Resonant Inelastic X-ray Scattering on elementary excitations

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TECHNISCHE UNIVERSITÄT DRESDEN

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

Autumn School in
Correlated Electrons
Jülich
13.09.2016

Outline

1. Introducing RIXS

2. Magnetic RIXS on low dimensional magnets

1. Introducing RIXS

Basic Scattering Process

Direct & Indirect RIXS

5 features of RIXS

Elementary Excitations Accessible to RIXS

Progress in past decade

2. Magnetic RIXS on low dimensional magnets

Quasi 2D cuprates

Quasi 1D cuprates

Quasi 2D iron pnictide

Quasi 2D iridate

Doped Cu & Fe systems

2. Magnetic RIXS on low dimensional magnets

Quasi 2D cuprates

Some theory

Some experiments

A (big) open problem

Basic Scattering Process

Direct and Indirect RIXS

What is *Resonant Inelastic X-ray scattering* — *RIXS*

X-ray scattering: *photon in → solid → photon out*

inelastic: $\omega_{out} < \omega_{in}$

resonant: *tune ω_{in} to an atomic absorption edge*

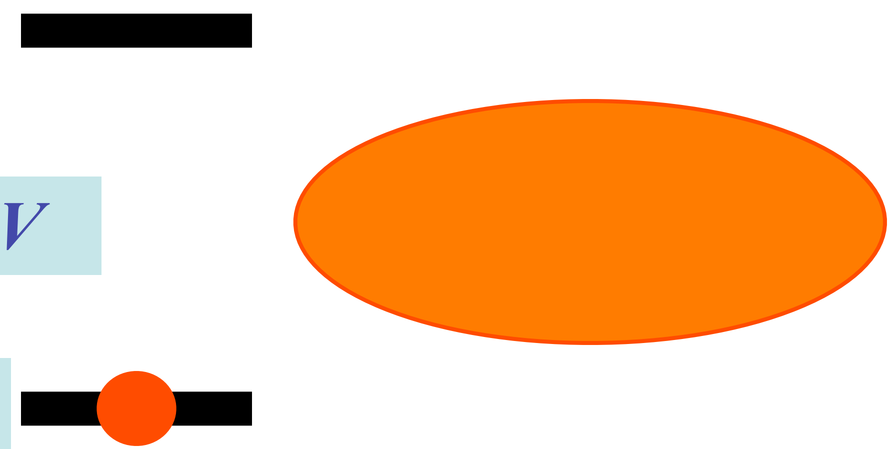
Cu

K-edge

4 p

~9 KeV

1 s



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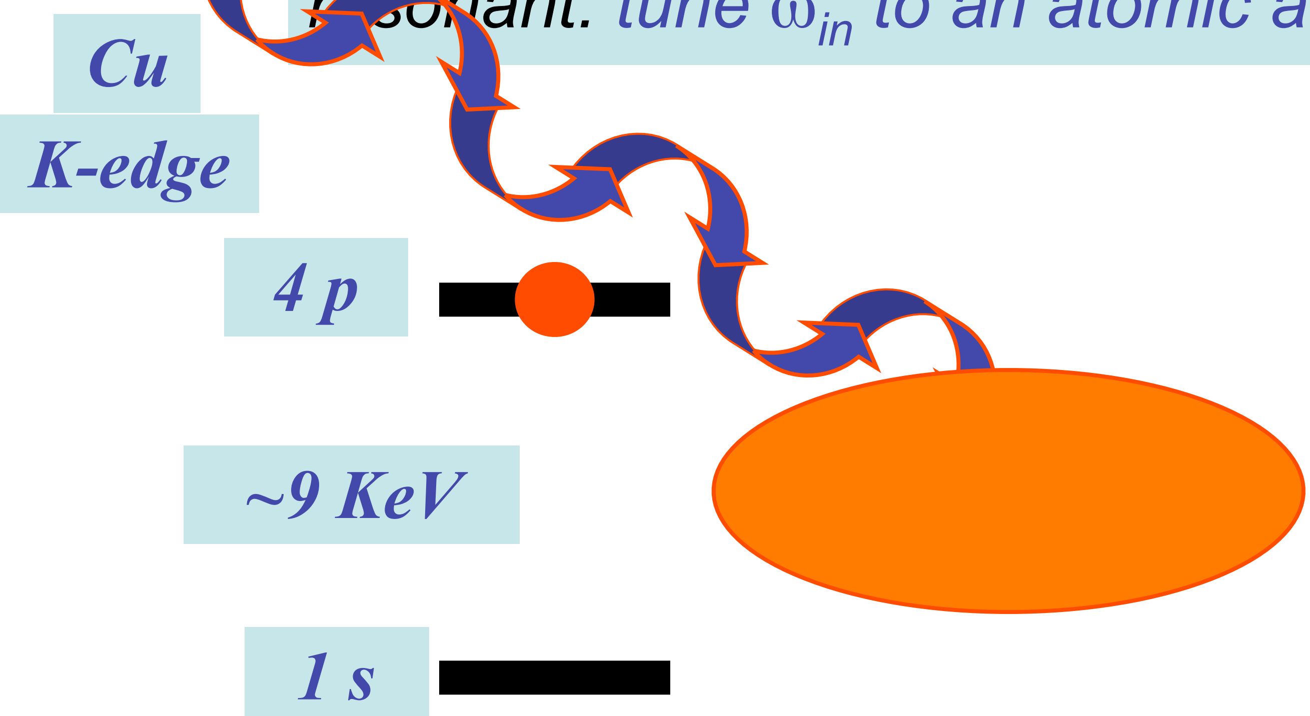


What is

Resonant

Inelastic

X-ray scattering

RIXS

X-ray scattering: *photon in → solid → photon out*

inelastic: $\omega_{out} < \omega_{in}$

resonant: tune ω_{in} *to an atomic absorption edge*

Cu

K-edge

INDIRECT

4 p

~9 KeV

1 s

Momentum transfer: q

Energy loss

What is *Resonant* *Inelastic* *X-ray scattering* — *RIXS*

X-ray scattering: *photon in→ solid → photon out*

inelastic: $\omega_{out} < \omega_{in}$

resonant: *tune ω_{in} to an atomic absorption edge*

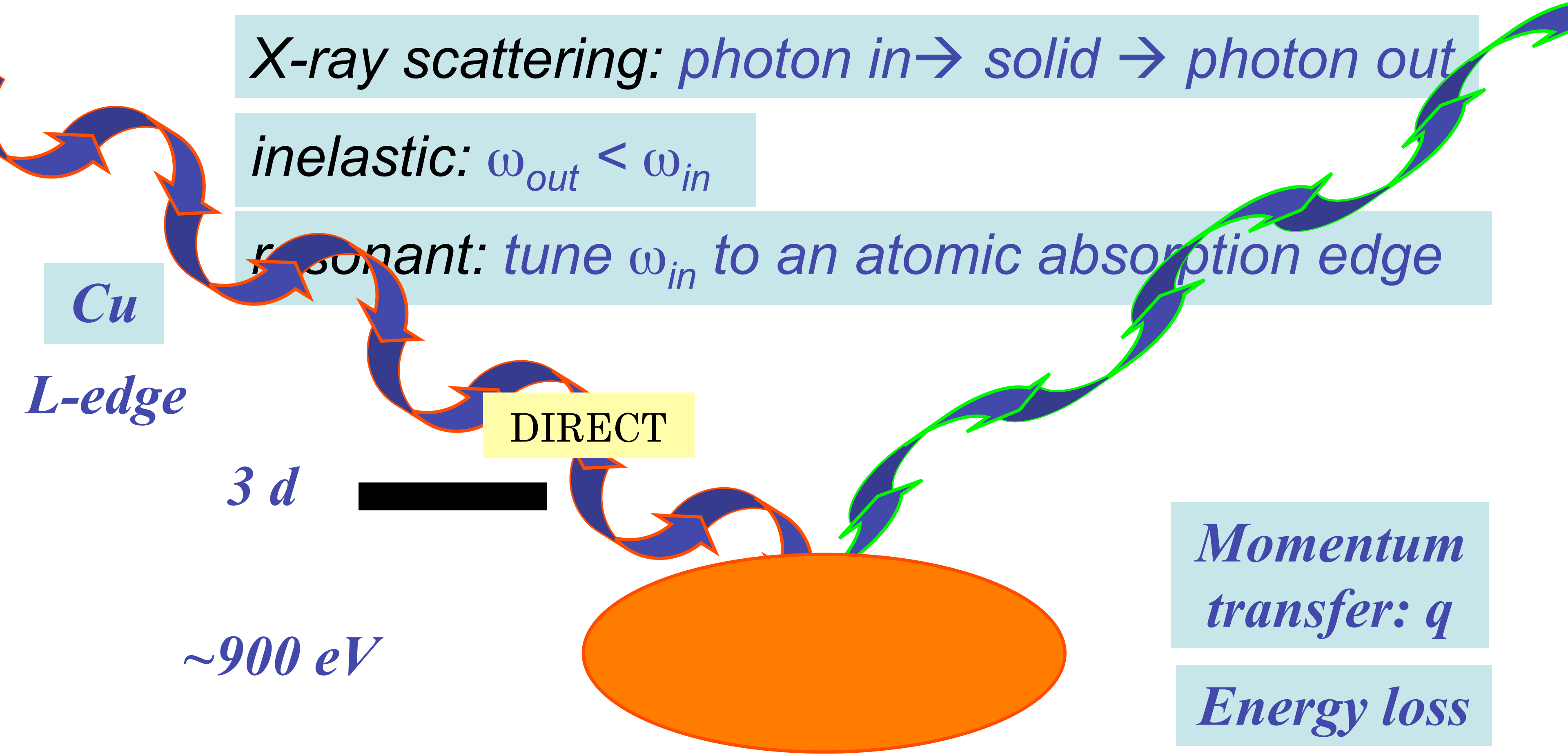
Cu

L-edge

3 d

~900 eV

2 p



Momentum transfer: q

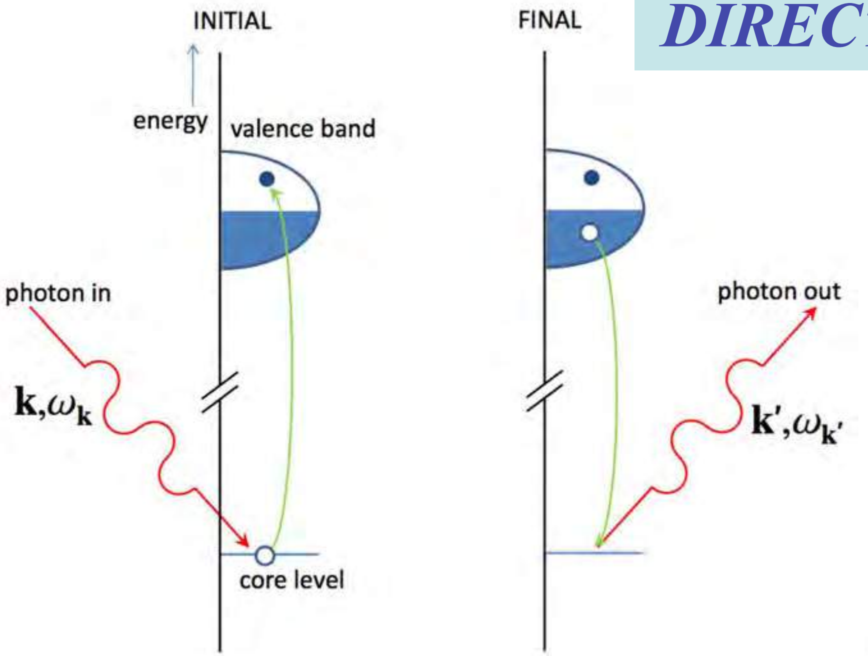
Energy loss

resolution < 100 meV

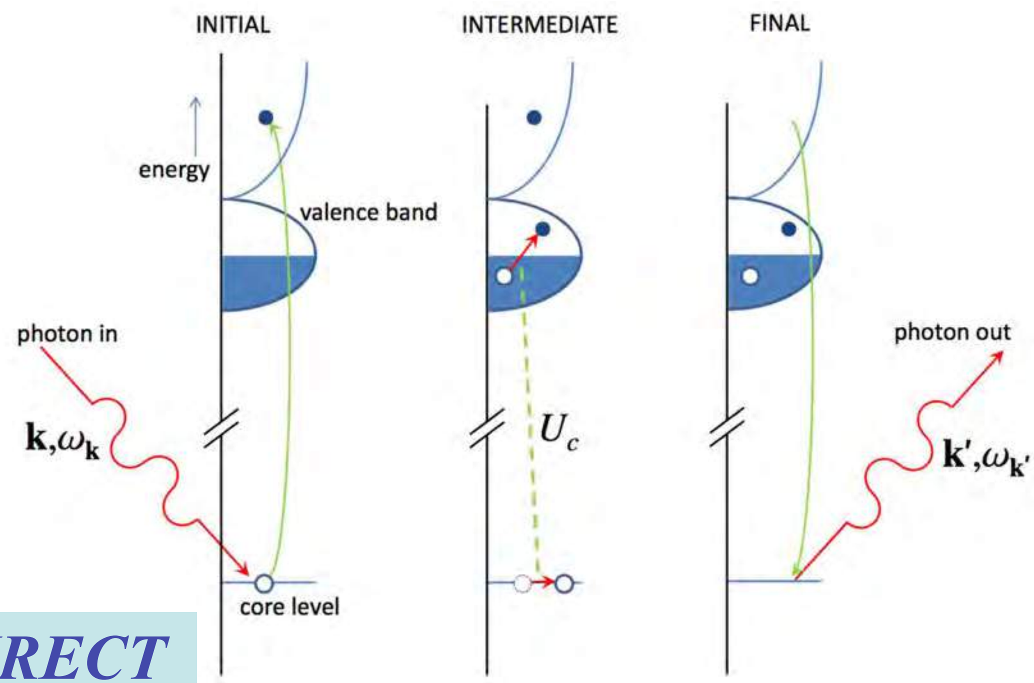
Direct and Indirect RIXS

DIRECT

scattering via absorption-emission matrix elements



scattering via intermediate state core-hole shake-up



INDIRECT

$$RIXS = |GS\rangle \rightarrow XAS \rightarrow |INTERMEDIATE\rangle \rightarrow XES \rightarrow |FS\rangle$$

Complicated state with strong core-hole potential

Local atomic transition

Local atomic transition

Contains chemical detail and atom specific physics

But:

$$RIXS = |GS\rangle \rightarrow .... \rightarrow |FS\rangle$$

Carries low energy, long wavelength, elementary excitations

Universal effective low energy (magnetic) behavior

RIXS amplitude F and intensity I

Intensity

Amplitude

$$I(\omega, \mathbf{k}, \mathbf{k}', \epsilon, \epsilon') = \sum_f |\mathcal{F}_{fg}(\mathbf{k}, \mathbf{k}', \epsilon, \epsilon', \omega_{\mathbf{k}})|^2 \times \delta(E_f + \hbar\omega_{\mathbf{k}'} - E_g - \hbar\omega_{\mathbf{k}})$$

ω_{loss}

momentum in

out

q_{loss}*=k'-k*

RIXS amplitude F and intensity I

polarization in

out

Intensity

Amplitude

$$I(\omega, \mathbf{k}, \mathbf{k}', \epsilon, \epsilon') = \sum_f |\mathcal{F}_{fg}(\mathbf{k}, \mathbf{k}', \epsilon, \epsilon', \omega_{\mathbf{k}})|^2$$
$$\times\, \delta(E_f + \hbar\omega_{\mathbf{k}'} - E_g - \hbar\omega_{\mathbf{k}})$$

ω_{loss}

momentum in

out

energy conservation

final energy

initial energy

ω_{out}

ω_{in}

2nd Order: Resonant Scattering

RIXS amplitude

$$\mathcal{F}_{fg}(\mathbf{k},\mathbf{k}',\boldsymbol{\epsilon},\boldsymbol{\epsilon}',\omega_{\mathbf{k}},\omega_{\mathbf{k}'}) = \sum_n \frac{\langle f|\mathcal{D}'^{\dagger}|n\rangle\langle n|\mathcal{D}|g\rangle}{E_g + \hbar\omega_{\mathbf{k}} - E_n + i\Gamma_n}$$

Kramers-Heisenberg expression

RIXS transition operator

$$\mathcal{D} = \frac{1}{im\omega_{\mathbf{k}}}\sum_{i=1}^{N} e^{i\mathbf{k}\cdot\mathbf{r}_i}\boldsymbol{\epsilon}\cdot\mathbf{p}_i,$$

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

Greens function expression for RIXS amplitude

RIXS amplitude

$$\mathcal{F}_{fg}(\mathbf{k},\mathbf{k}',\epsilon,\epsilon',\omega_{\mathbf{k}},\omega_{\mathbf{k}'}) = \sum_n \frac{\langle f|\mathcal{D}'^{\dagger}|n\rangle\langle n|\mathcal{D}|g\rangle}{E_g+\hbar\omega_{\mathbf{k}}-E_n+i\Gamma_n}$$

Greens function

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

=intermediate state propagator

with

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

so that:

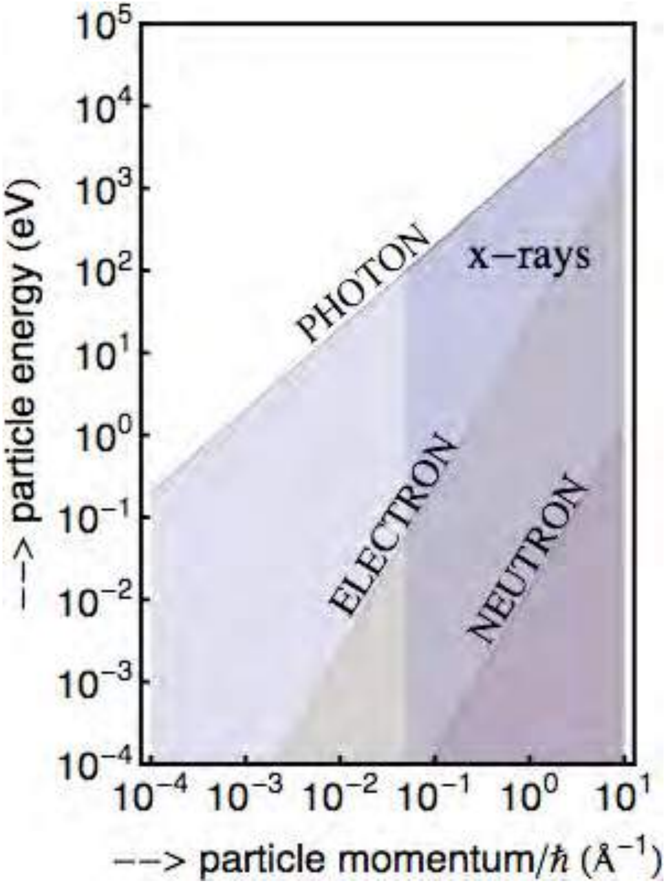
$$\mathcal{F}_{fg} = \langle f|\mathcal{D}'^{\dagger}G(z_{\mathbf{k}})\mathcal{D}|g\rangle$$

5 distinguishing features of RIXS

Why *Inelastic Scattering*

- *with X-rays*
- *at a resonance*

X-rays: *momentum ~ Å^{-1}*



probe charged excitations

have angular momentum l=1

polarization dependence

Solid: *Lattice spacings ~ Å*

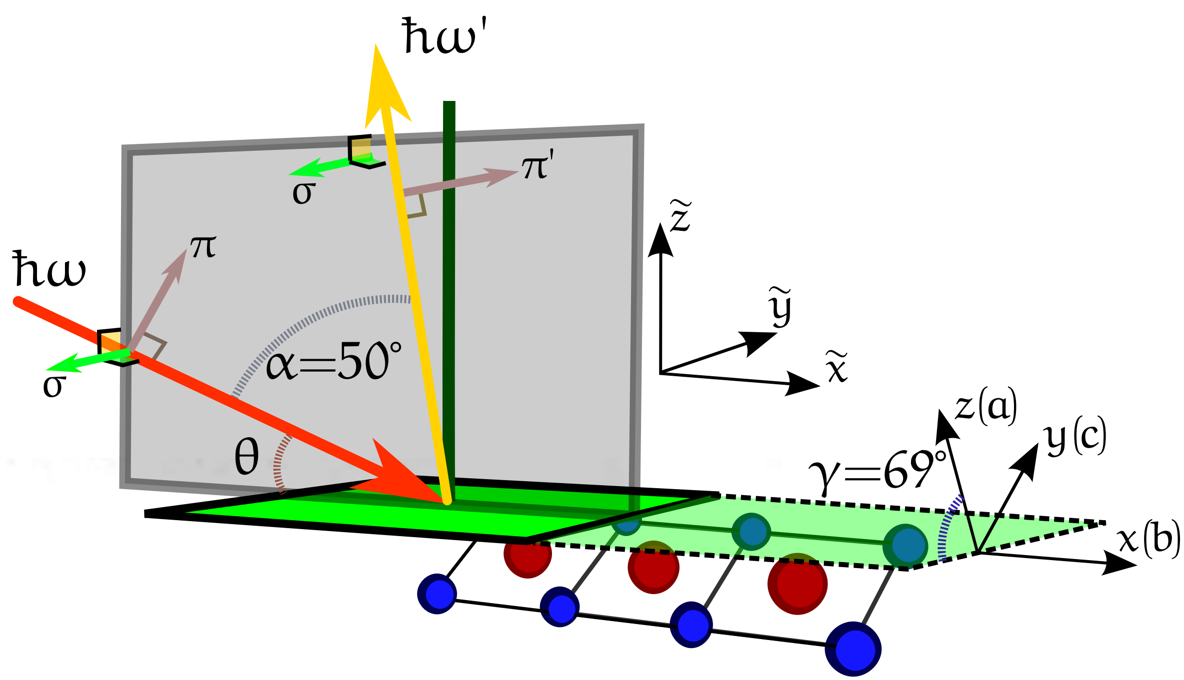
Brillouin zone ~ Å^{-1}

Visible Photons: *momentum ~ 10^{-3} Å^{-1}*

X-rays at 10 keV *momentum ~ 5 Å^{-1}*

several BZ's

1. RIXS exploits both the *energy and momentum* dependence of the photon scattering cross-section. Comparing the energies of a neutron, electron, and photon, each with a wavelength on the order of the relevant length scale in a solid, *i.e.* the interatomic lattice spacing, which is on the order of a few Angstroms, it is obvious that an x-ray photon has much more energy than an equivalent neutron or electron,



2. RIXS can utilize the *polarization* of the photon: the nature of the excitations created in the material can be disentangled through polarization analysis of the incident and scattered photons, which allows one, through the use of various selection rules, to characterize the symmetry and nature of the excitations. To date, no experimental facility allows the polarization of the scattered photon to be measured, though the incident photon polarization is frequently varied. It is important to note that a polarization change of a photon is necessarily related to an angular momentum change. Conservation of angular momentum means that any angular momentum lost by the scattered photons has been transferred to elementary excitations in the solid.

