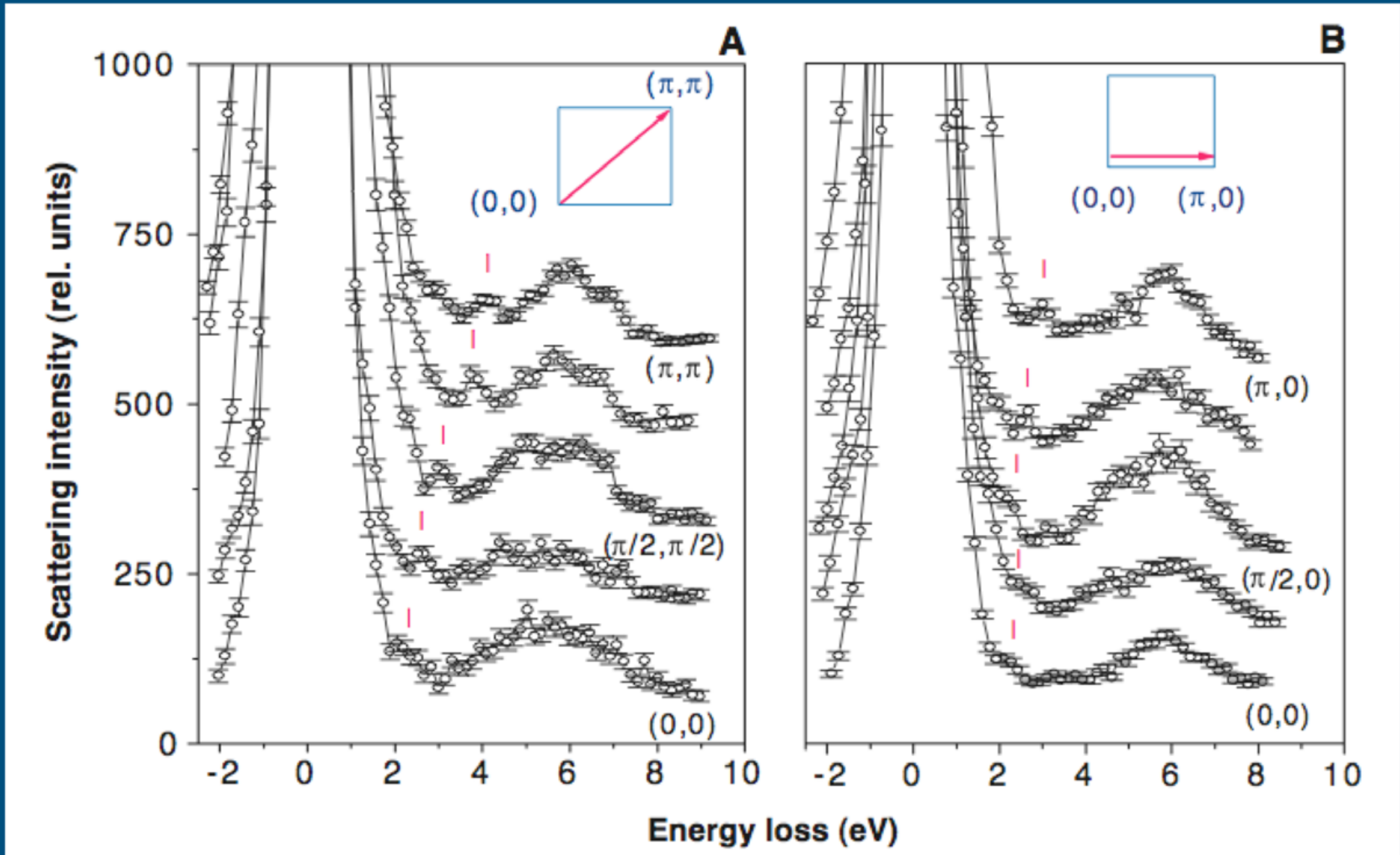
Complementary to Raman and neutrons

- Momentum dependence
- Tune X-rays to different resonances
- Small samples
- Why not?

Outline

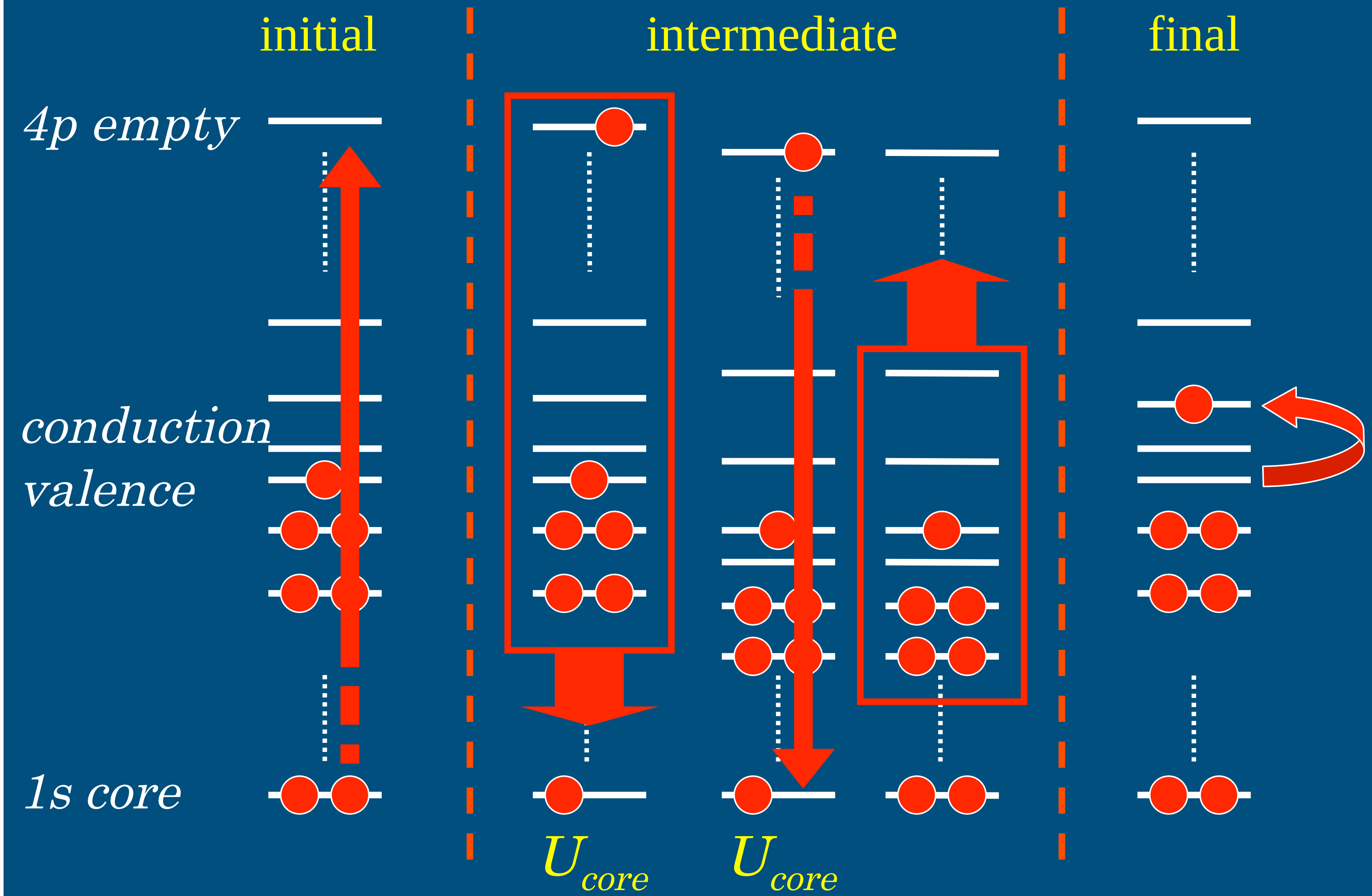
1. Framework: the UCL expansion
   What can one measure?
2. Charge excitations
3. Magnetic RIXS on La_2CuO_4
   Coupling of core-hole to spins
   - Cu K-edge: indirect RIXS
   - Cu L-edge: direct RIXS
4. Orbital RIXS on $LaMnO_3$