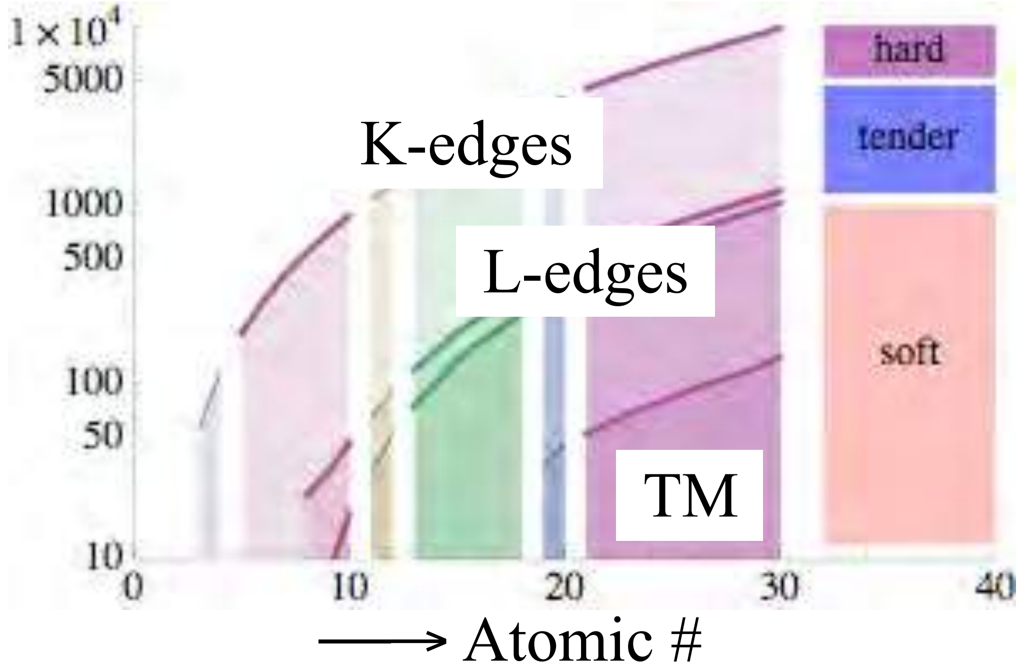
Why Inelastic Scattering with X-rays at a resonance

At resonance:

enhanced loss features

choose element & electronic shell

X-ray Absorption Edges



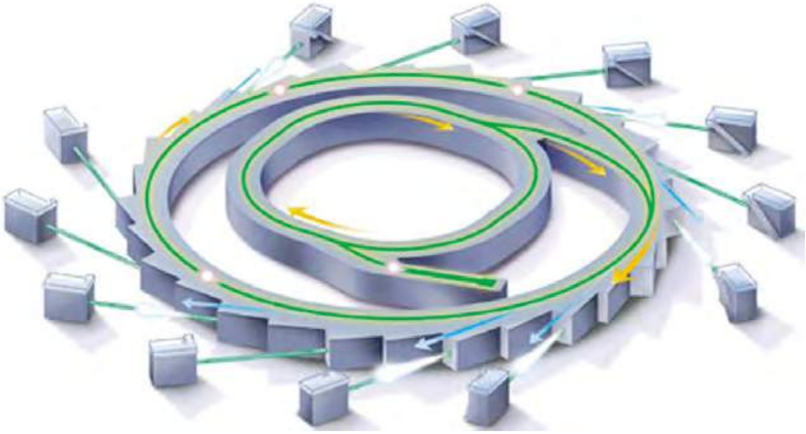
X-ray penetration depth: ~microns

RIXS is bulk sensitive

Tunable X-ray sources

synchrotron

ESRF Grenoble



Tunable X-ray sources

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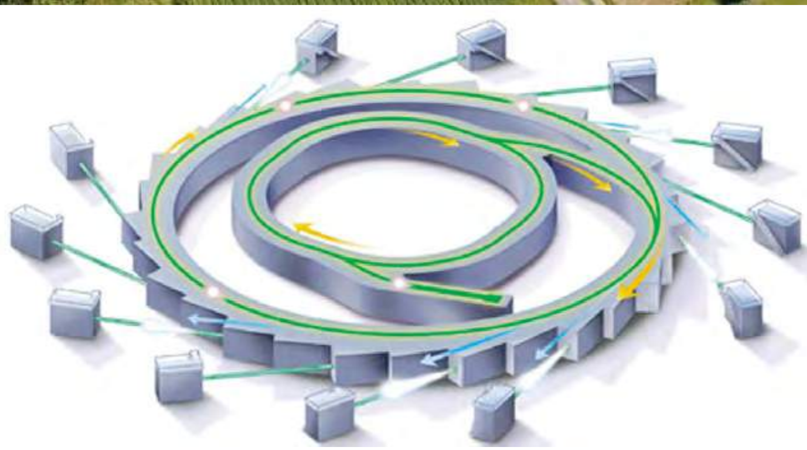
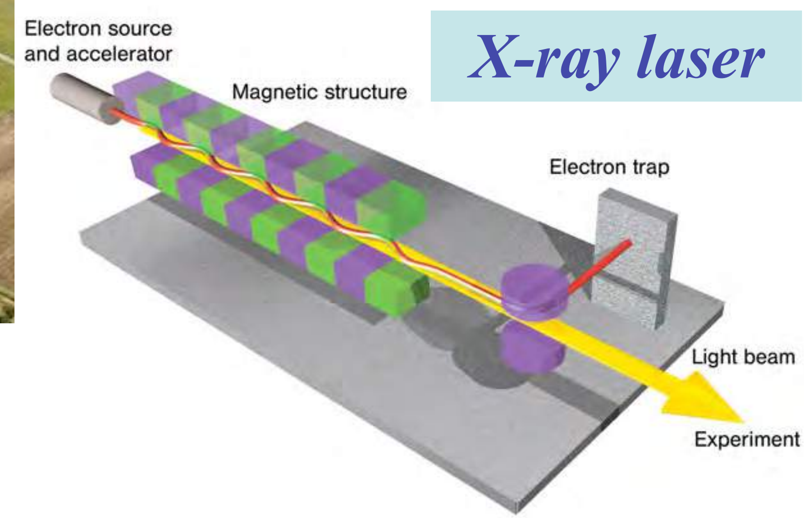
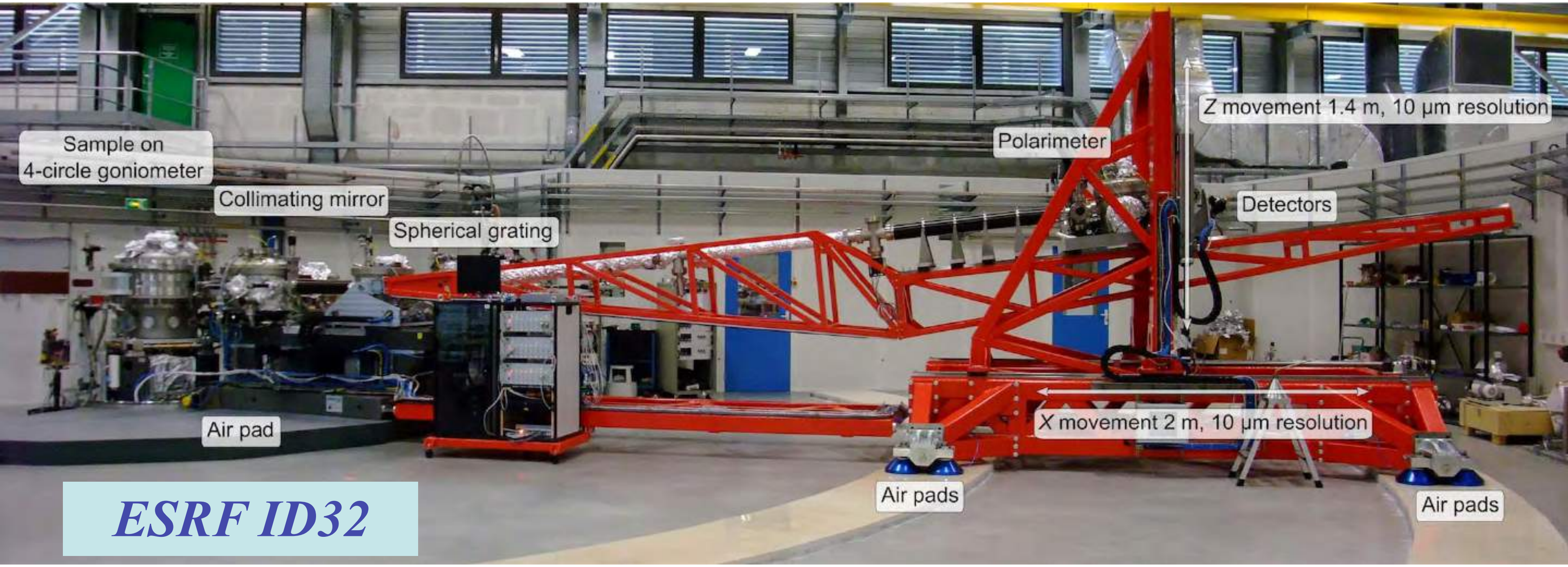


X-ray laser



LCLS, Stanford

X-ray spectrometers



ESRF ID32

Soft RIXS: TM L-edges

3. RIXS is *element and orbital specific*: Chemical sensitivity arises by tuning the incident photon energy to specific atomic transitions of the different types of atoms in a material. Such transitions are called absorption edges. RIXS can even differentiate between the same chemical element at sites with inequivalent chemical bondings, with different valencies or at inequivalent crystallographic positions if the absorption edges in these cases are distinguishable.

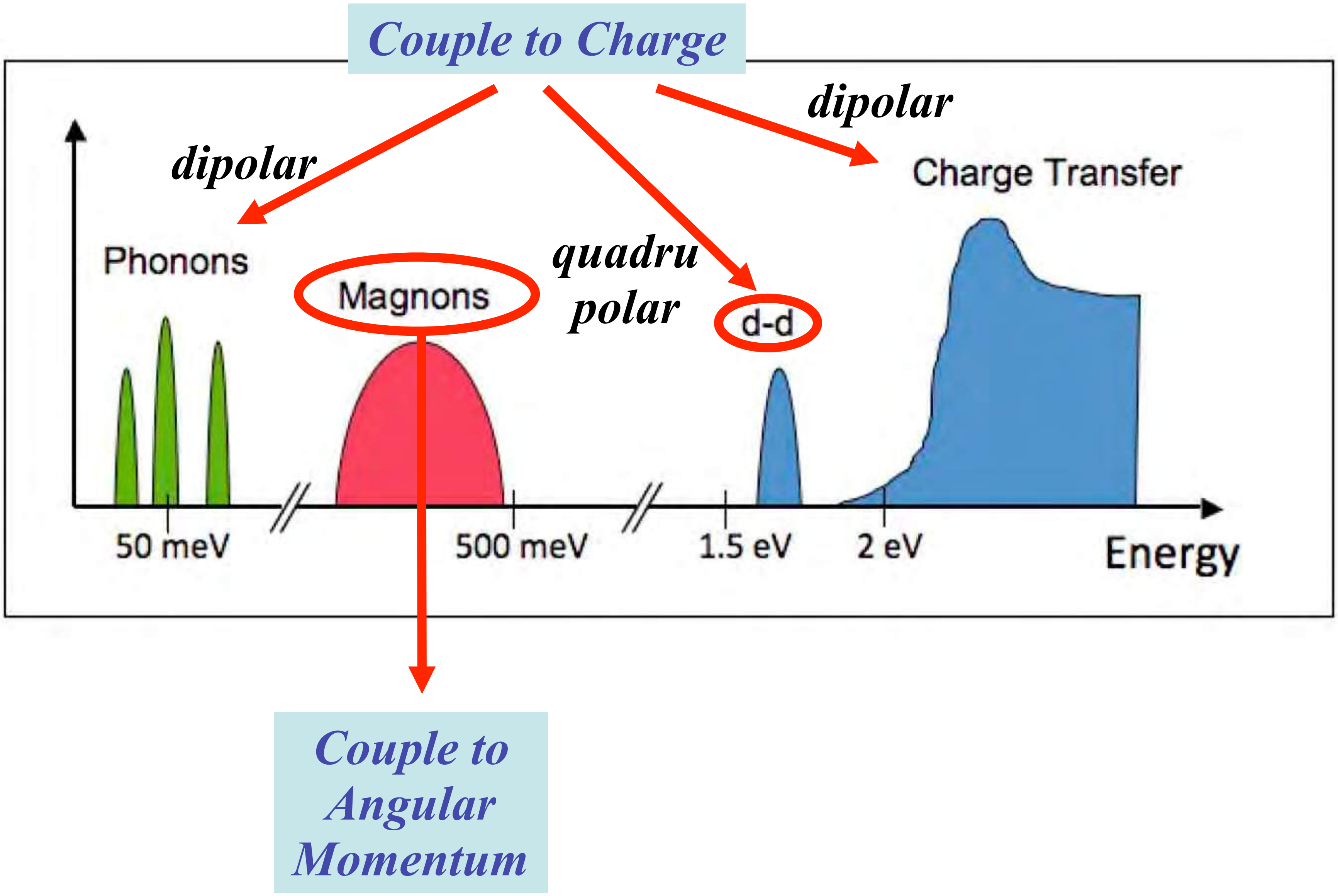
4. RIXS is *bulk sensitive*: the penetration depth of resonant x-ray photons is material and scattering geometry-specific, but typically is on the order of a few μm in the hard x-ray regime (for example at transition metal K-edges) and on the order of 0.1 μm in the soft x-ray regime (e.g transition metal L-edges).

5. RIXS needs only *small sample volumes*: the photon-matter interaction is relatively strong, compared to for instance the neutron-matter interaction strength. In addition, photon sources deliver many orders of magnitude more particles per second, in a much smaller spot, than do neutron sources. These facts make RIXS possible on very small volume samples, thin films, surfaces and nano-objects, in addition to bulk single crystal or powder samples.

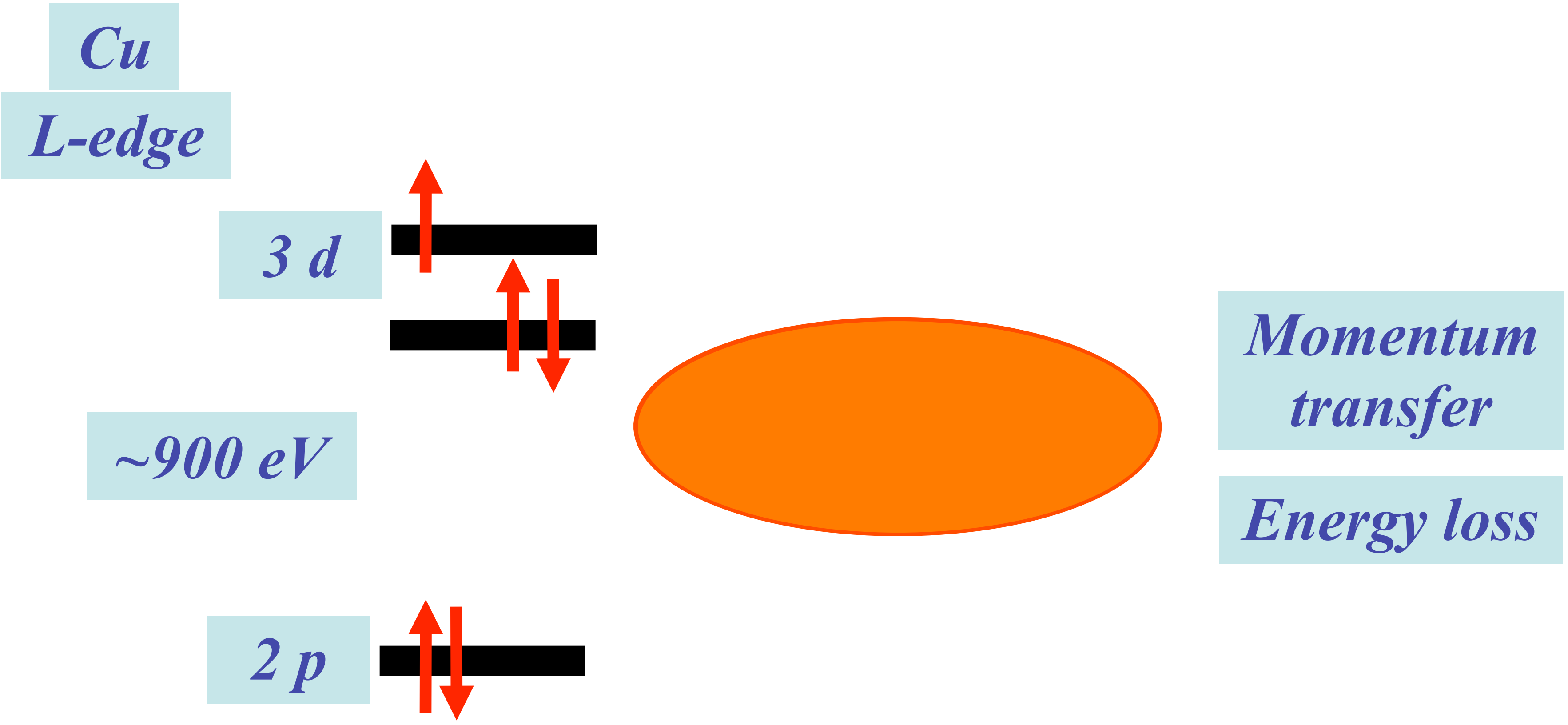
Elementary Excitations

accessible to RIXS

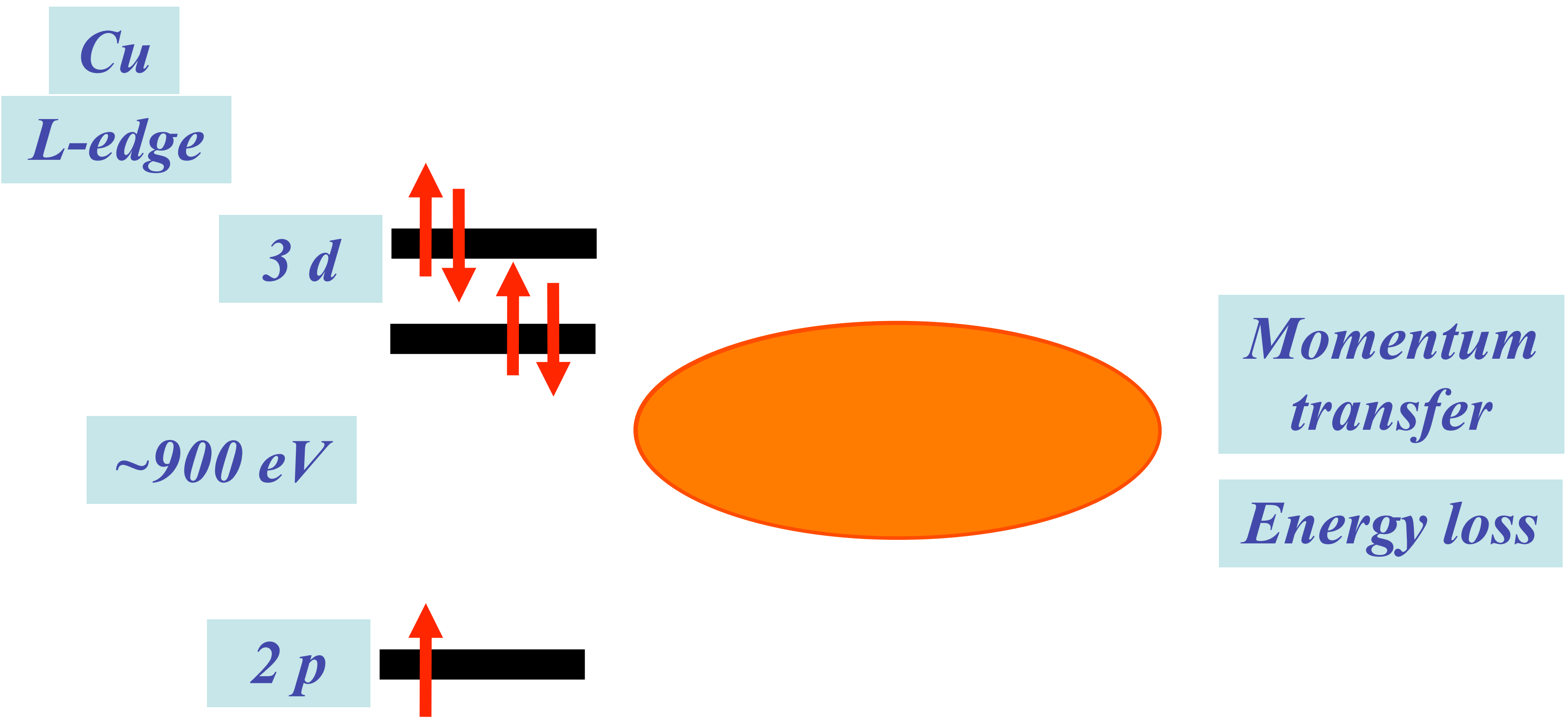
Elementary Excitations in TMO: Schematic



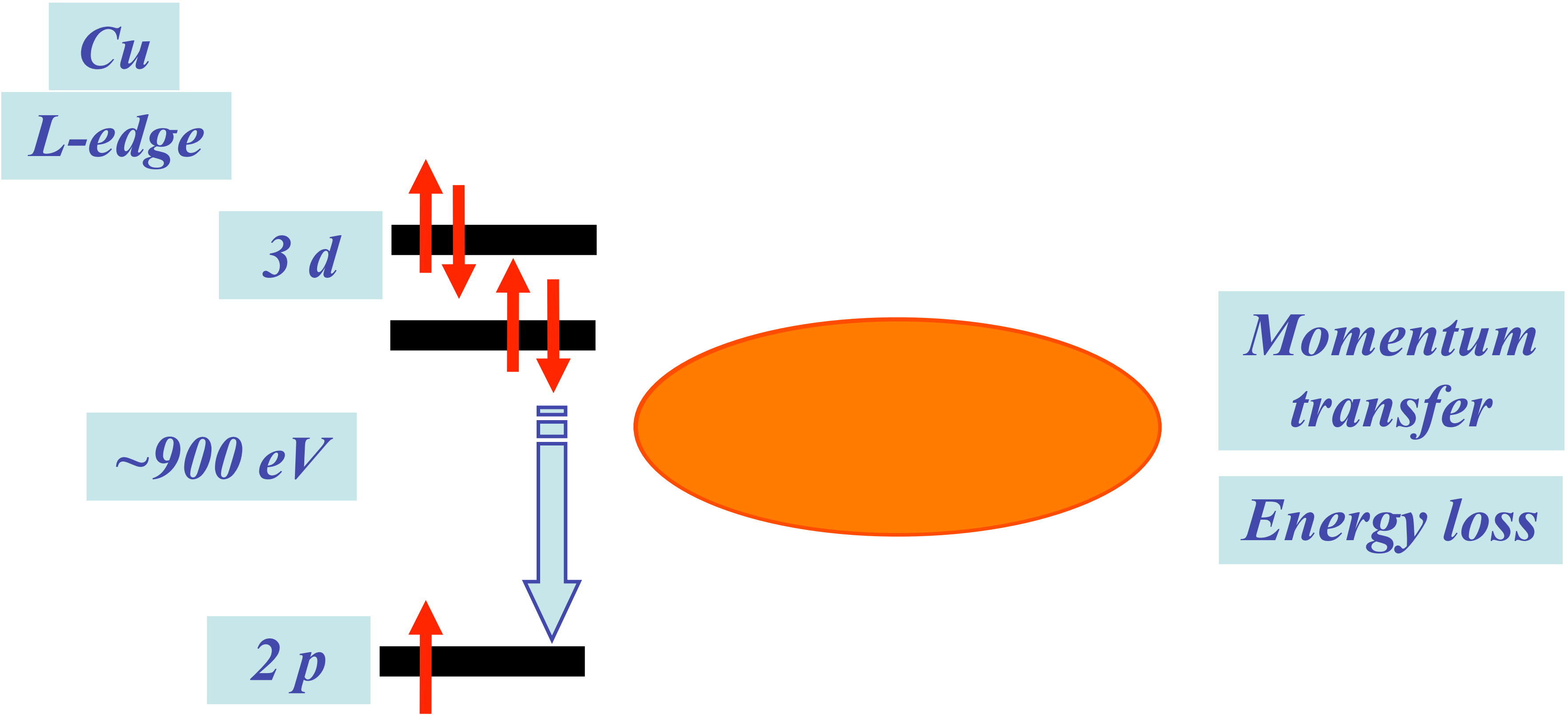
Direct RIXS @ TM L-edges



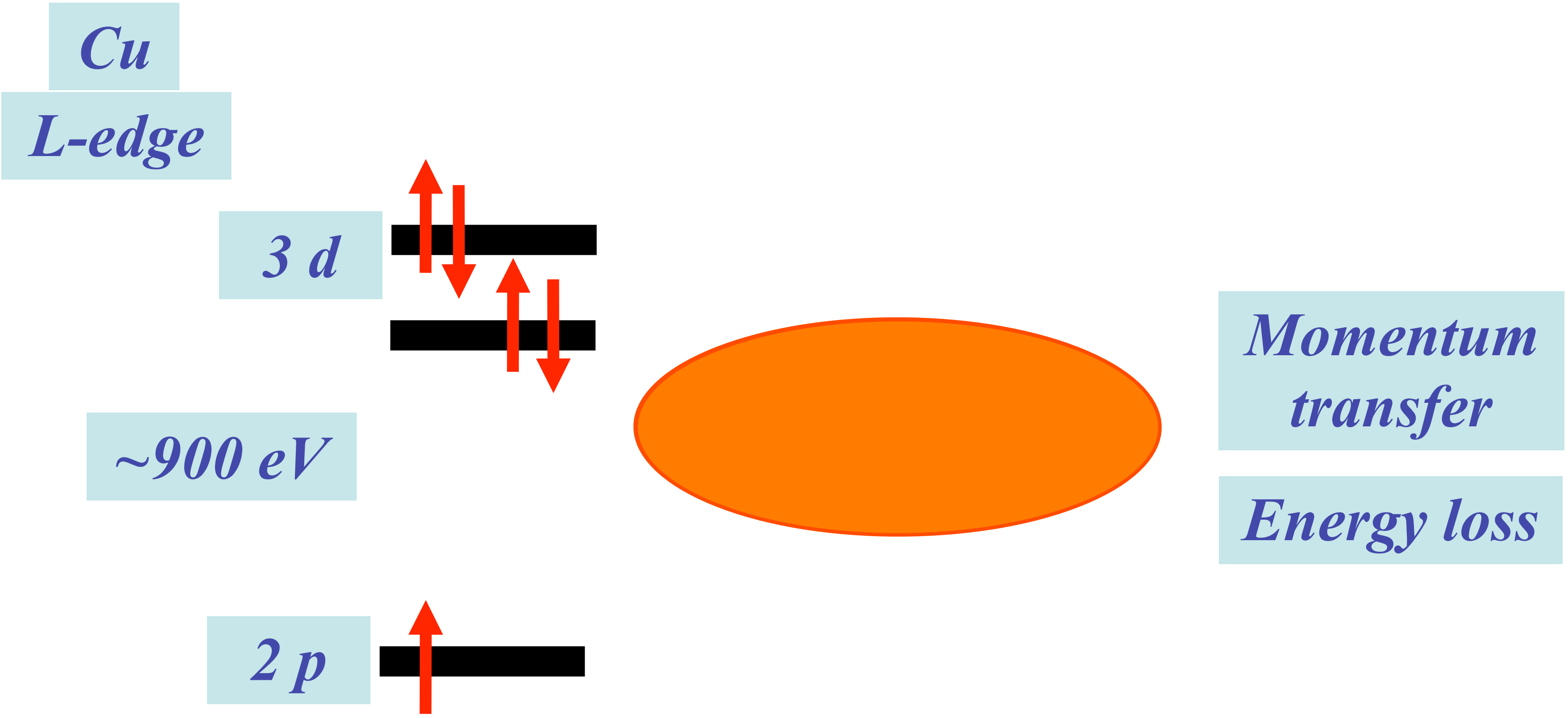
Direct RIXS @ TM L-edges



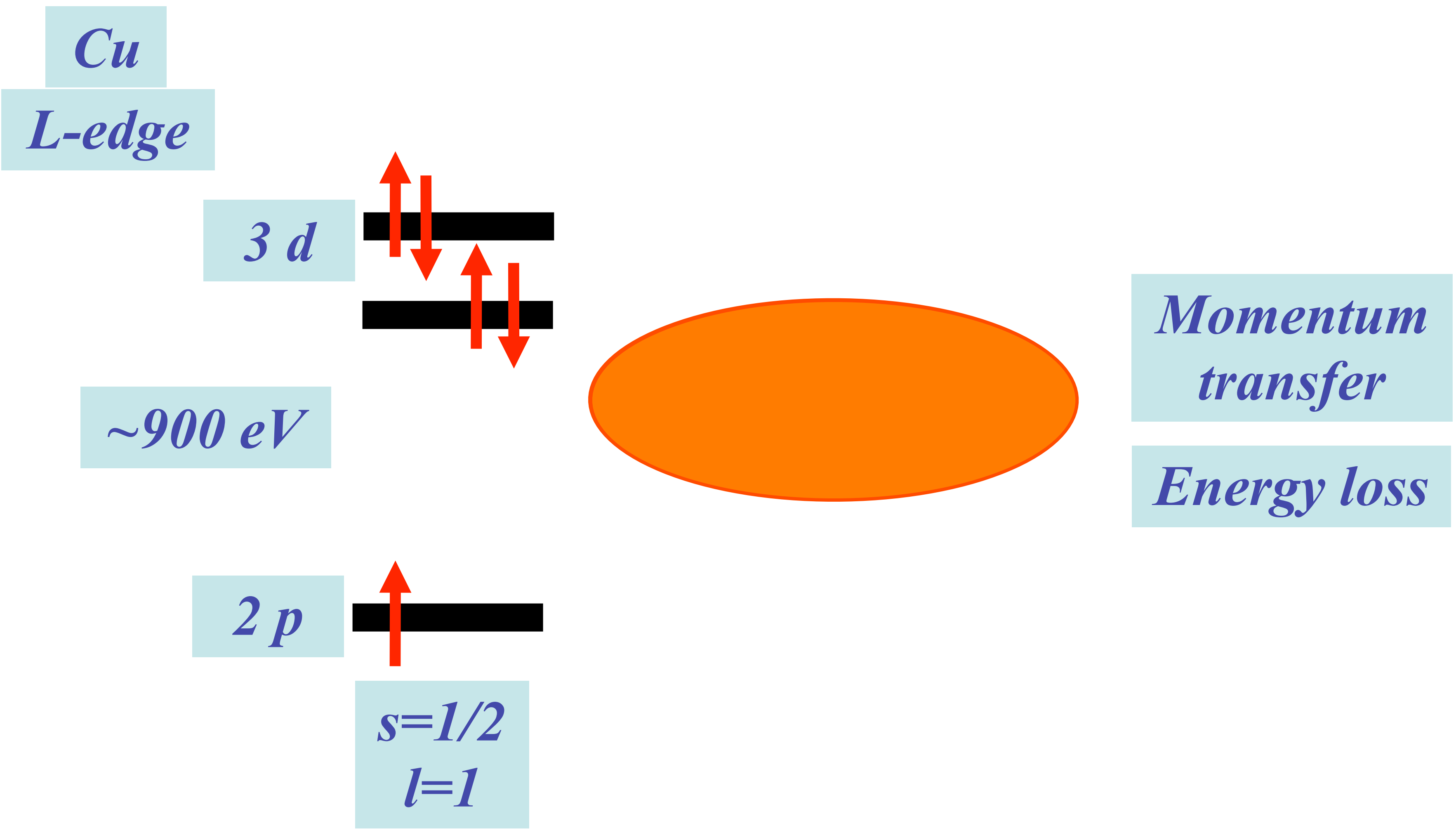
Direct RIXS @ TM L-edges



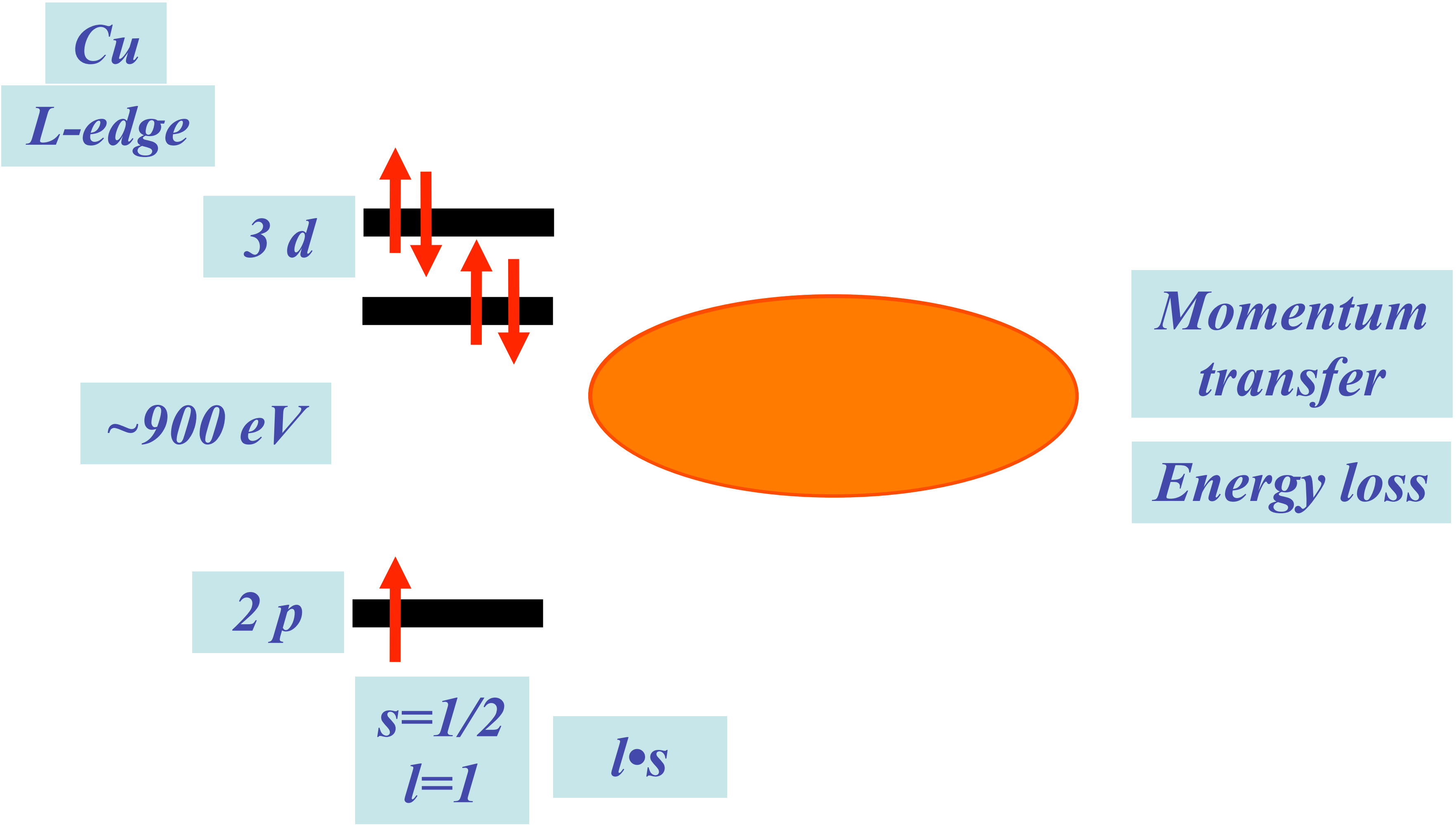
Direct RIXS @ TM L-edges



Direct RIXS @ TM L-edges



Direct RIXS @ TM L-edges



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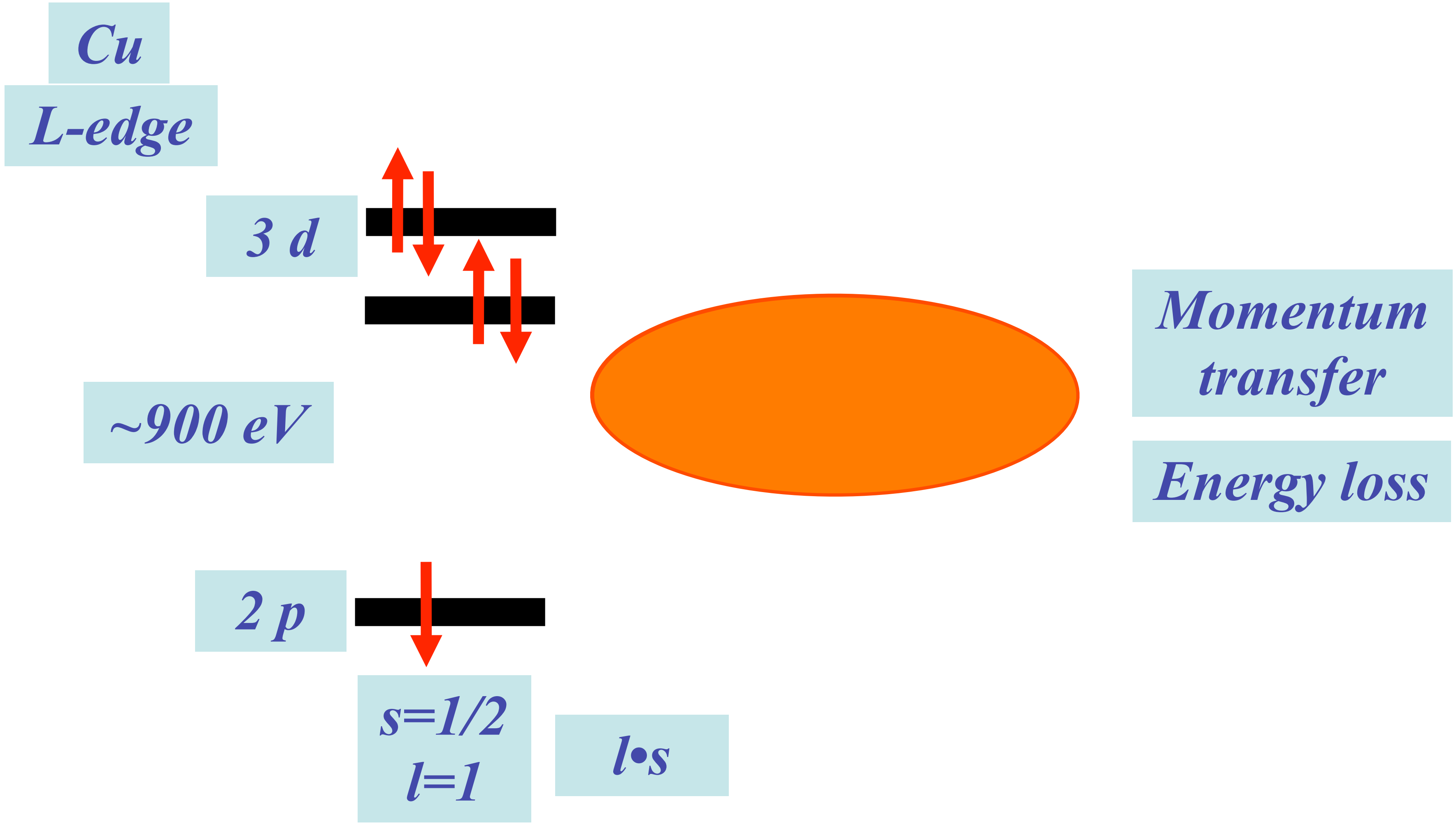