Typical RIXS spectra

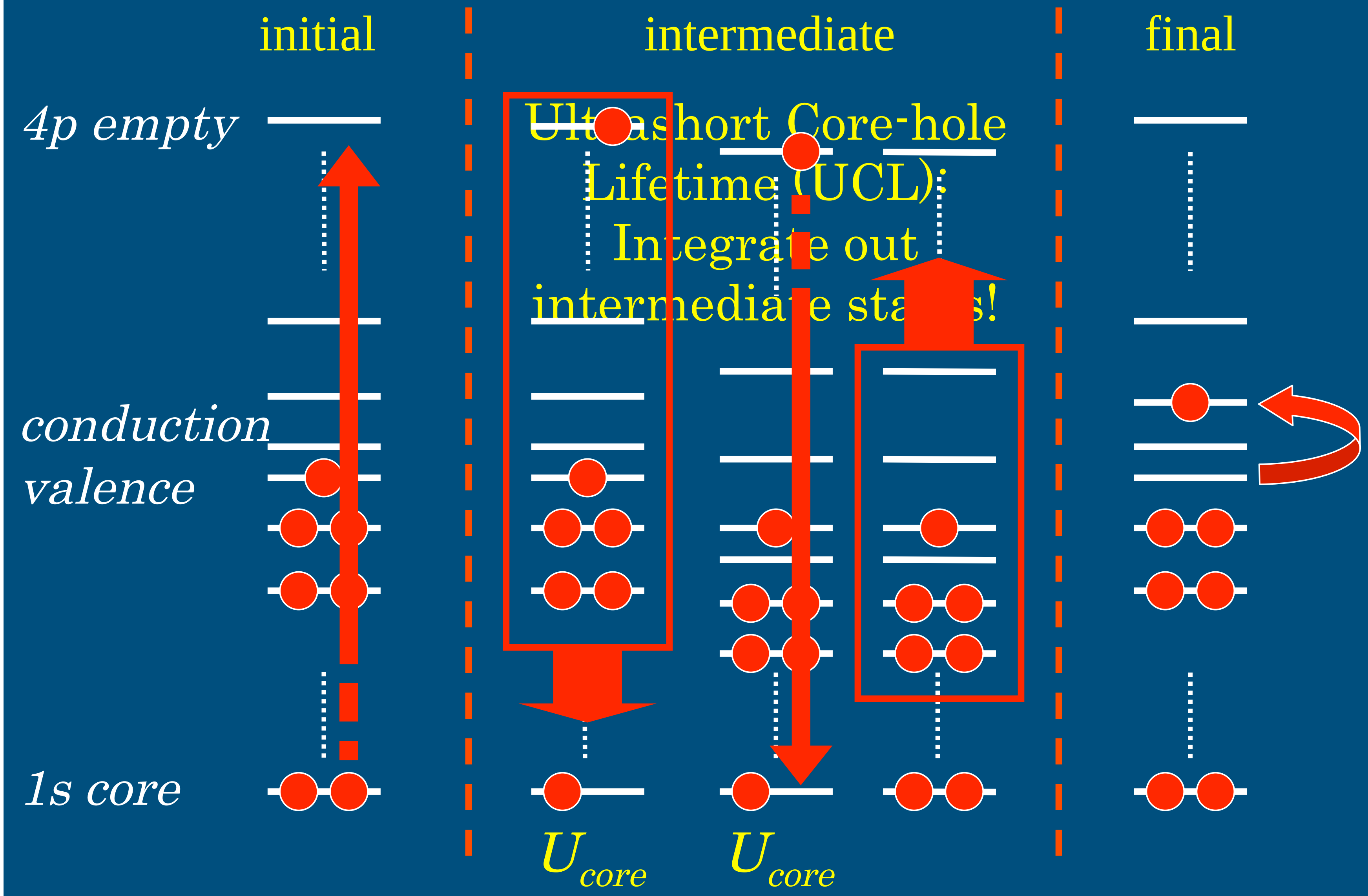


M.Z. Hasan *et al.* Science **288**, 1811 (2000)

What happens in detail



What happens in detail



Theory

- Kramers-Heisenberg relation

$$\left.\frac{d^2\sigma}{d\Omega d\omega}\right|_{\mathrm{res}}(\mathbf{q},\omega) \propto \left\langle \sum_f \left|A_{fi}\right|^2 \delta(\omega-\omega_{fi}) \right\rangle_T$$

with

$$A_{fi} = \omega_{\mathrm{res}} \sum_n \frac{\langle f|\,\hat{O}\,|n\rangle \langle n|\,\hat{O}\,|i\rangle}{\omega_{\mathrm{det}} - E_n - i\Gamma}$$

Detuning from resonance

Intermediate states

Inverse core-hole lifetime

$\Gamma \approx 0.25 - 3\ \mathrm{eV}$

Theory - UCL expansion

Scattering amplitude

$$A_{fi} = \omega_{\text{res}} \sum_n \frac{\langle f|\,\hat{O}\,|n\rangle \langle n|\,\hat{O}\,|i\rangle}{\omega_{\text{det}} - E_n - i\Gamma}$$

$$\Delta := \omega_{\text{det}} - i\Gamma$$

$$= \omega_{\text{res}} \sum_n \langle f|\,\hat{O}\,|n\rangle \langle n|\,\hat{O}\,|i\rangle \frac{1}{\Delta - E_n}$$

Using the Ultrashort Core-hole Lifetime gives

$$= \frac{\omega_{\text{res}}}{\Delta} \sum_n \langle f|\,\hat{O}\,|n\rangle \langle n|\,\hat{O}\,|i\rangle \frac{1}{1 - E_n/\Delta}$$

$$= \frac{\omega_{\text{res}}}{\Delta} \sum_n \sum_l \langle f|\,\hat{O}\,|n\rangle (E_n/\Delta)^l \langle n|\,\hat{O}\,|i\rangle$$

$$= \frac{\omega_{\text{res}}}{\Delta} \sum_l \frac{1}{\Delta^l} \langle f|\,\hat{O} H_{\text{int}}^l \hat{O}\,|i\rangle$$

What does RIXS measure?

We obtain $$A_{fi} = \frac{\omega_{\text{res}}}{\Delta} \sum_{l=0}^{\infty} \frac{1}{\Delta^l} \langle f | \hat{O} H^l_{\text{int}} \hat{O} | i \rangle$$

Intermediate state Hamiltonian

$$H_{\text{int}} = H_0 + H'$$

Valence electrons → Arbitrary range hopping & Coulomb interactions

Interaction of valence electrons with core-hole → Assume immobile core-hole, and local core-hole potential

At T = 0, approximate H^l_{int} for both strong and weak potentials

J. van den Brink & M. van Veenendaal, EPL **73**, 121 (2006)
L. Ament, F. Forte & J. van den Brink, PRB **75**, 115118 (2007)

What does RIXS measure?