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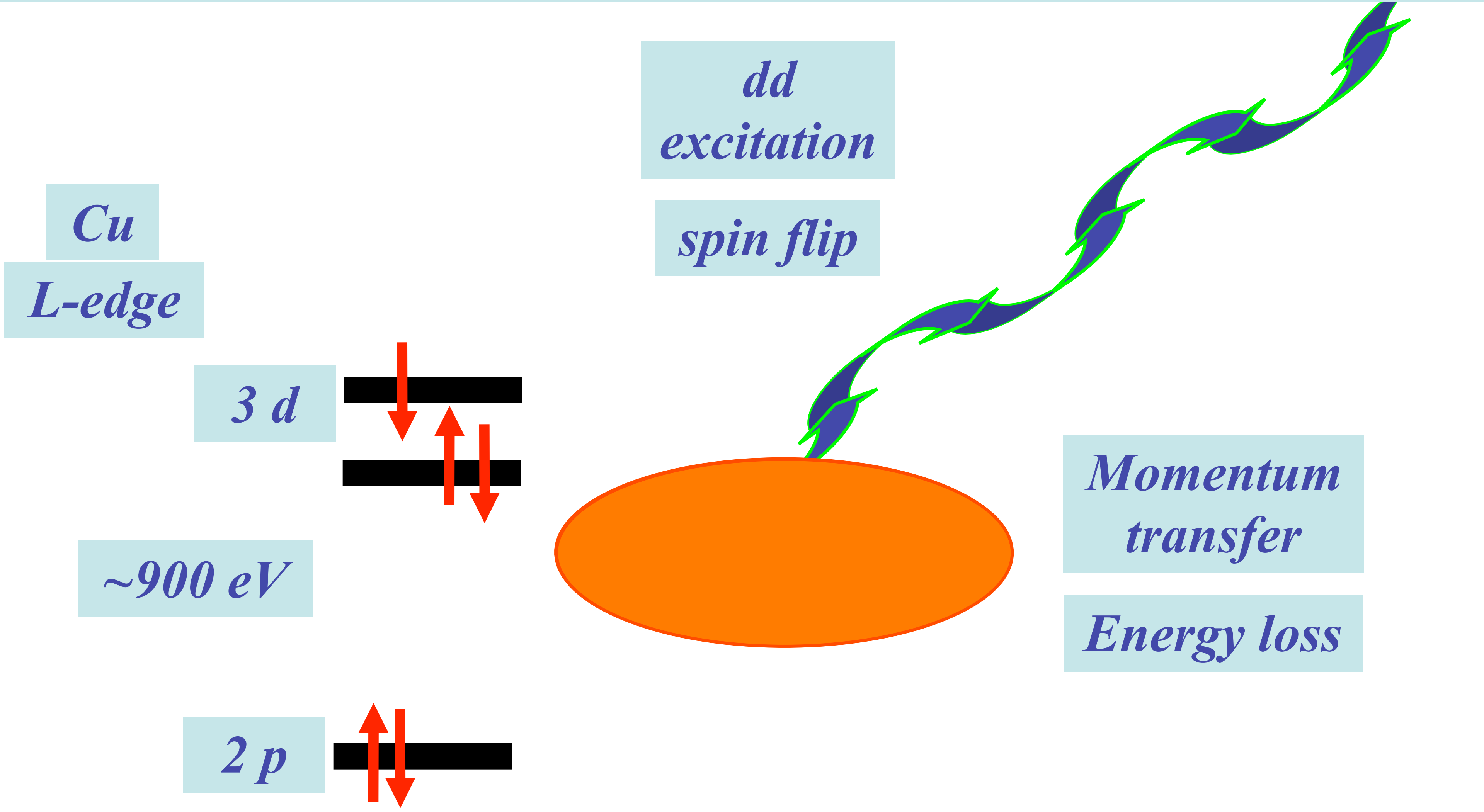

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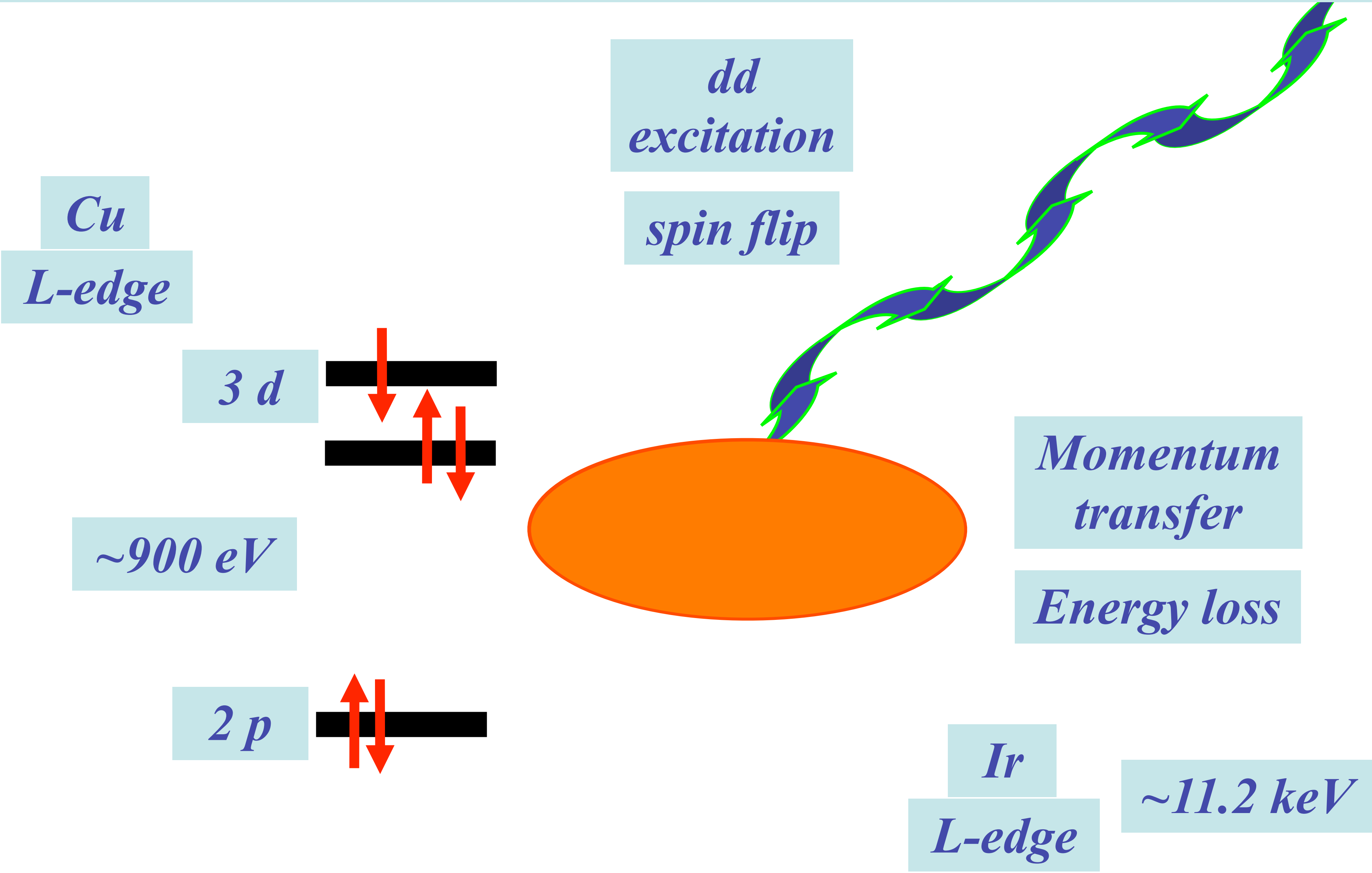
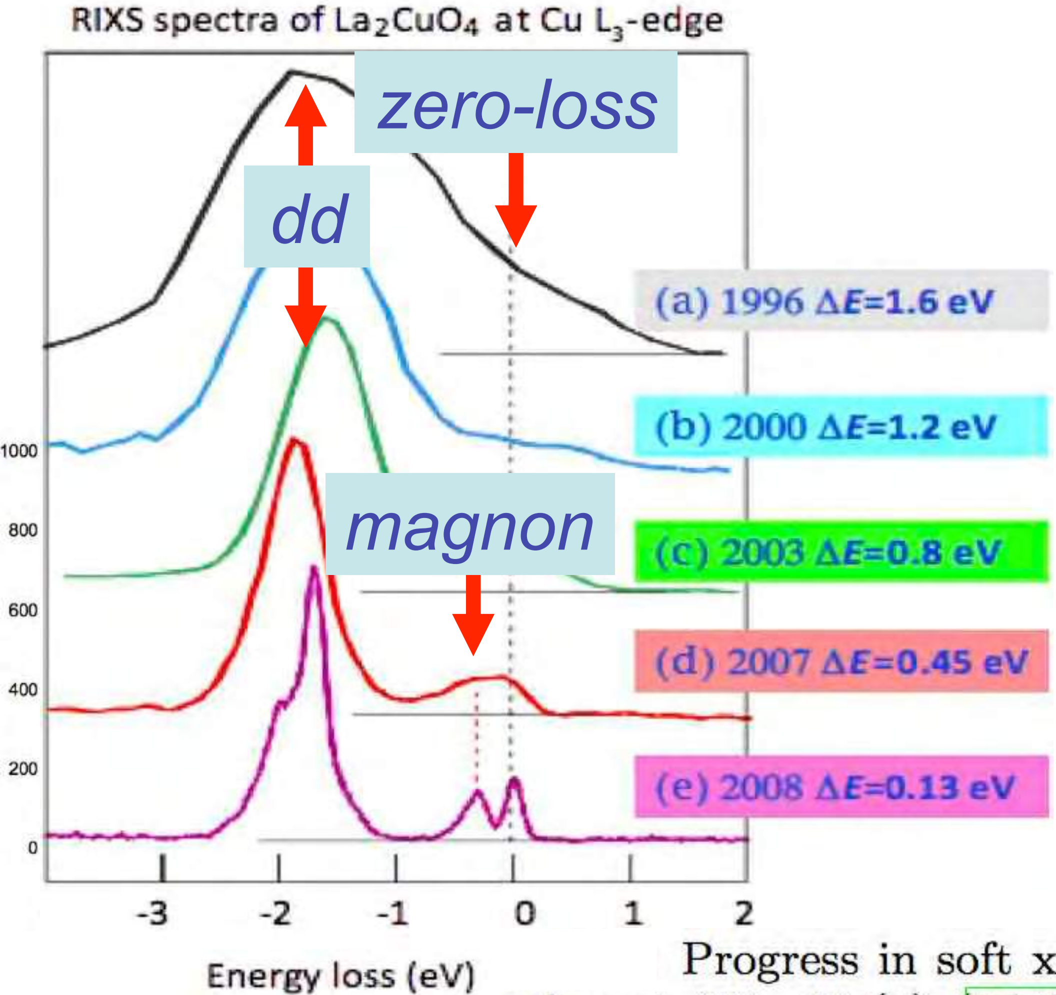


In principle RIXS can probe a very broad class of intrinsic excitations of the system under study – as long as these excitations are overall charge neutral. This constraint arises from the fact that in RIXS the scattered photons do not add or remove charge from the system under study.

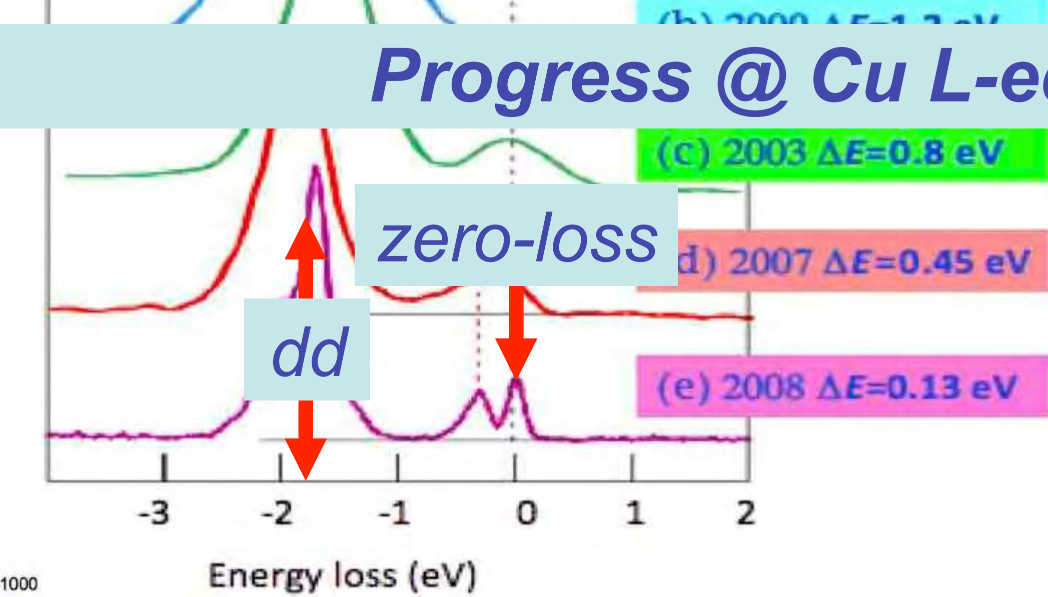
Progress in the Past Decades

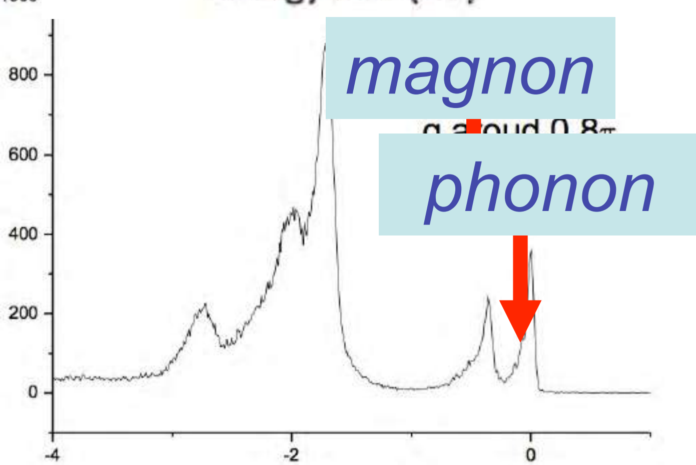
Progress @ Cu L-edge resolution



Progress in soft x-ray RIXS resolution at the Cu L-edge at 931 eV (a) (Ichikawa *et al.*, 1996), BLBB @ Photon Factory (b) I511-3 @ MAX II (Duda *et al.*, 2000b), (c) AXES @ ID08, ESRF(Ghiringhelli *et al.*, 2004) (d) AXES @ ID08, ESRF(Braicovich *et al.*, 2009), (e) SAXES @ SLS (Ghiringhelli *et al.*, 2010). Courtesy of G. Ghiringhelli and L. Braicovich.

Progress @ Cu L-edge resolution





COURTESY
Lucio Braicovich
ESRF Beamline ID32
13.07.2015

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Summary part I

- Direct and Indirect RIXS
- RIXS measures excitation energy & momentum
- Polarization in/out dependence can be studied
- Element and orbital sensitive
- Bulk sensitive & needs small sample volumes
- Measures charge neutral elementary excitations

  spin, orbital, lattice, charge excitons
- Great progress in resolution in the past decade

2. Magnetic RIXS on low dimensional magnets

Quasi 2D cuprates

Quasi 1D cuprates

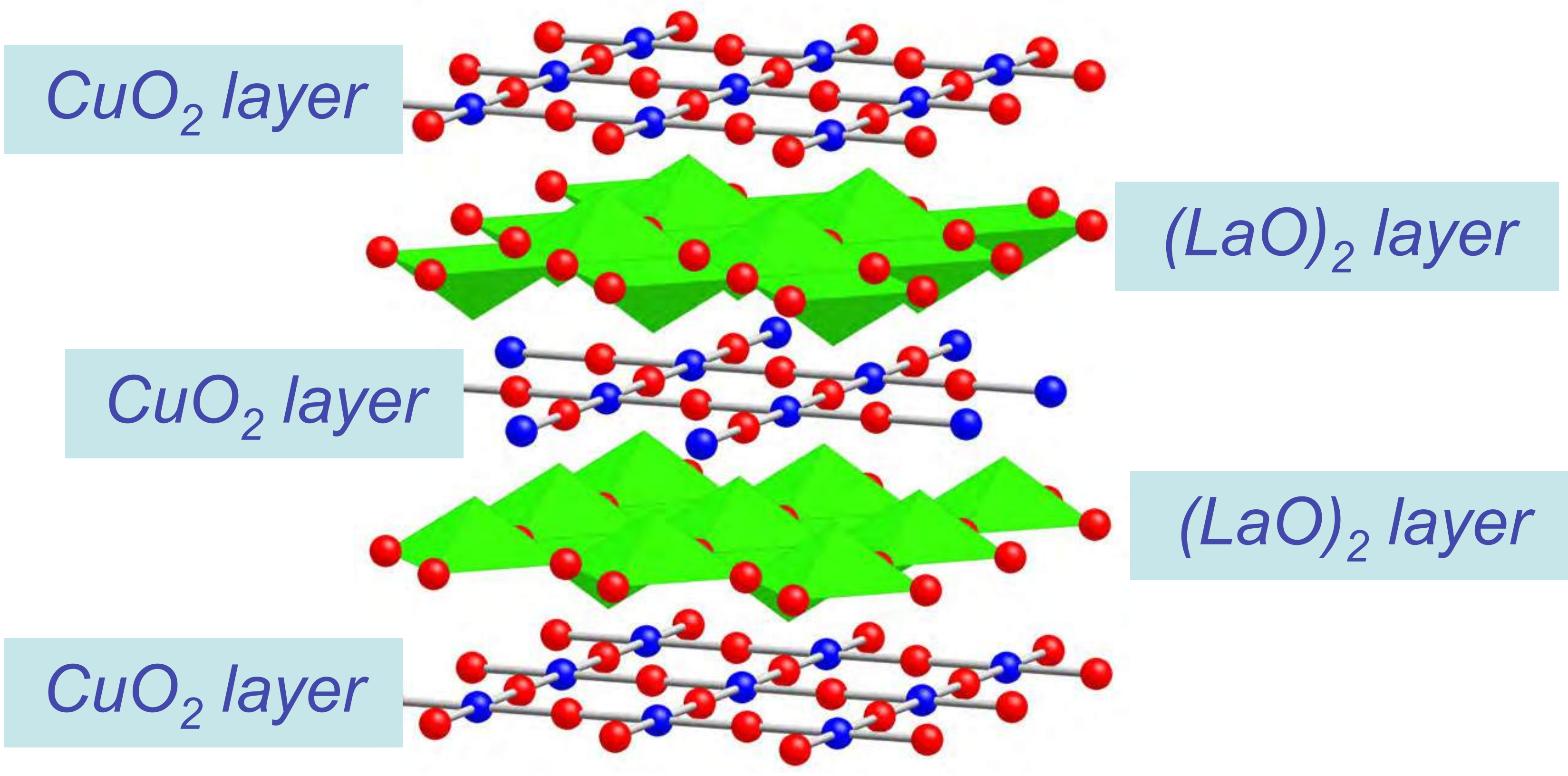
Quasi 2D iron pnictide

Quasi 2D iridate

Doped Cu & Fe systems

Quasi 2D cuprates

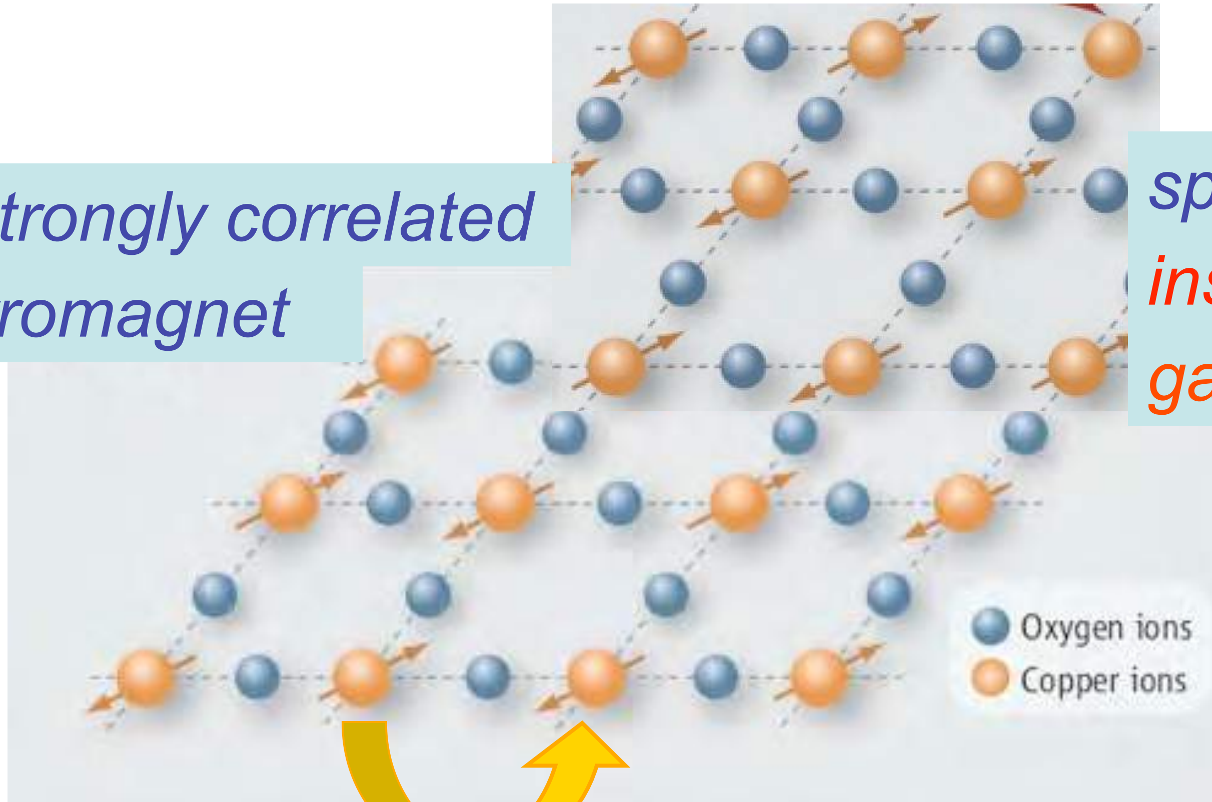
La_2CuO_4 crystal structure



La_2CuO_4 magnetic structure

strongly correlated

antiferromagnet



spin 1/2

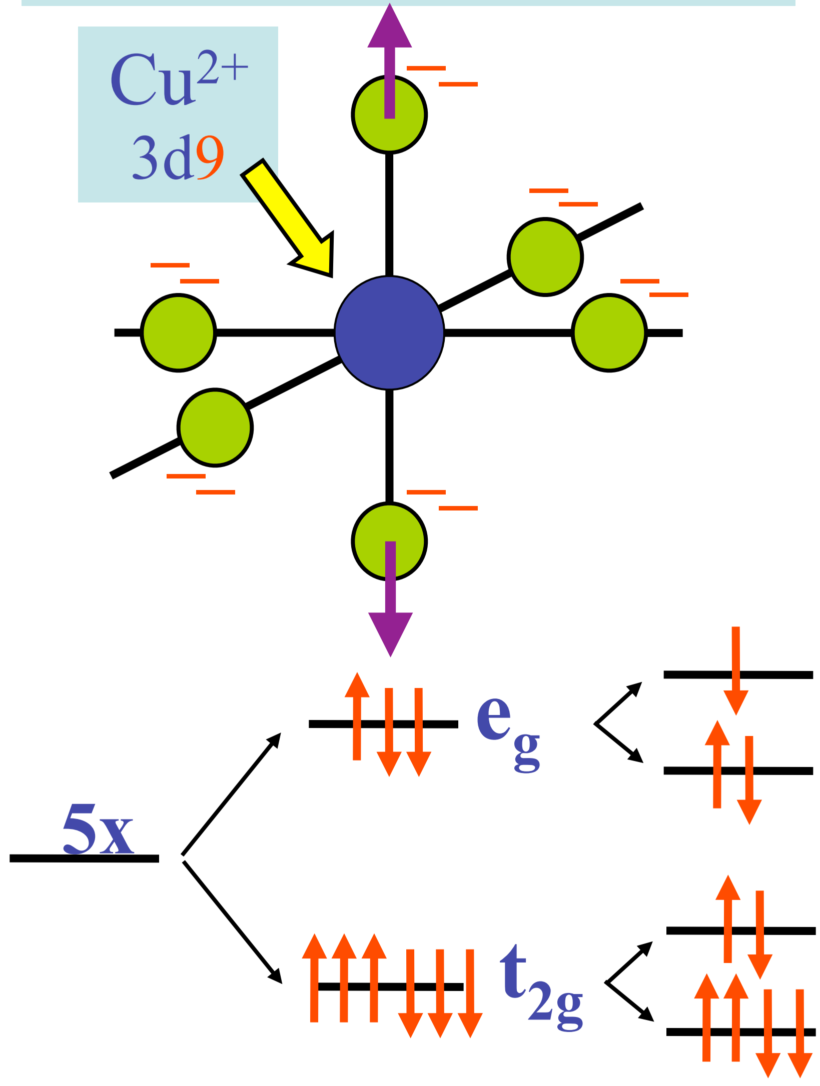
insulator

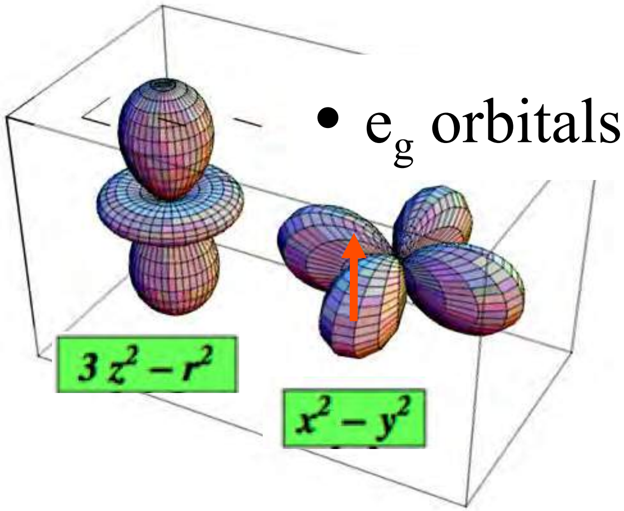
gap ~2 eV

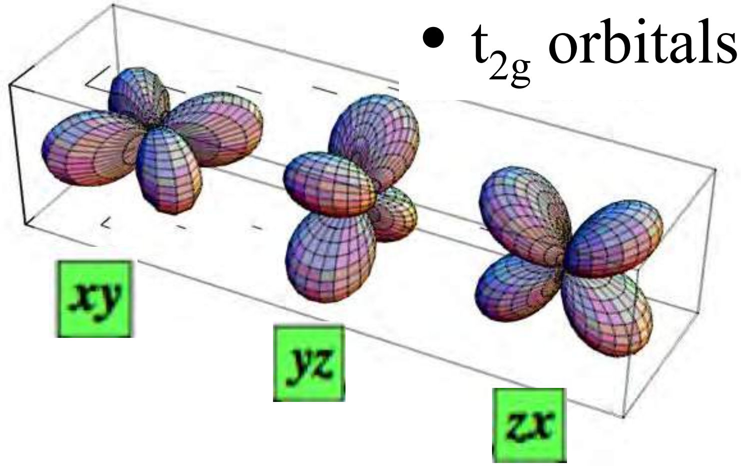
U ~ 6 eV

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

Cubic Crystal field splitting







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

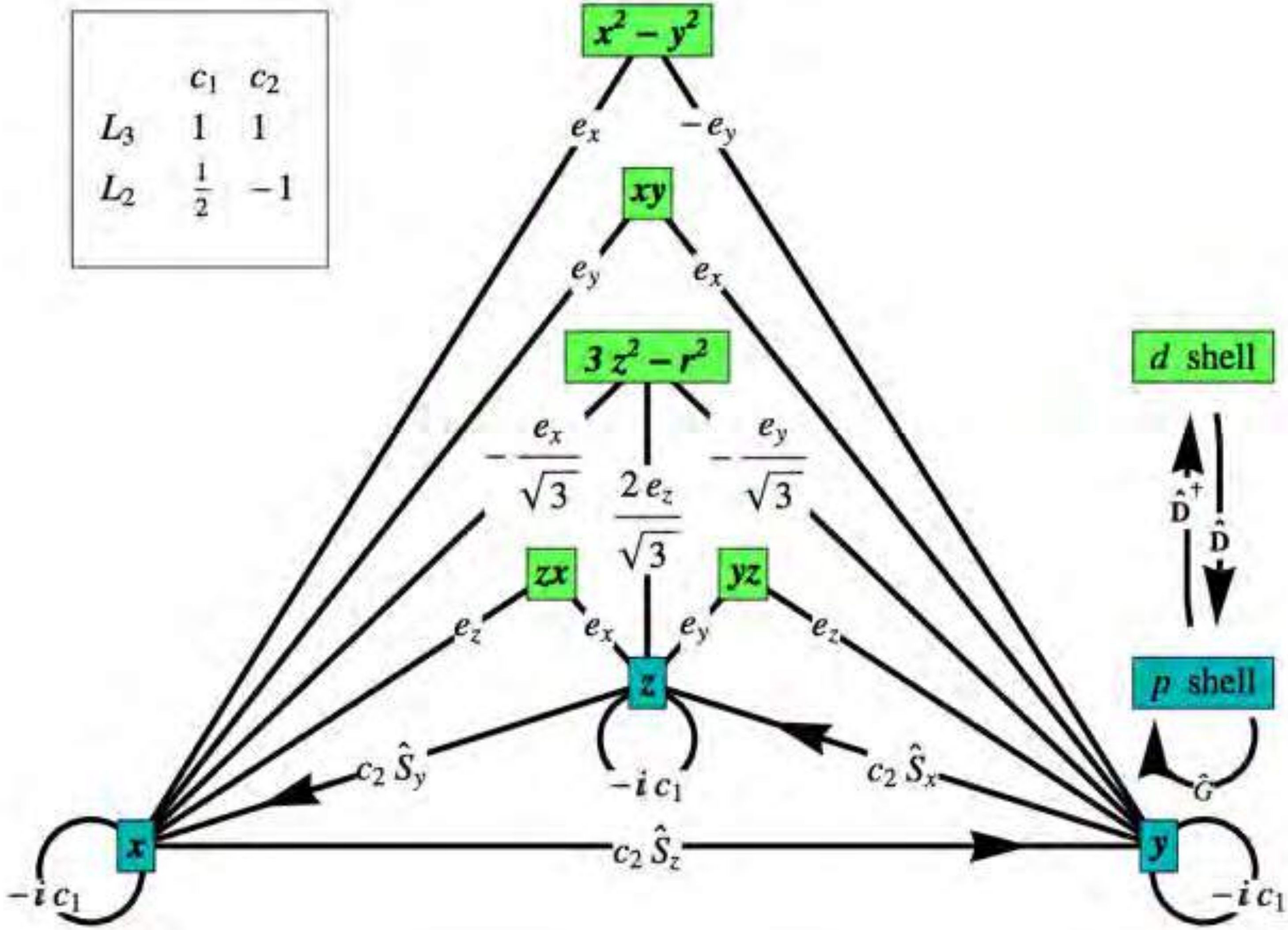
$$G(z_{\mathbf{k}}) = \frac{1}{z_{\mathbf{k}} - H} = \sum_n \frac{|n\rangle\langle n|}{z_{\mathbf{k}} - E_n}$$

$\backsimeq$ *constant*

RIXS response governed by (dipole) transition operators

RIXS amplitude @ transition metal L-edge

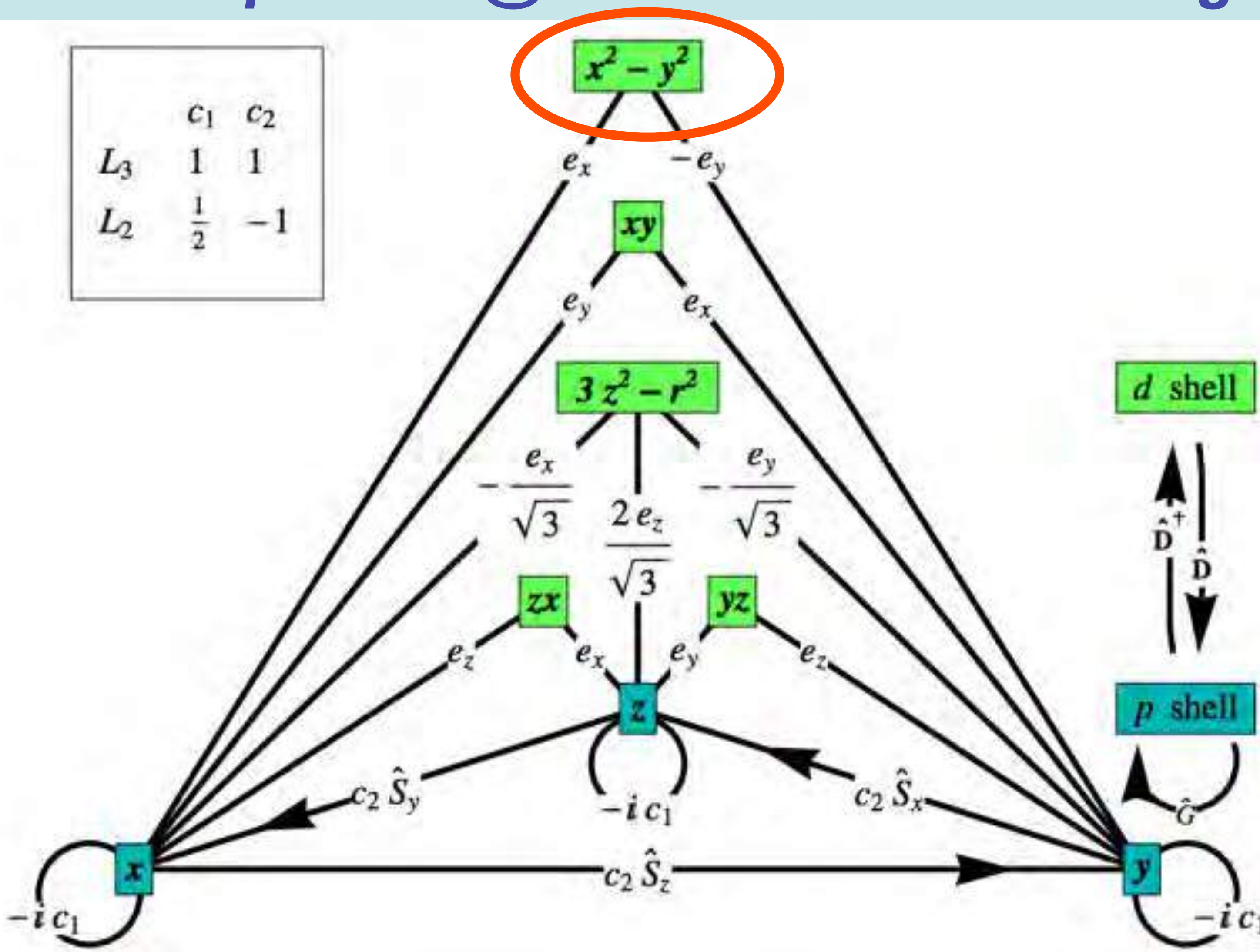
	c_1	c_2
L_3	1	1
L_2	$\frac{1}{2}$	-1



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

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

RIXS amplitude @ transition metal L-edge



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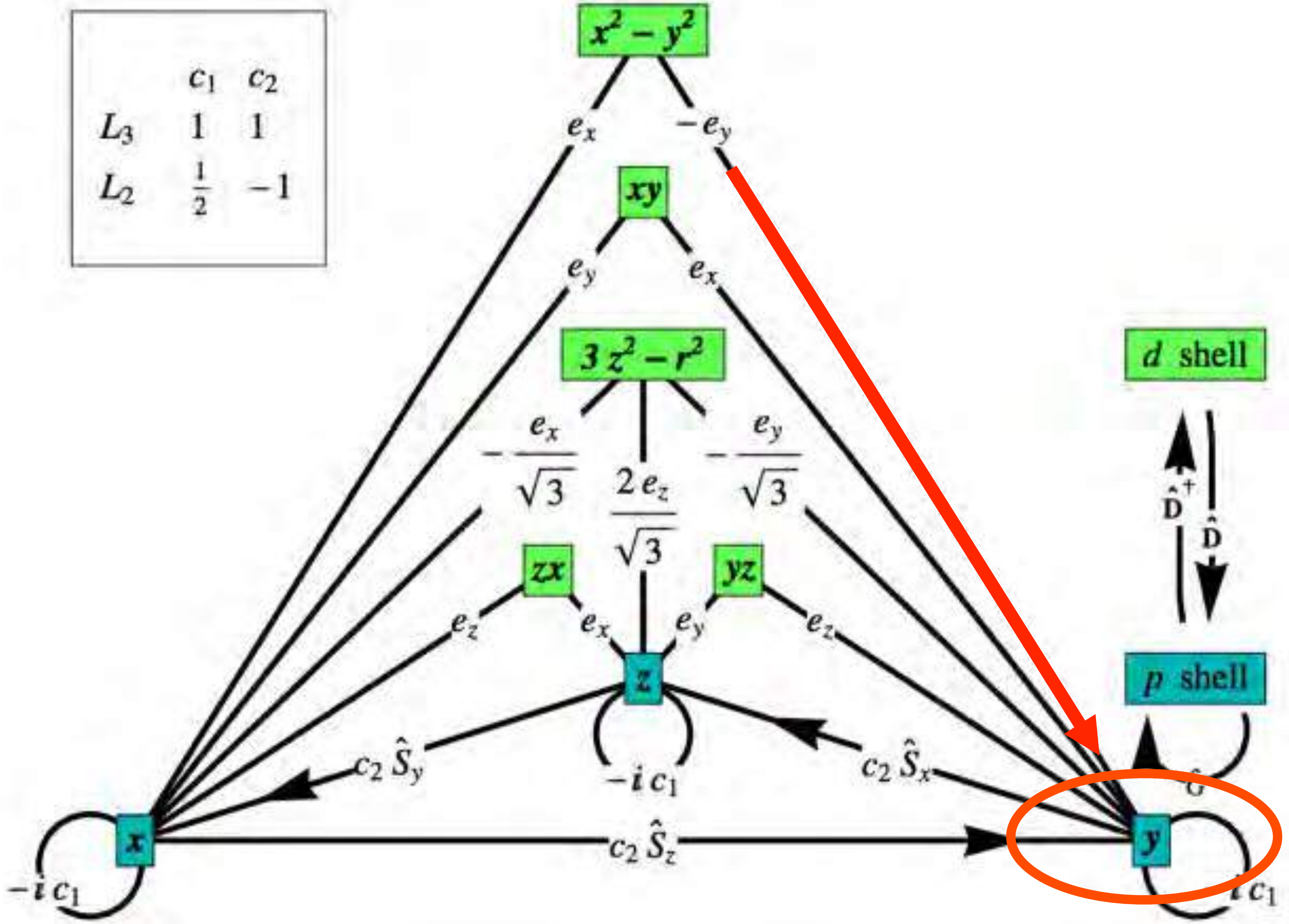