Spinless fermions: $A_{fi} = P(\omega) \langle f | \rho_{\mathbf{q}} | 0 \rangle$

Charge density

with $P(\omega) = \dfrac{\omega_{\text{res}} U_c}{(\Delta - U_c)(\Delta - \omega)}$

Resonant prefactor

$$\frac{d^2\sigma}{d\omega d\Omega} \propto |P(\omega)|^2 S(\mathbf{q}, \omega)$$

Fermions with spin:

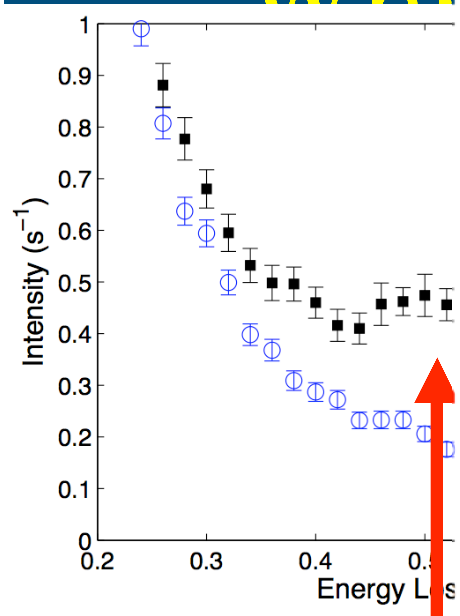
$$A_{fi} = P_1(\omega) \langle f | \rho_{\mathbf{q}} | i \rangle + P_2(\omega) \langle f | \mathbf{S}^2_{\mathbf{q}} | i \rangle$$

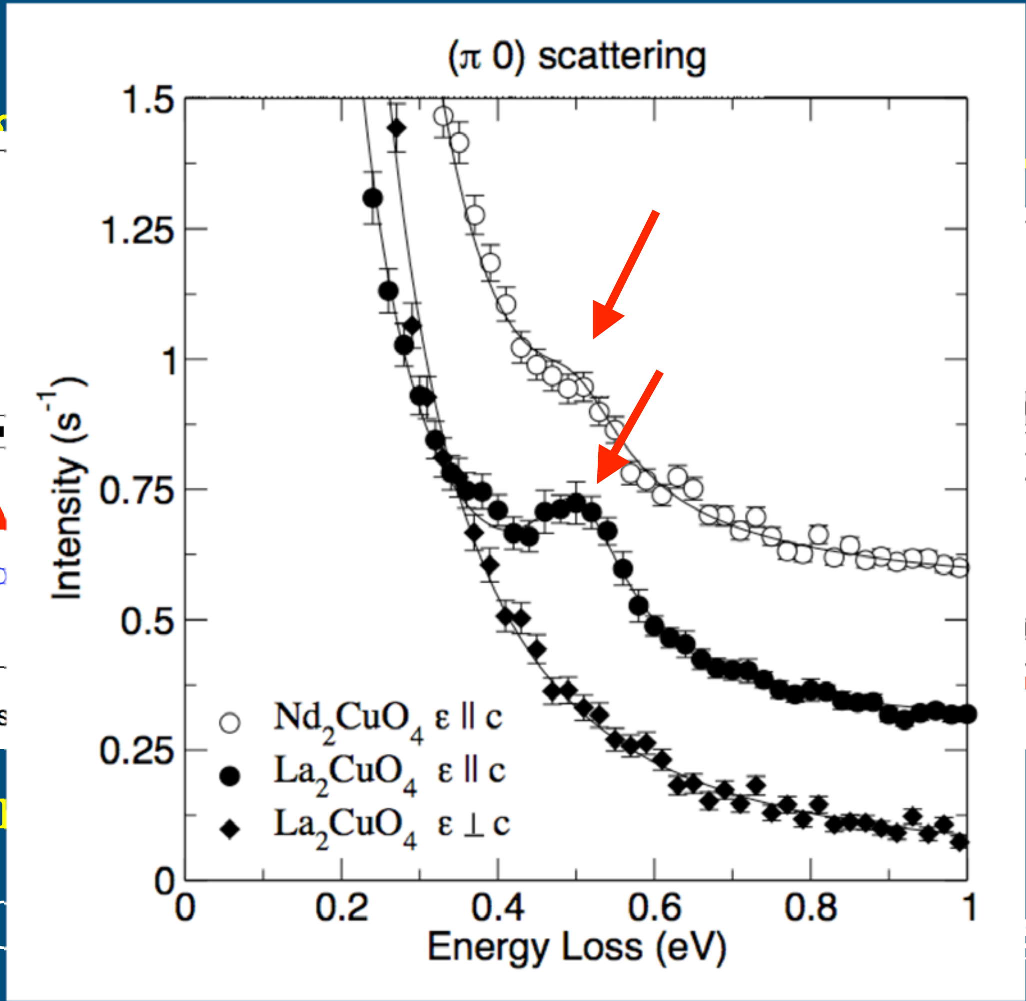
Charge density

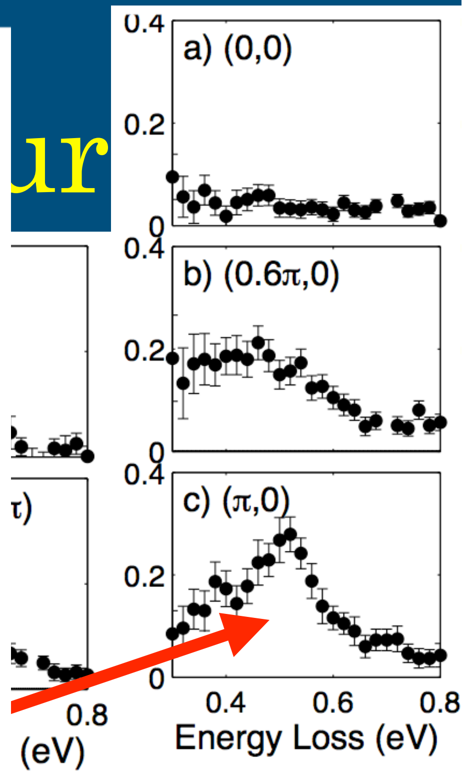
Longitudinal spin density

J. van den Brink & M. van Veenendaal, EPL **73**, 121 (2006)
L. Ament, F. Forte & J. van den Brink, PRB **75**, 115118 (2007)

Wh... ur







Can we p... ones?

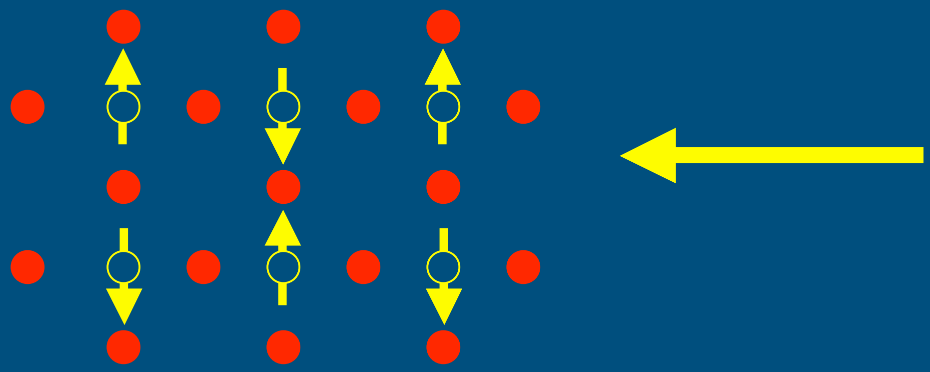
Data on ... gap (~ 2 eV)

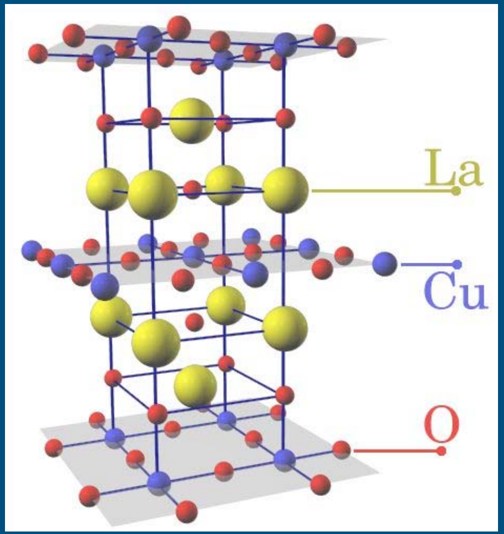
500 meV peak! Phonons?
d-d excitation? Nd_2CuO_4: no shift
Magnons?

J. van den Brink & M. van Veenendaal, EPL **73**, 121 (2006)
L. Ament, F. Forte & J. van den Brink, PRB **75**, 115118 (2007)
J. Hill *et al.*, arXiv: 0709.3274

Magnetic RIXS on La_2CuO_4

Perovskite layers of CuO_2:





Cu 3d e_g holes at low energy:

Single band Hubbard model $\rightarrow H_0 = J \sum_{<i,j>} \mathbf{S}_i \cdot \mathbf{S}_j$

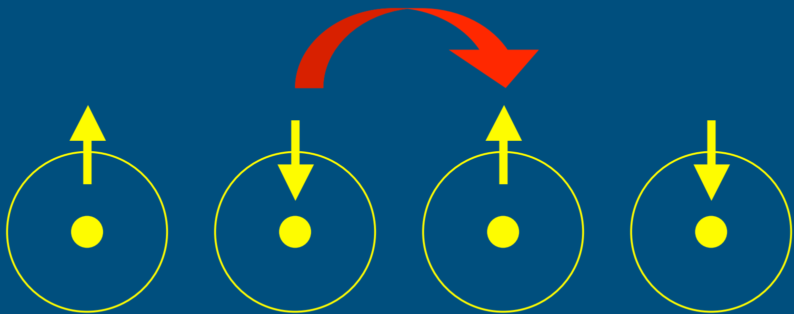
Ground state is antiferromagnetically ordered

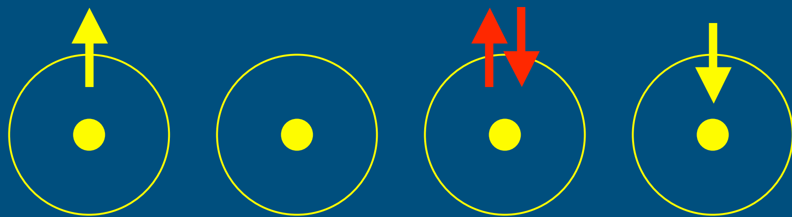
Collective excitations: magnons

Magnetic RIXS on La_2CuO_4

How does the core-hole couple to the spins?

Go back to Hubbard model!



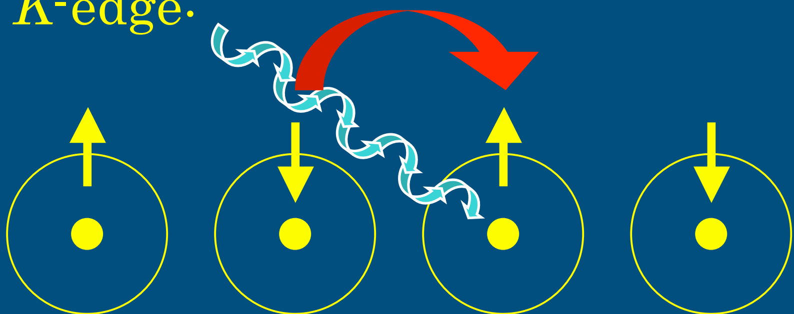


Hopping amplitude t

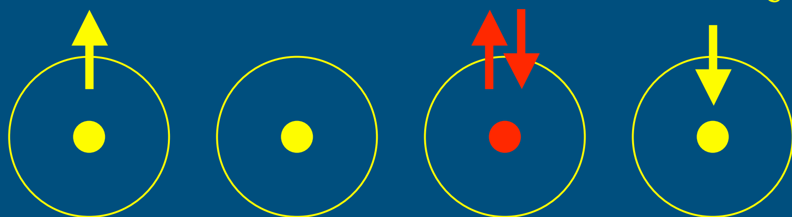
Coulomb repulsion U

Core-hole locally modifies superexchange!

K-edge:



Coulomb repulsion $U - U_c$



Magnetic RIXS on La_2CuO_4

How does the core-hole couple to the spins?

Indirect RIXS locally modifies superexchange!

To first order in UCL:

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

$$\frac{d^2\sigma}{d\omega d\Omega} \propto |A_{fi}|^2 \Rightarrow$$

4-spin correlation function

At T = 0, 2-magnon excitations are probed:

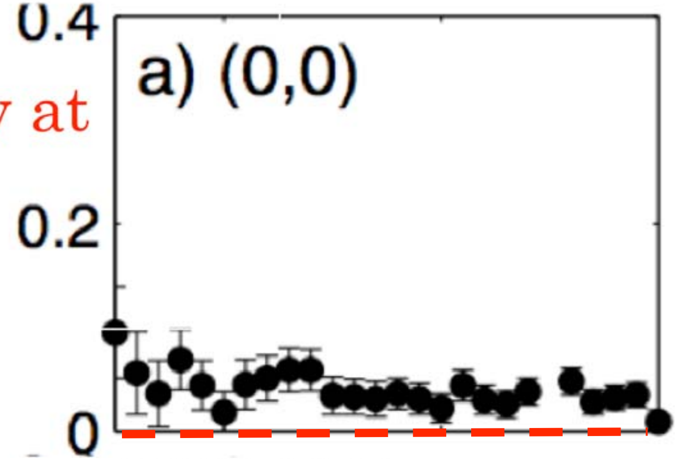
$$\propto \eta J \langle f| \sum_{\mathbf{k}} f(\mathbf{k}) \alpha^\dagger_{\mathbf{k}} \alpha^\dagger_{-\mathbf{k}-\mathbf{q}} |0\rangle$$