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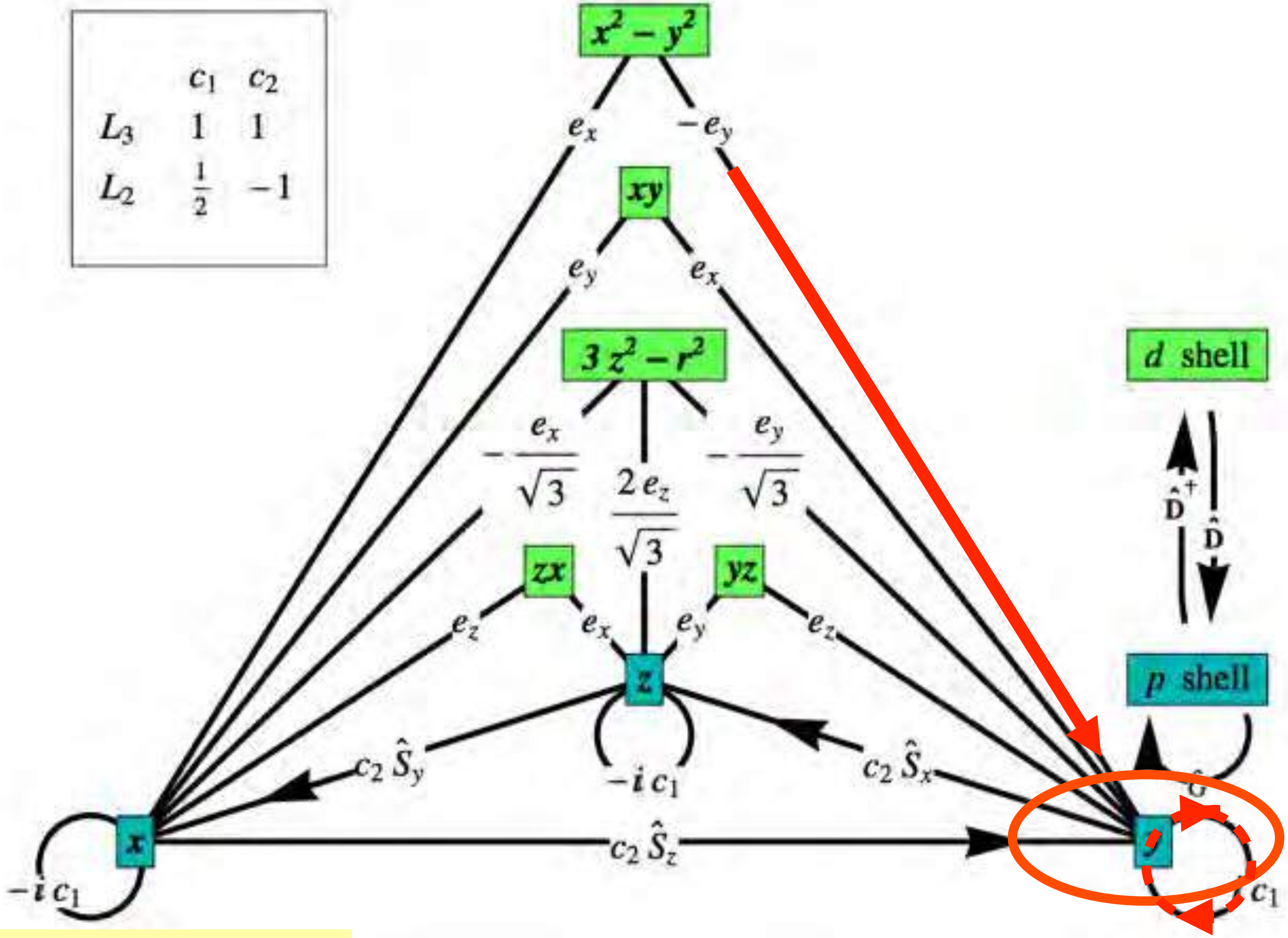

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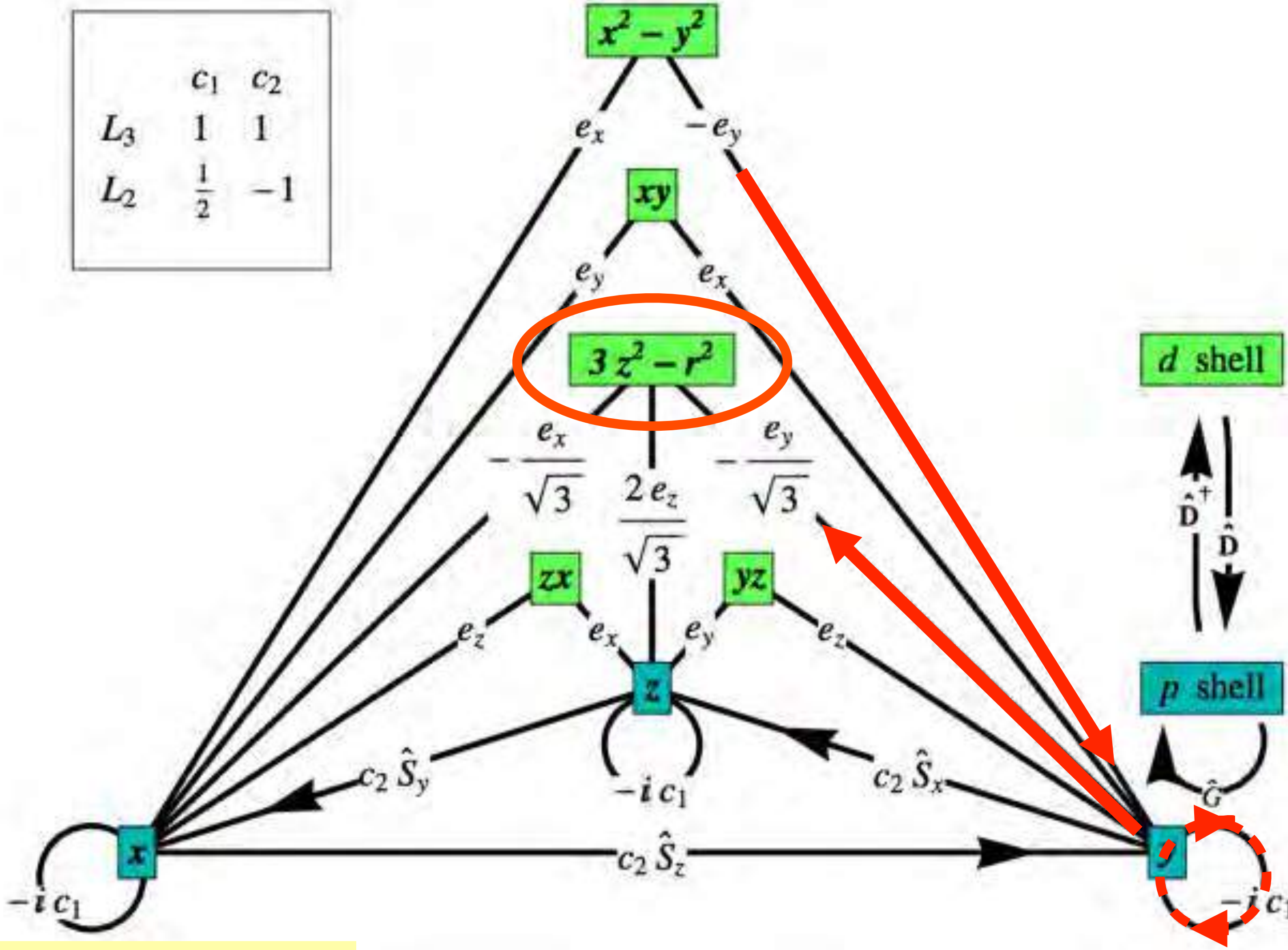
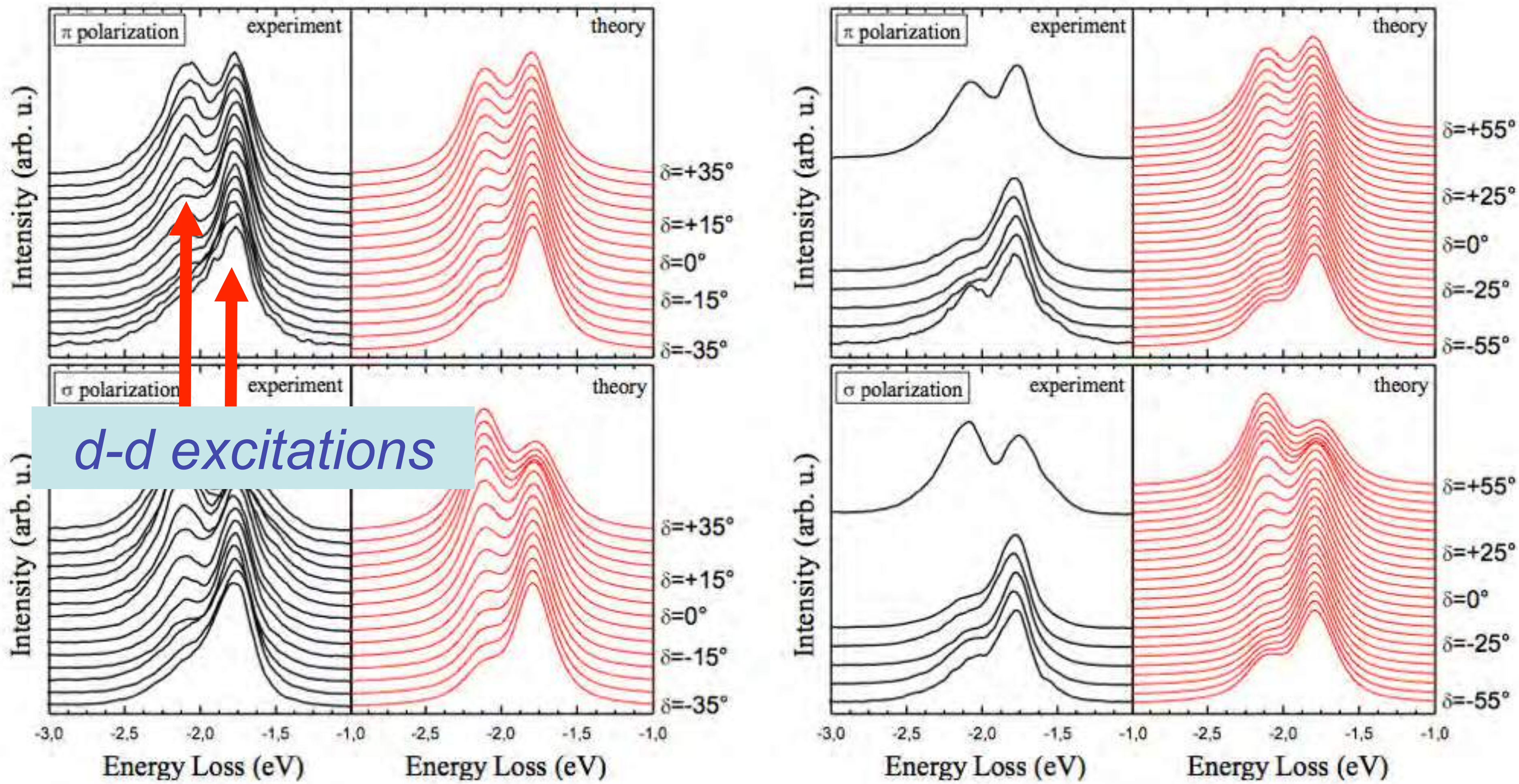


Ament, Ghiringhelli, Moretti, Braicovich & JvdB, PRL 103, 117003 (2009)

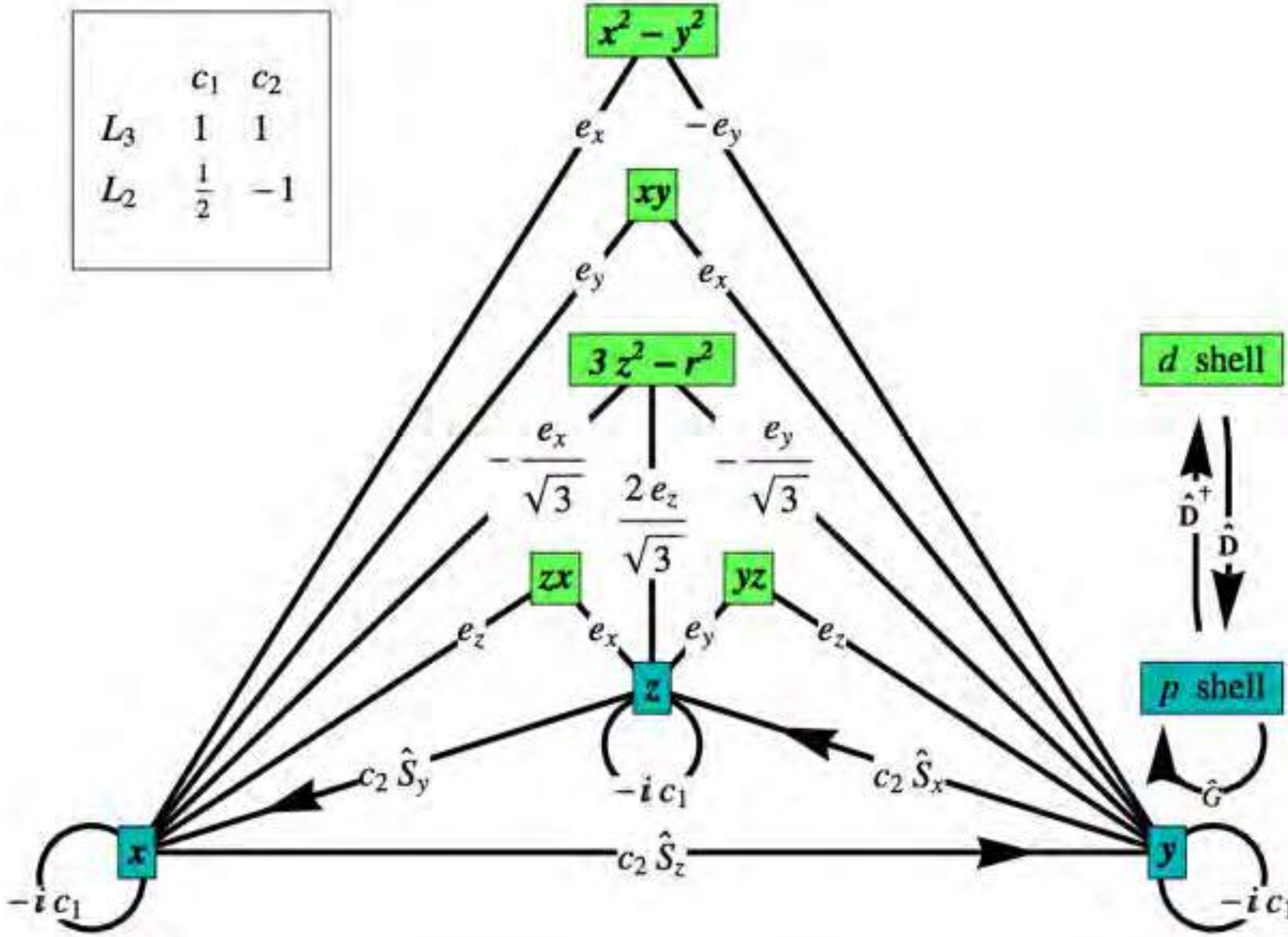
Marra, Wohlfeld & JvdB, PRL 109, 117401 (2012)

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)

RIXS spin-flip amplitude @ transition metal L-edge



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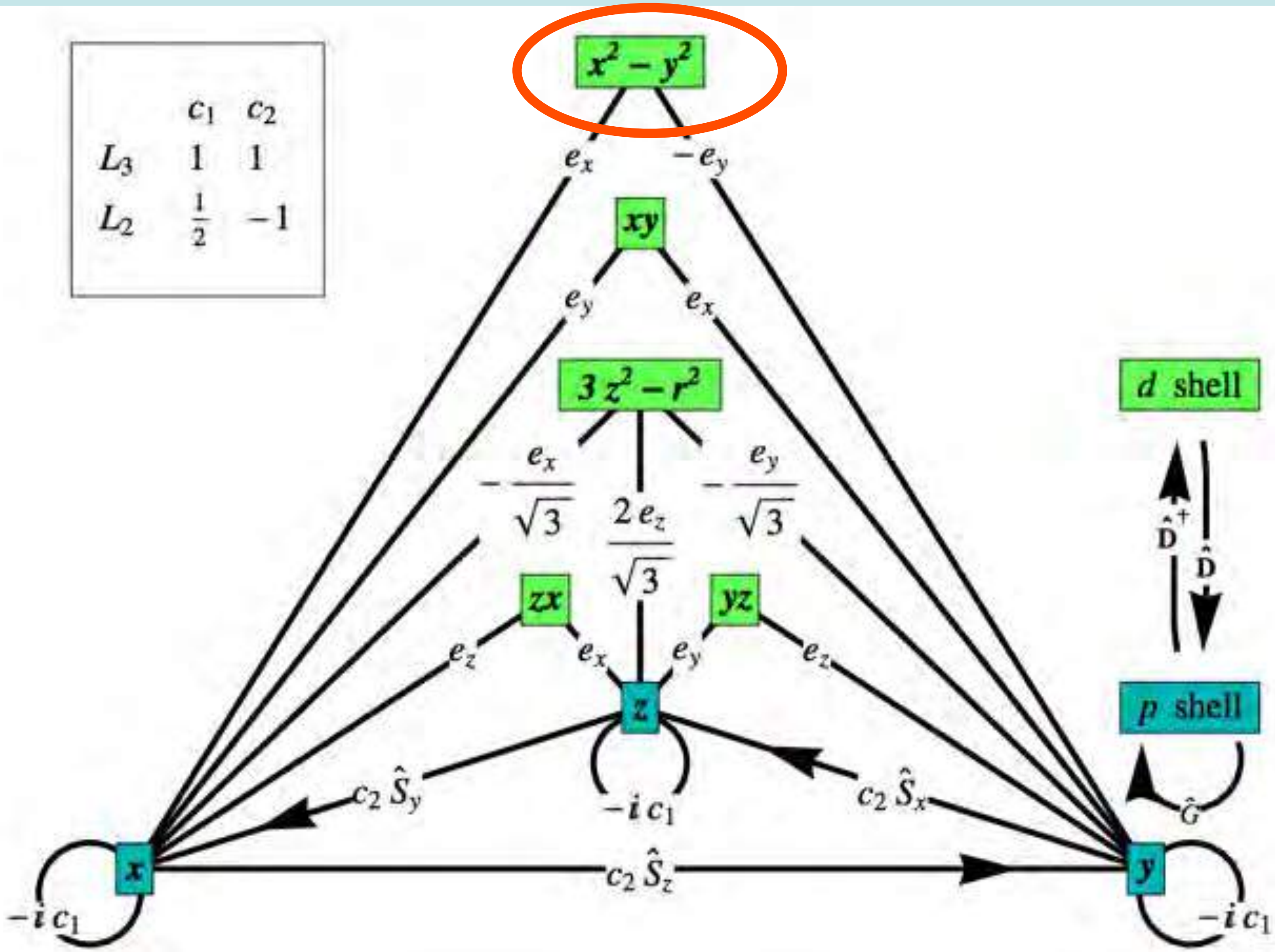