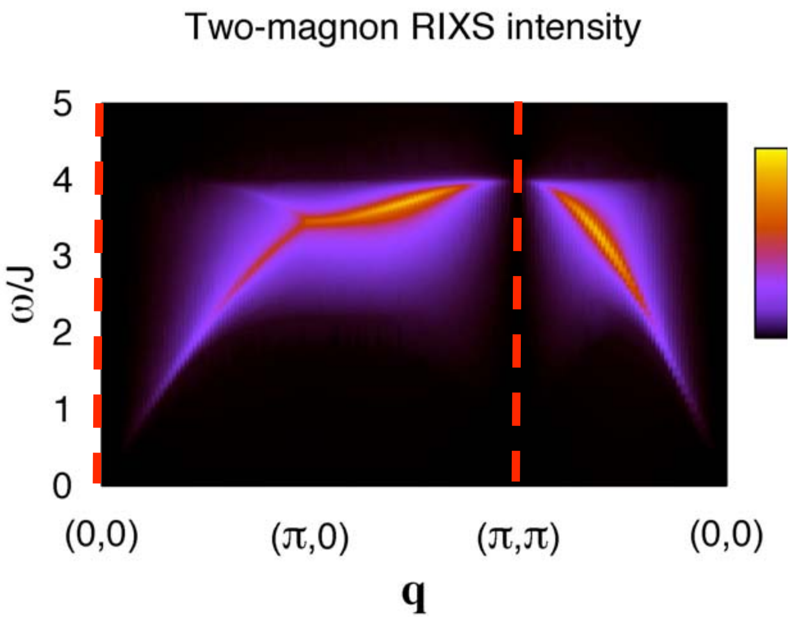
J. van den Brink, cond-mat/0510140

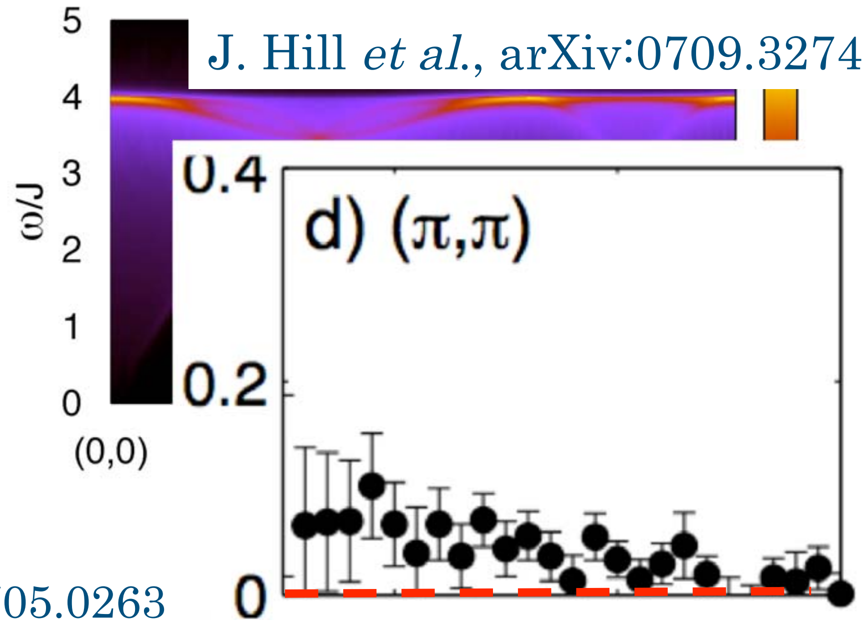
F. Forte, L. Ament & J. van den Brink, arXiv:0705.0263

Magnetic RIXS on La_2CuO_4

Selection rule: vanishing intensity at
$\mathbf{q} = (0,0)$ and $\mathbf{q} = (\pi,\pi)$







J. Hill *et al.*, arXiv:0709.3274

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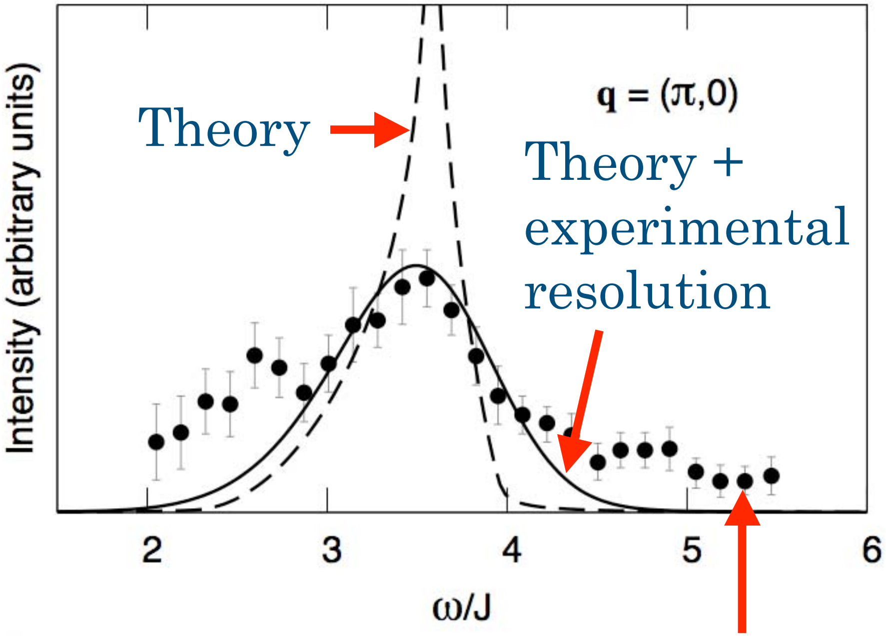
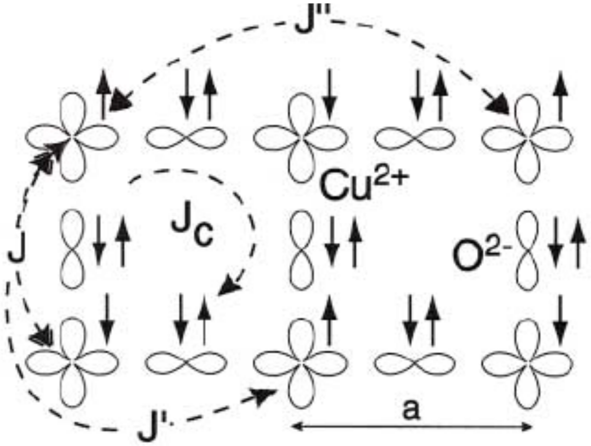


J. Hill *et al.*, arXiv:0709.3274

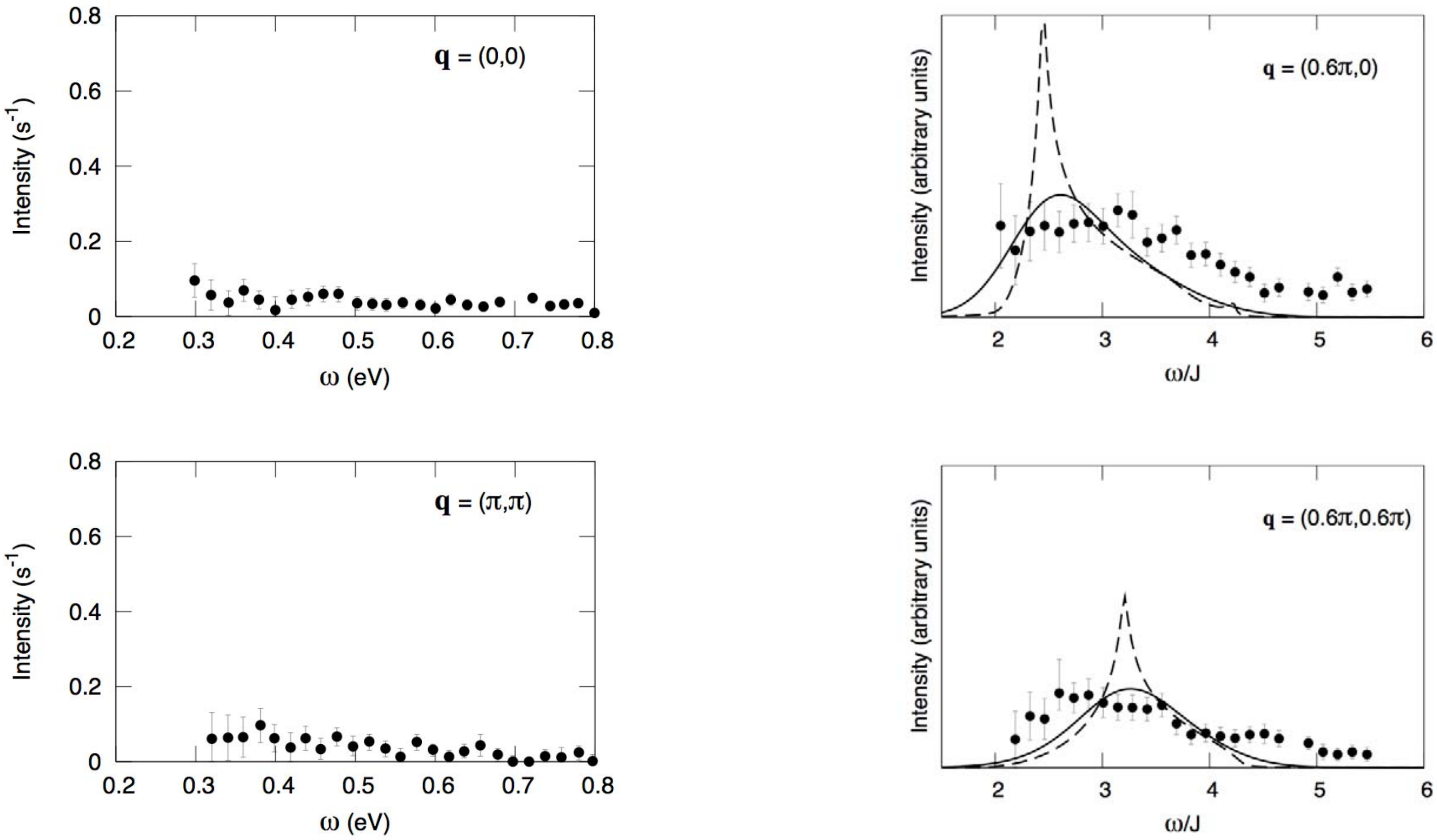
Improve theory with longer range interactions:



$J = 146.3$ meV, $J' = J'' = 2$ meV, and $J_c = 61$ meV

Values from neutron scattering.
Coldea *et al.*, PRL **86**, 5377 (2001)

Magnetic RIXS on La_2CuO_4



F. Forte, L. Ament & J. van den Brink, arXiv:0705.0263
J. Hill *et al.*, arXiv:0709.3274

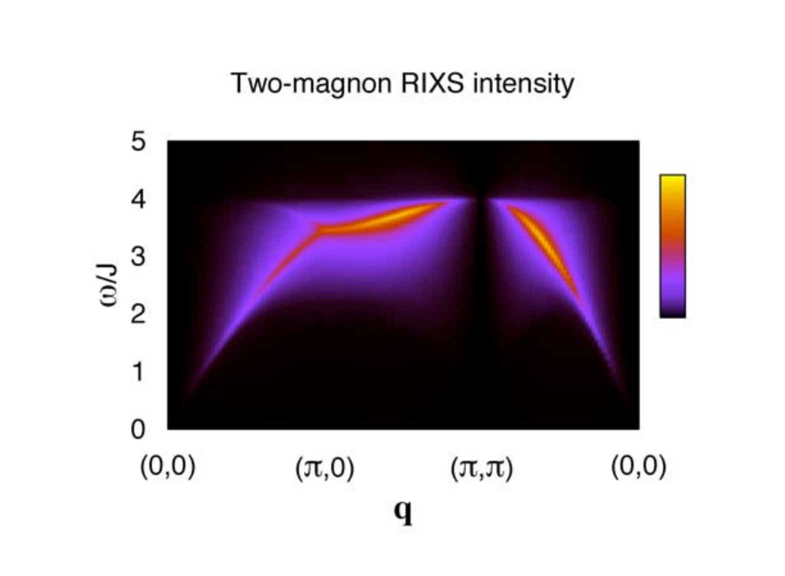
Magnetic RIXS on La_2CuO_4

L-edge: direct RIXS - photoexcited electron in 3d-shell.
G. Ghiringhelli *et al.* are measuring this right now!

Sneak preview:

1. Dispersive feature similar to *K*-edge RIXS, becoming elastic in the low-q limit.
2. Another feature appearing at
3. This feature disappears off-re

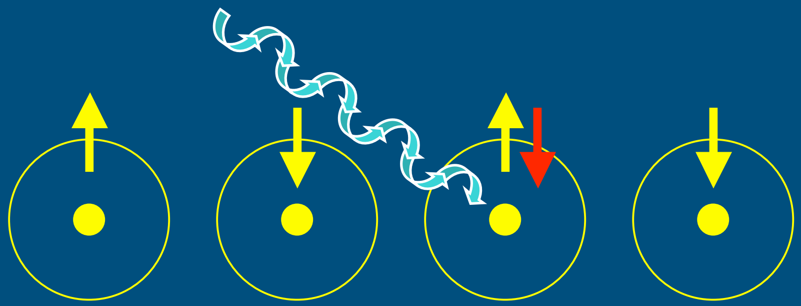
What about the th

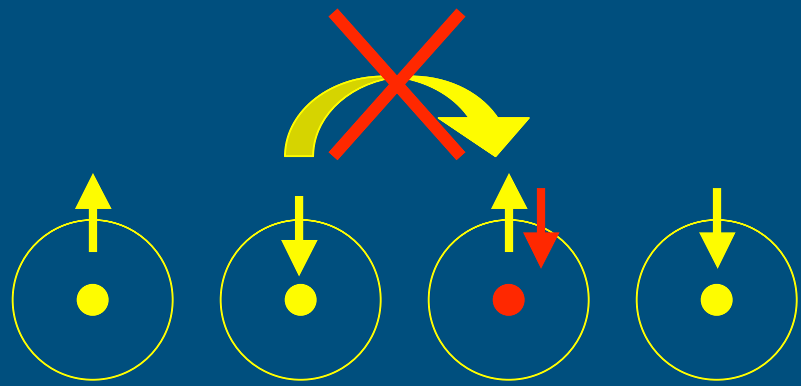


Magnetic RIXS on La_2CuO_4

L-edge: direct RIXS - core-electron ends up in 3d-state

Back to Hubbard model!





$$J' = 0$$

The extra 3d-electron locally blocks superexchange

The UCL results for the K-edge and L-edge are the same!

Magnetic RIXS on La_2CuO_4

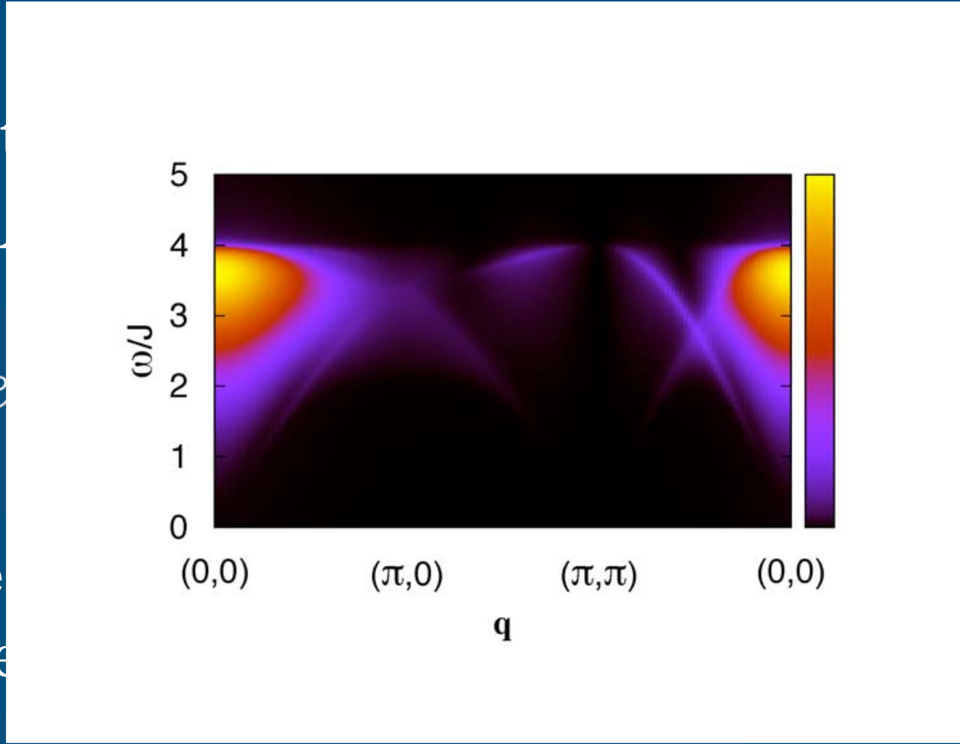
L-edge has more intensity t

Can we go beyond the UCL

$$A^{(1)}_{fi} \propto \frac{\omega_{\text{res}} J}{\Delta^2} \langle f | \sum_{i,j} e$$