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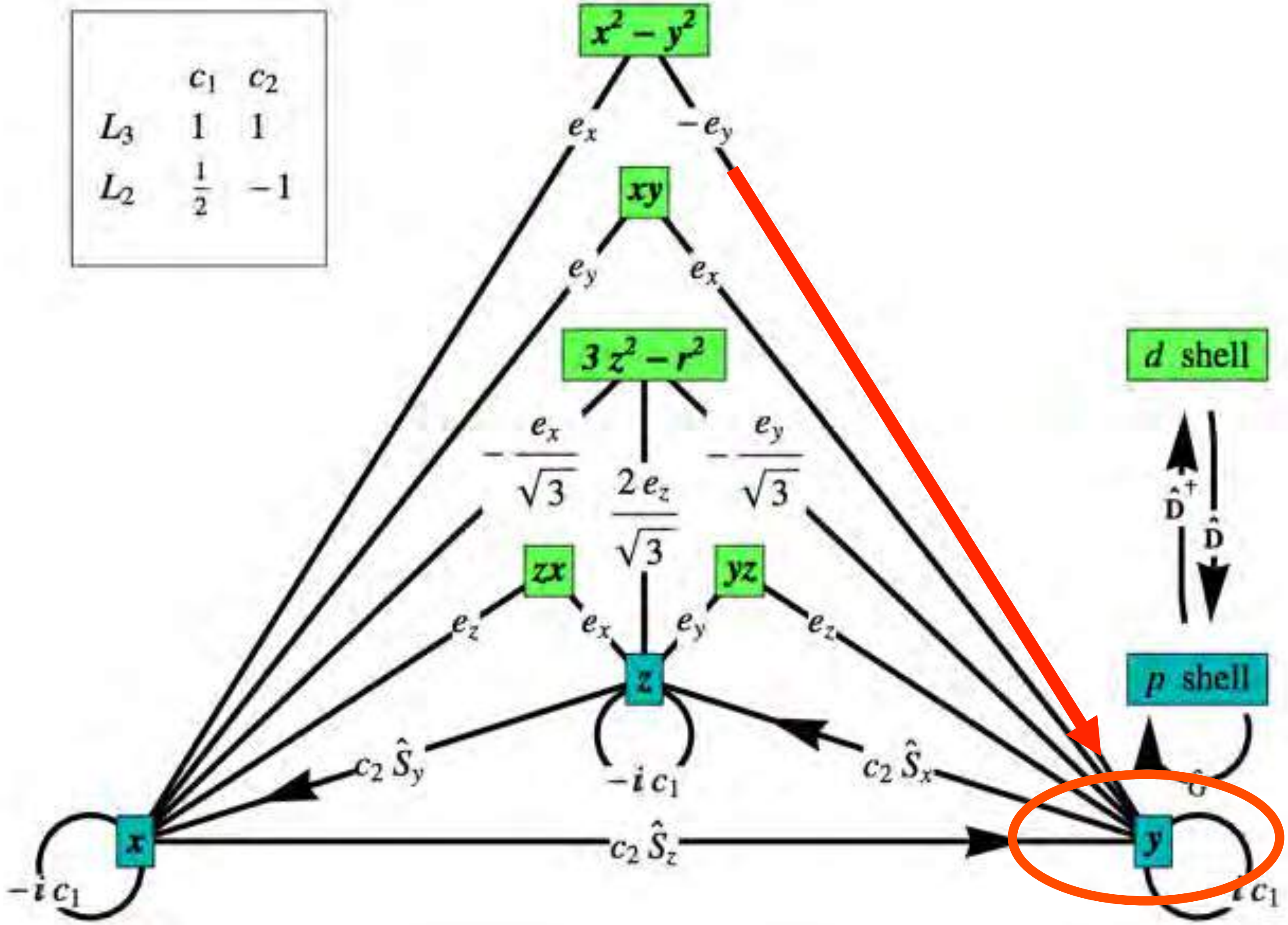

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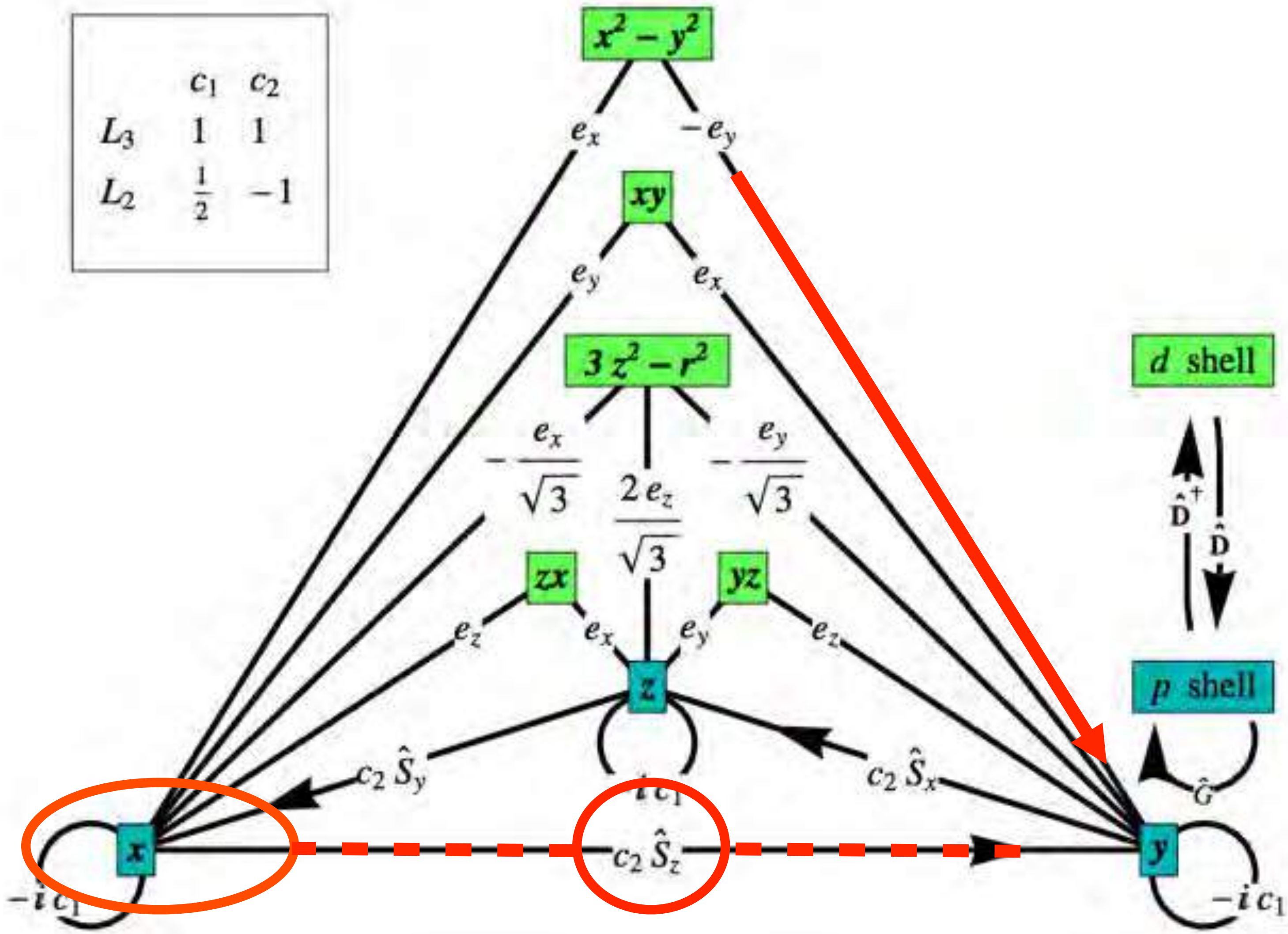

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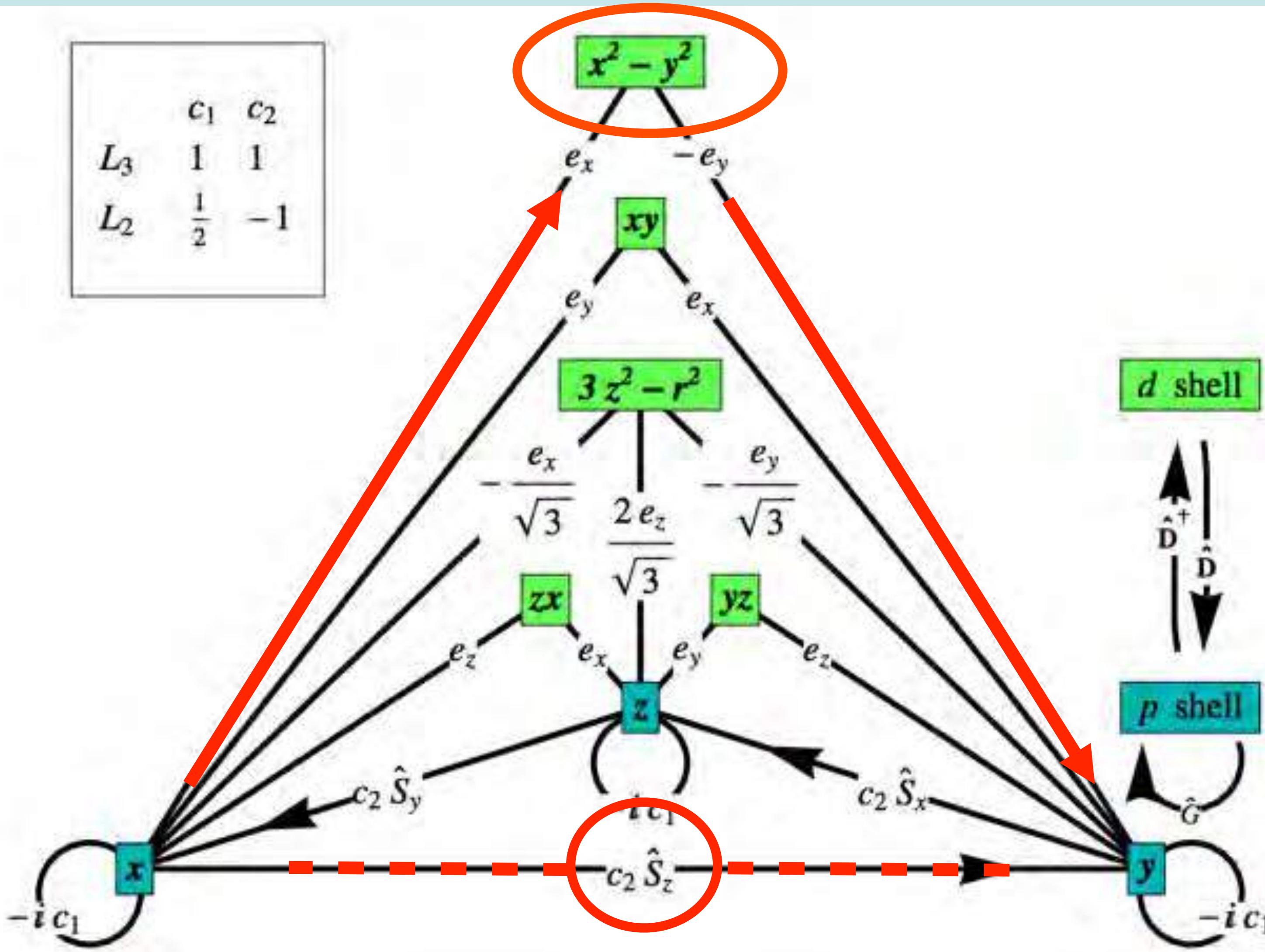
Marra, Wohlfeld & JvdB, PRL 109, 117401 (2012)

RIXS spin-flip amplitude @ transition metal L-edge



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

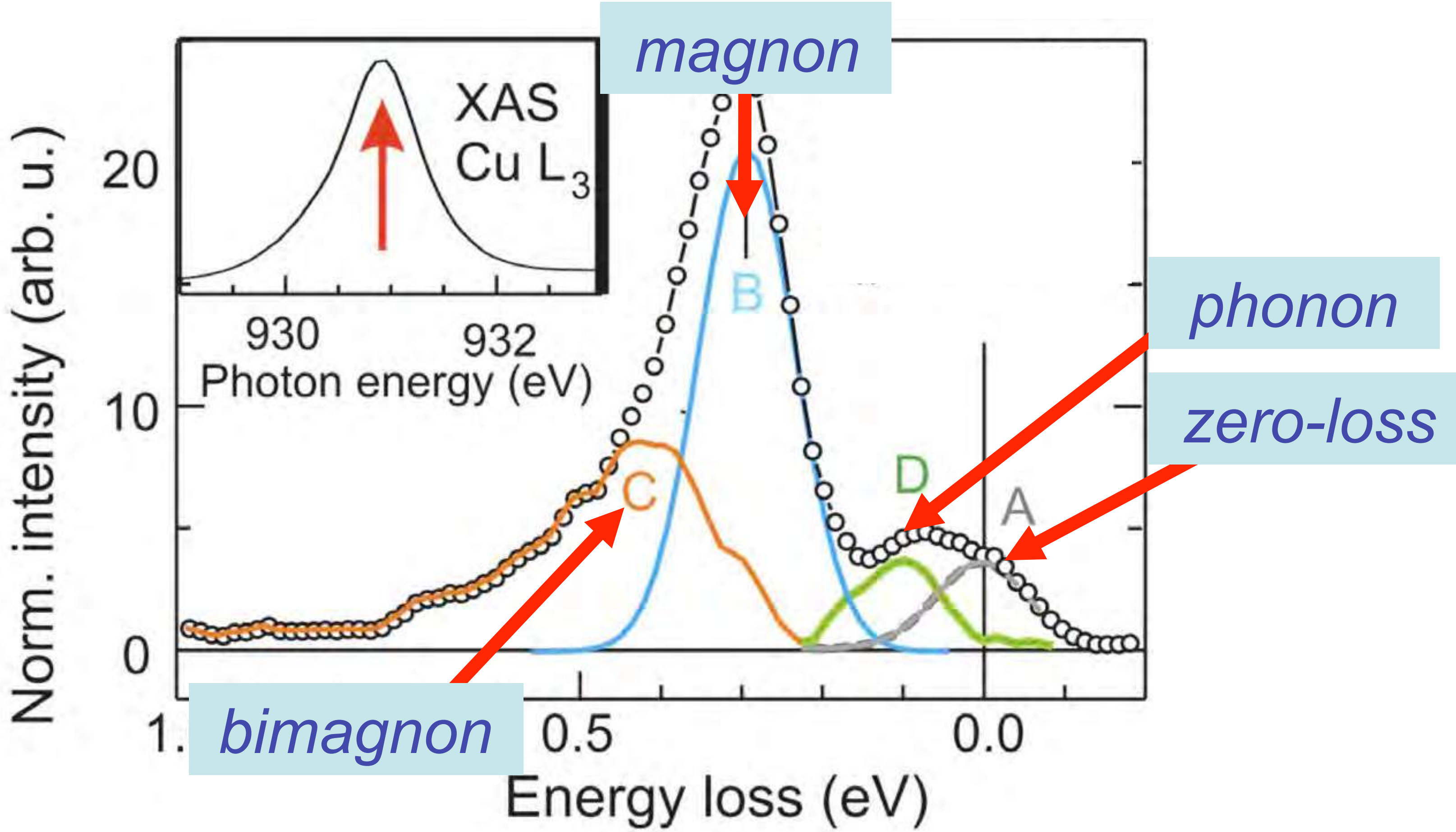
x^2-y^2 spin *NOT // z*:
pure spin flip

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

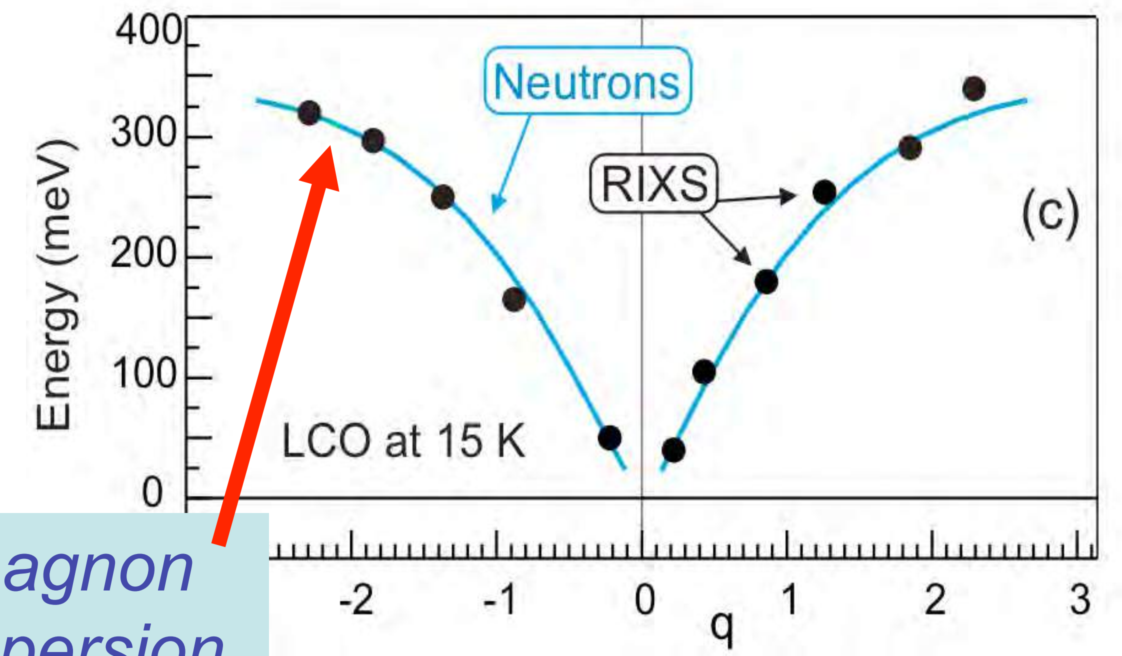
Magnetic RIXS on La_2CuO_4 @ Cu L-edge

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

CuO's are such special cases...