Reminder
$\Delta := \omega_{\text{in}} - i\Gamma$

Off-re
$A^{(1)}_{fi}$ be



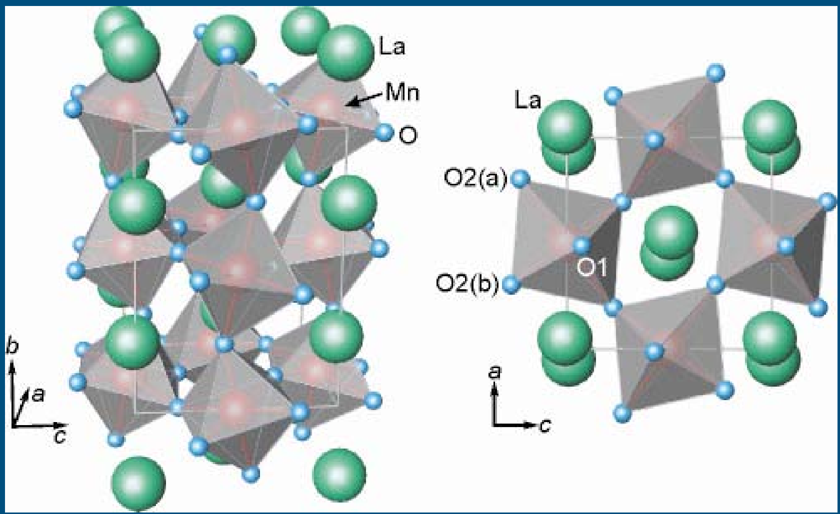
$$A^{(2)}_{fi} \propto \frac{\omega_{\text{res}} J^2}{\Delta^3} \langle f | \sum_{i,j,k} e^{i\mathbf{q}\cdot\mathbf{R}_i} (\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_i \cdot \mathbf{S}_k) | i \rangle$$

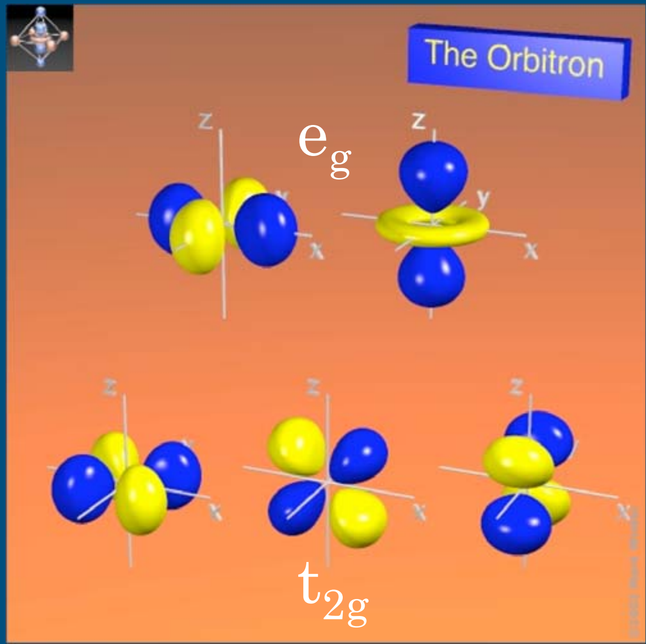
Currently, $\mathbf{q} = 0$ is being measured by G. Ghiringhelli *et al.*

To be continued!

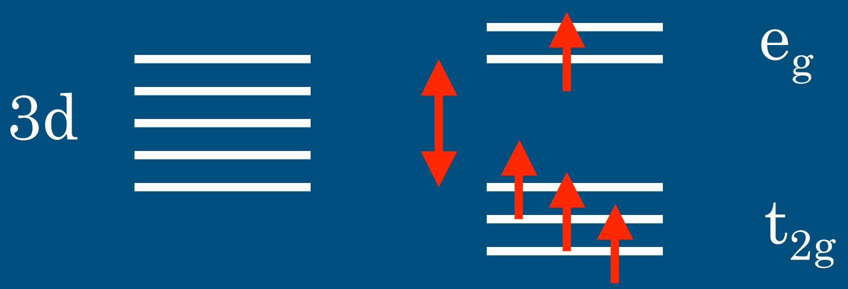
Outlook - Orbital RIXS

- $LaMnO_3$ has a cubic structure:





- MnO_6 octahedron induces crystal field:



$LaMnO_3$: A-type AFM order: spins align FM in each layer.

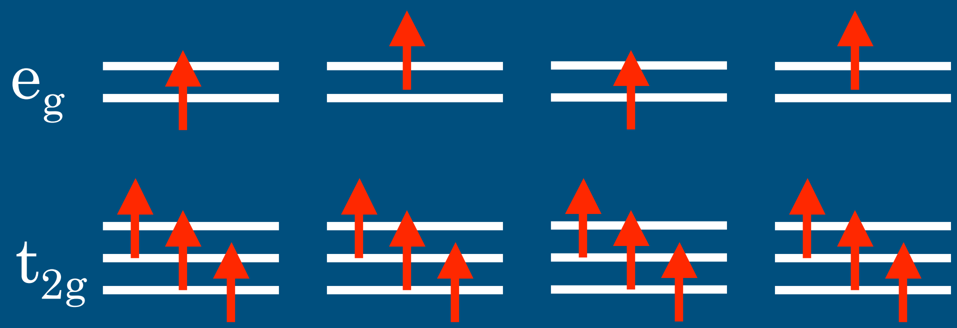
Orbital RIXS

Kugel-Khomskii model without Hund’s rule coupling:

$$H \propto \sum_{<i,j>} \left(\mathbf{S}_i \cdot \mathbf{S}_j + \frac{1}{4} \right) H^{ij}_{\mathrm{orb}}$$

To first order, orbitals of different layers decouple!

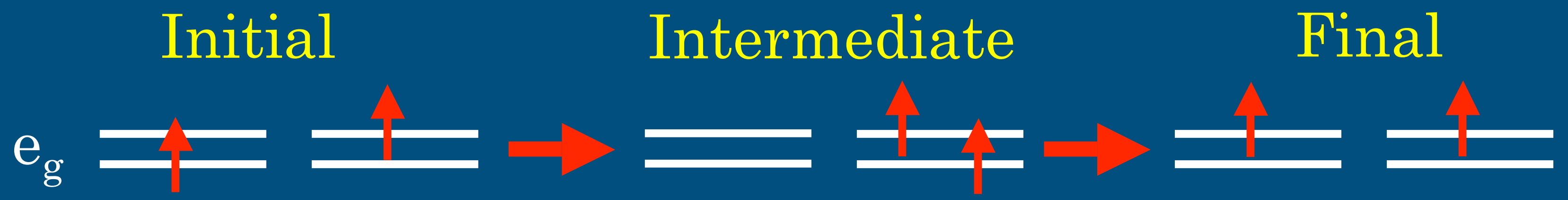
- e_g orbitals order ‘antiferro-orbitally’:



Order by superexchange and/or Jahn-Teller distortion of octahedra

- Excitations: e_g orbital waves (analogous to spin waves)

Orbital RIXS

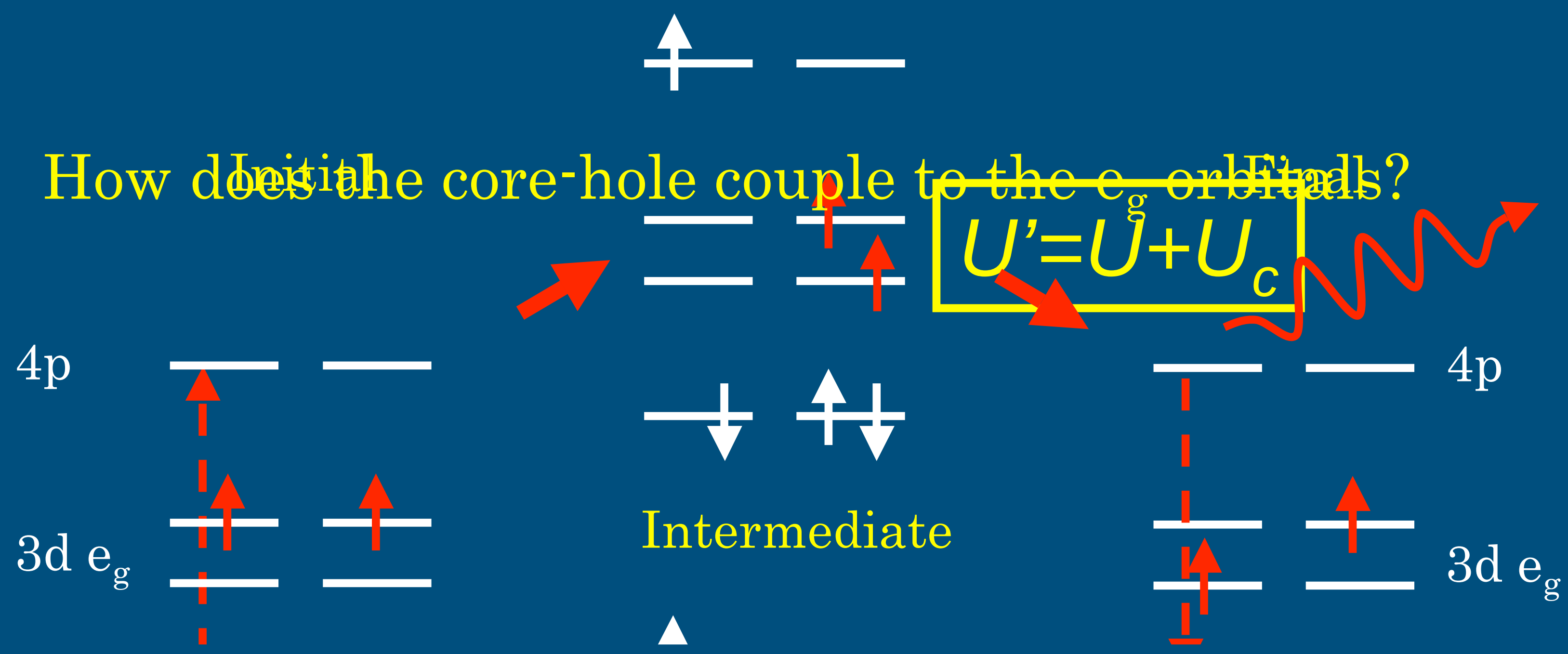


$$H_{\text{orb}} \sim S_i^+ S_j^z$$

Looks like Heisenberg, but no conservation of pseudo-S^z. This leads to single orbiton excitations.

S. Ishihara & S. Maekawa, PRB **62**, 2338 (2000)
J. van den Brink, P. Horsch, F. Mack & A. M. Oles, PRB **59**, 6795 (1999)

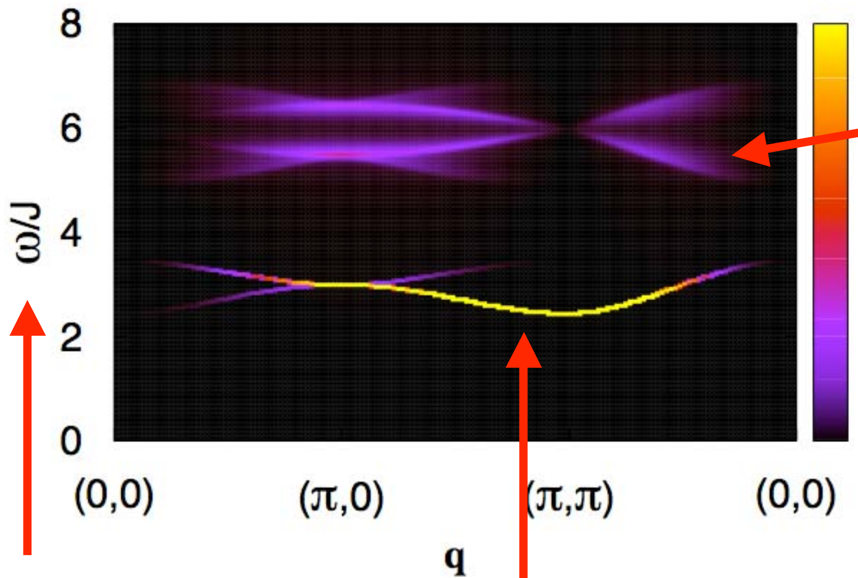
Orbital RIXS



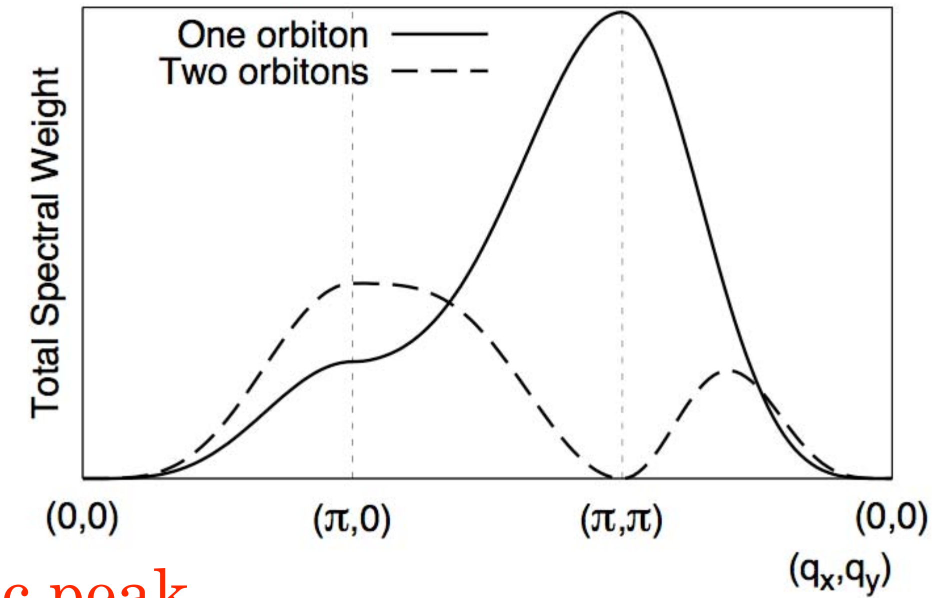
Again, core-hole locally modifies superexchange!

Orbital RIXS

Orbital RIXS spectrum for $LaMnO_3$



Two-orbiton continuum



J = 25 meV

One-orbiton peak

Experimental limitations:
Resolution = 100 meV, inelastic peak

Conclusions

- Experimental progress is enormous
- Theoretical spectra can be easily obtained with the UCL expansion using e.g. a model Hamiltonian
- We now know which charge correlation functions we measure in indirect RIXS
- Both direct and indirect RIXS are a new probe for magnons (La_2CuO_4)
- We can possibly detect orbitons in the manganites!