Magnetic direct RIXS on La_2CuO_4 @ Cu L-edge

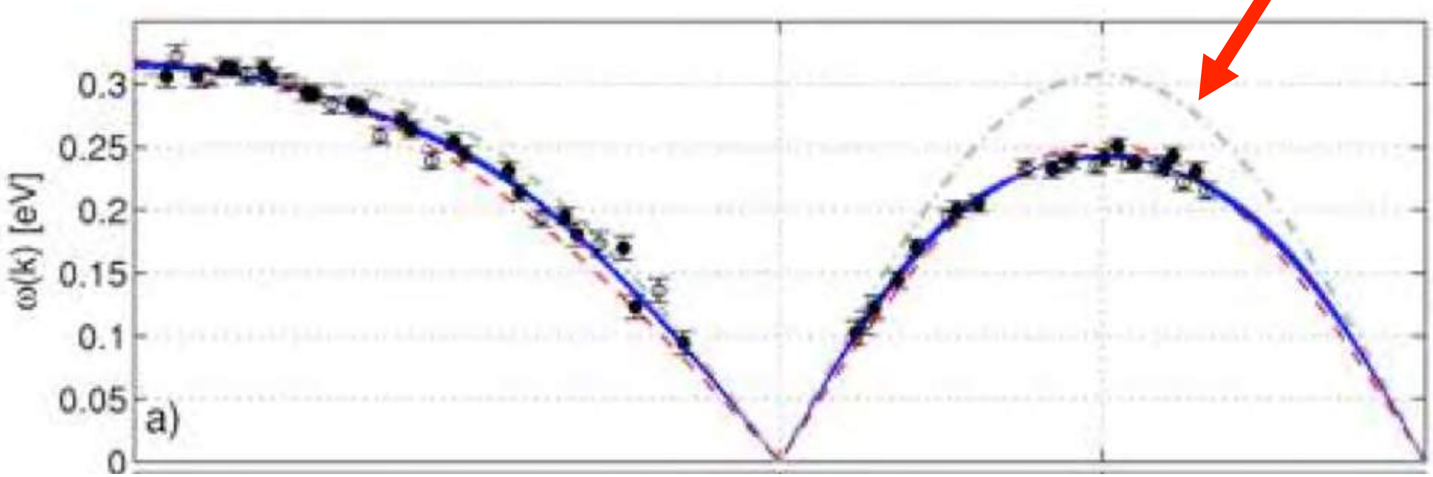


magnon dispersion

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

RIXS magnon dispersion of $Sr_2CuO_2Cl_2$

deviation from simple Heisenberg

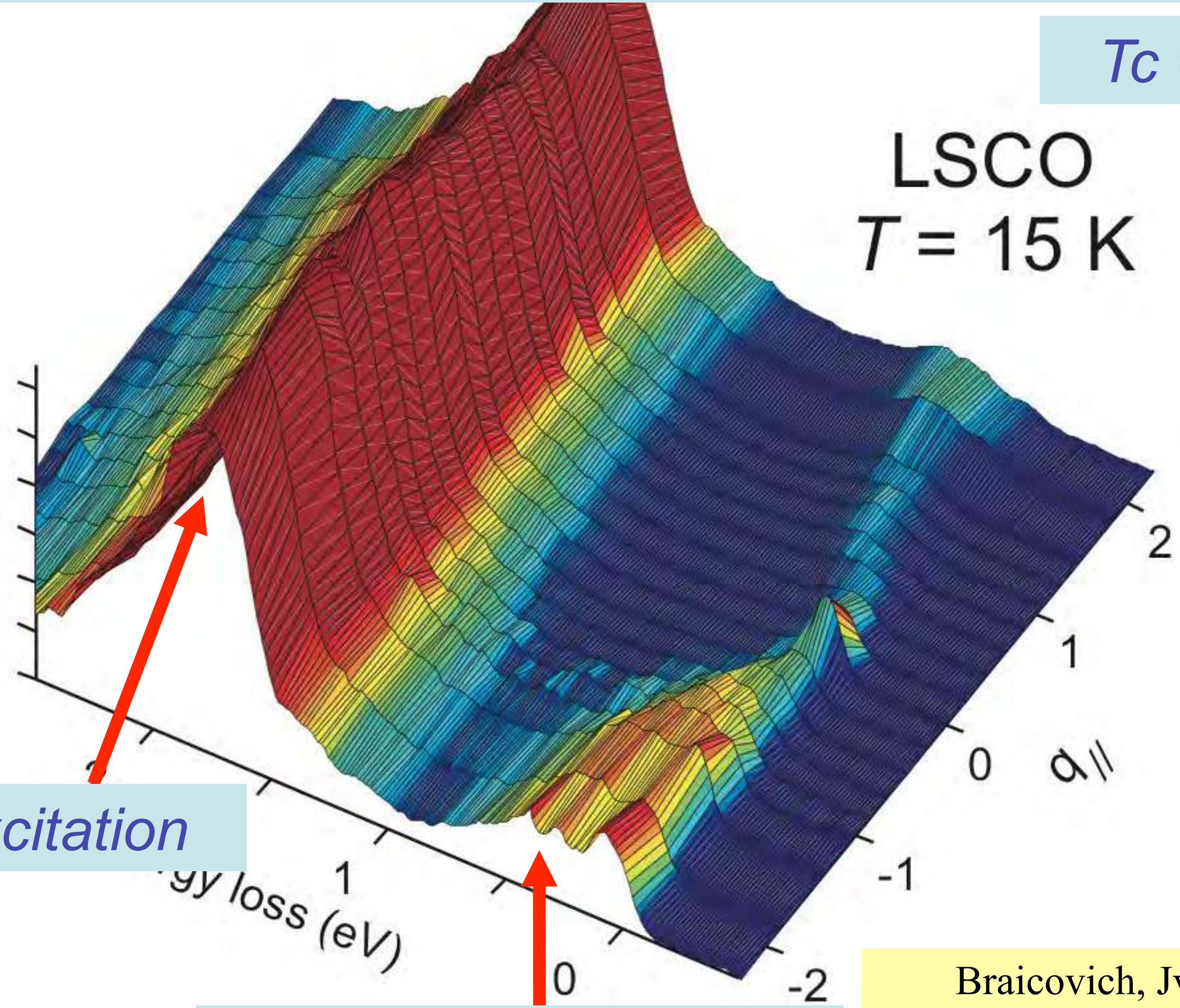


—> transferred momentum q

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

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

Tc = 21K



d-d excitation

paramagnon

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

nature physics

ARTICLES

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

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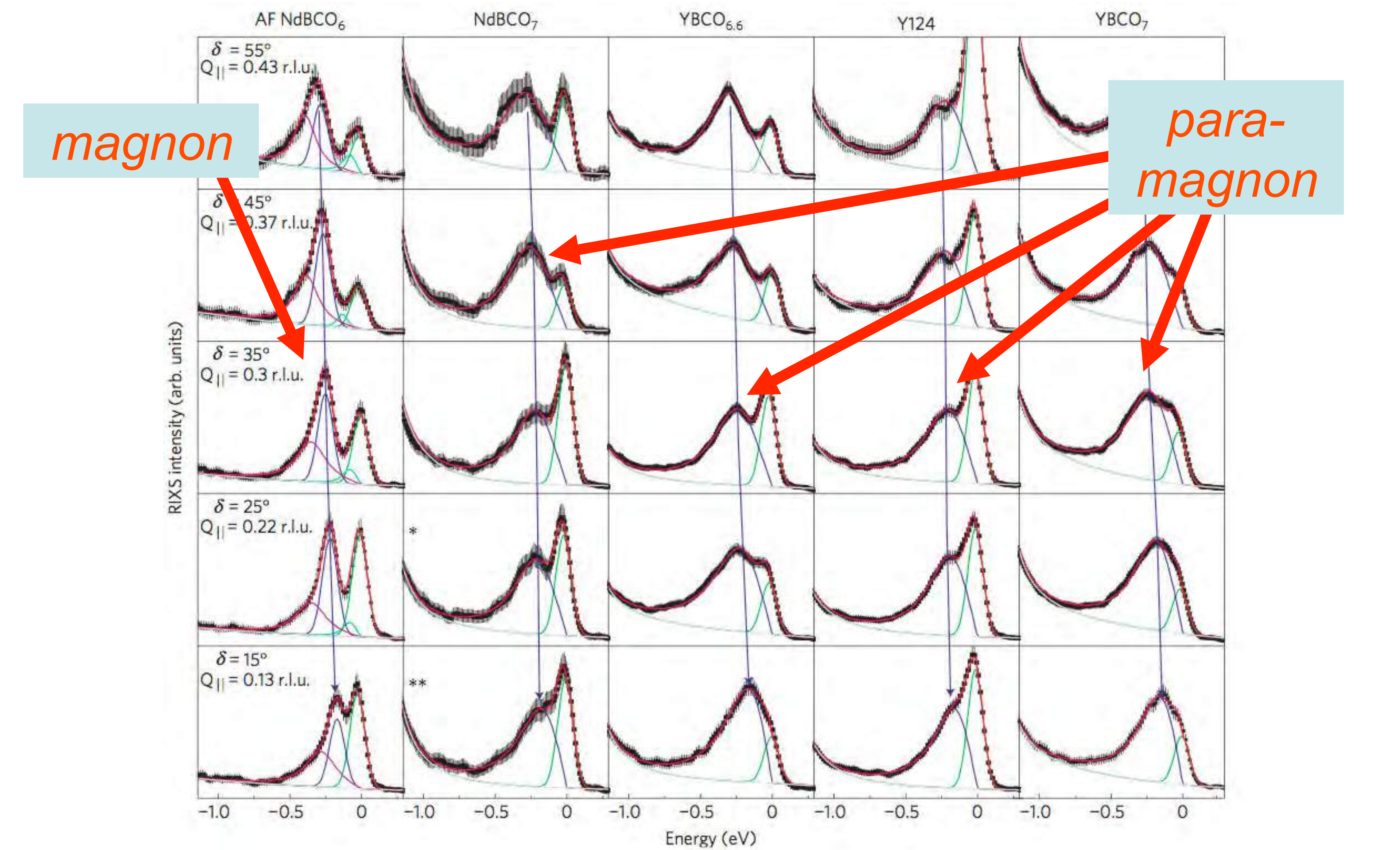

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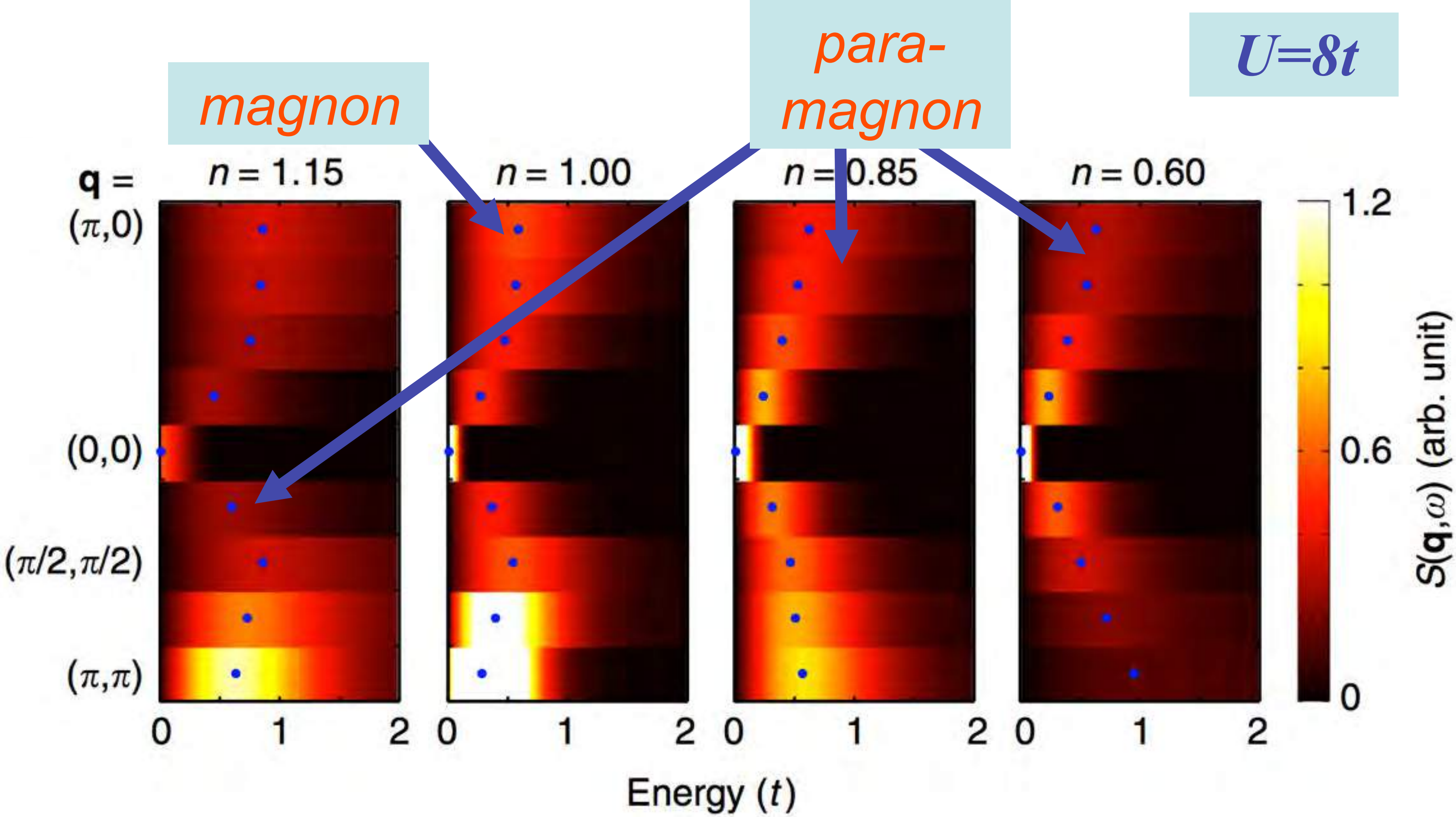
Dynamical structure factor Hubbard model, QMC



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

Summary part 2

- 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

RIXS amplitude/intensity

Interaction light & matter

RIXS amplitude F and intensity I

Intensity

Amplitude

$$I(\omega, \mathbf{k}, \mathbf{k}', \boldsymbol{\epsilon}, \boldsymbol{\epsilon}') = \sum_f |\mathcal{F}_{fg}(\mathbf{k}, \mathbf{k}', \boldsymbol{\epsilon}, \boldsymbol{\epsilon}', \omega_{\mathbf{k}})|^2 \times \delta(E_f + \hbar\omega_{\mathbf{k}'} - E_g - \hbar\omega_{\mathbf{k}})$$

ω_{loss}

momentum in

out

$q_{loss}=k'-k$

RIXS amplitude F and intensity I

polarization in

out

Intensity

Amplitude

$$I(\omega, \mathbf{k}, \mathbf{k}', \boldsymbol{\epsilon}, \boldsymbol{\epsilon}') = \sum_f |\mathcal{F}_{fg}(\mathbf{k}, \mathbf{k}', \boldsymbol{\epsilon}, \boldsymbol{\epsilon}', \omega_{\mathbf{k}})|^2$$
$$\times\, \delta(E_f + \hbar\omega_{\mathbf{k}'} - E_g - \hbar\omega_{\mathbf{k}})$$

ω_{loss}

momentum in

out

energy conservation

final energy

initial energy

ω_{out}

ω_{in}

Interaction of light and matter: Hamiltonian

kinetic

Zeeman

$$H = \sum_{i=1}^{N} \Big[\frac{(\mathbf{p}_i + e\mathbf{A}(\mathbf{r}_i))^2}{2m} + \frac{e\hbar}{2m} \boldsymbol{\sigma}_i \cdot \mathbf{B}(\mathbf{r}_i) + \frac{e\hbar}{2(2mc)^2} \times$$
$$\boldsymbol{\sigma}_i \cdot \Big(\mathbf{E}(\mathbf{r}_i) \times (\mathbf{p}_i + e\mathbf{A}(\mathbf{r}_i)) - (\mathbf{p}_i + e\mathbf{A}(\mathbf{r}_i)) \times \mathbf{E}(\mathbf{r}_i) \Big) \Big]$$
$$+ \frac{e\hbar^2 \rho(\mathbf{r}_i)}{8(mc)^2 \epsilon_0} + H_{\text{Coulomb}} + \sum_{\boldsymbol{\kappa},\boldsymbol{\varepsilon}} \hbar\omega_{\boldsymbol{\kappa}} \left(a^\dagger_{\boldsymbol{\kappa}\boldsymbol{\varepsilon}} a_{\boldsymbol{\kappa}\boldsymbol{\varepsilon}} + \frac{1}{2} \right)$$

spin-orbit coupling

Darwin

free photons

electron-nucleus
electron-electron

vector potential

plane wave

$$\mathbf{A}(\mathbf{r}) = \sum_{\boldsymbol{\kappa},\boldsymbol{\varepsilon}} \sqrt{\frac{\hbar}{2\mathcal{V}\epsilon_0\omega_{\boldsymbol{\kappa}}}} \left(\boldsymbol{\varepsilon}\, a_{\boldsymbol{\kappa}\boldsymbol{\varepsilon}} e^{i\boldsymbol{\kappa}\cdot\mathbf{r}} + \boldsymbol{\varepsilon}^* a^\dagger_{\boldsymbol{\kappa}\boldsymbol{\varepsilon}} e^{-i\boldsymbol{\kappa}\cdot\mathbf{r}} \right)$$

Lowest order perturbing Hamiltonian: small A

$$H' = \sum_{i=1}^{N} \left[\frac{e}{m}\mathbf{A}(\mathbf{r}_i)\cdot\mathbf{p}_i + \frac{e^2}{2m}\mathbf{A}^2(\mathbf{r}_i) + \frac{e\hbar}{2m}\boldsymbol{\sigma}_i\cdot\nabla\times\mathbf{A}(\mathbf{r}_i) - \frac{e^2\hbar}{(2mc)^2}\boldsymbol{\sigma}_i\cdot\frac{\partial\mathbf{A}(\mathbf{r}_i)}{\partial t}\times\mathbf{A}(\mathbf{r}_i)\right],$$

small

Fermi Golden Rule, to second order:

transition rate

$$w = \frac{2\pi}{\hbar}\sum_{\mathrm{f}}\left|\langle \mathrm{f}|\, H'\,|\mathrm{g}\rangle + \sum_n \frac{\langle \mathrm{f}|\, H'\,|n\rangle\,\langle n|\, H'\,|\mathrm{g}\rangle}{E_{\mathrm{g}} - E_n}\right|^2 \delta(E_{\mathrm{f}} - E_{\mathrm{g}})$$

2nd Order...

$$H' = \sum_{i=1}^{N} \frac{e}{m}\mathbf{A}(\mathbf{r}_i)\cdot\mathbf{p}_i + \frac{e^2}{2m}\mathbf{A}^2(\mathbf{r}_i) + \frac{e\hbar}{2m}\boldsymbol{\sigma}_i\cdot\nabla\times\mathbf{A}(\mathbf{r}_i)$$

Fermi Golden Rule, to second order:

transition rate

$$w = \frac{2\pi}{\hbar}\sum_{\mathrm{f}}\left|\langle \mathrm{f}|\,H'\,|\mathrm{g}\rangle + \sum_{n}\frac{\langle \mathrm{f}|\,H'\,|n\rangle\,\langle n|\,H'\,|\mathrm{g}\rangle}{E_{\mathrm{g}}-E_n}\right|^2\delta(E_{\mathrm{f}}-E_{\mathrm{g}})$$

2nd Order...

$$H' = \sum_{i=1}^{N} \frac{e}{m}\mathbf{A}(\mathbf{r}_i)\cdot\mathbf{p}_i + \frac{e\hbar}{2m}\boldsymbol{\sigma}_i \cdot \nabla \times \mathbf{A}(\mathbf{r}_i)$$

Fermi Golden Rule, to second order:

transition rate

$$w = \frac{2\pi}{\hbar}\sum_{\mathrm{f}}\left|\langle \mathrm{f}|\, H'\,|\mathrm{g}\rangle + \sum_{n}\frac{\langle \mathrm{f}|\, H'\,|n\rangle\,\langle n|\, H'\,|\mathrm{g}\rangle}{E_{\mathrm{g}} - E_n}\right|^2 \delta(E_{\mathrm{f}} - E_{\mathrm{g}})$$

2nd Order: Resonant Scattering I

RIXS amplitude

$$\frac{e^2\hbar}{2m^2\mathcal{V}\epsilon_0\sqrt{\omega_{\mathbf{k}}\omega_{\mathbf{k}'}}}\sum_n\sum_{i,j=1}^{N}$$

$$\times\frac{\langle f|\, e^{-i\mathbf{k}'\cdot\mathbf{r}_i}\left(\boldsymbol{\epsilon}'^*\cdot\mathbf{p}_i-\frac{i\hbar}{2}\boldsymbol{\sigma}_i\cdot\mathbf{k}'\times\boldsymbol{\epsilon}'^*\right)|n\rangle}{E_g+\hbar\omega_{\mathbf{k}}-E_n+i\Gamma_n}$$

$$\times\langle n|\, e^{i\mathbf{k}\cdot\mathbf{r}_j}\left(\boldsymbol{\epsilon}\cdot\mathbf{p}_j+\frac{i\hbar}{2}\boldsymbol{\sigma}_j\cdot\mathbf{k}\times\boldsymbol{\epsilon}\right)|g\rangle$$

small

RIXS transition operator

$$\mathcal{D}=\frac{1}{im\omega_{\mathbf{k}}}\sum_{i=1}^{N}e^{i\mathbf{k}\cdot\mathbf{r}_i}\boldsymbol{\epsilon}\cdot\mathbf{p}_i,$$

RIXS amplitude

$$\mathcal{F}_{fg}(\mathbf{k},\mathbf{k}',\boldsymbol{\epsilon},\boldsymbol{\epsilon}',\omega_{\mathbf{k}},\omega_{\mathbf{k}'})=\sum_n\frac{\langle f|\,\mathcal{D}'^{\dagger}\,|n\rangle\,\langle n|\,\mathcal{D}\,|g\rangle}{E_g+\hbar\omega_{\mathbf{k}}-E_n+i\Gamma_n}$$

Kramers-Heisenberg expression

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

2nd Order: Resonant Scattering II

RIXS amplitude

$$\mathcal{F}_{fg}(\mathbf{k},\mathbf{k}',\boldsymbol{\epsilon},\boldsymbol{\epsilon}',\omega_{\mathbf{k}},\omega_{\mathbf{k}'}) = \sum_n \frac{\langle f|\,\mathcal{D}'^{\dagger}\,|n\rangle\,\langle n|\,\mathcal{D}\,|g\rangle}{E_g + \hbar\omega_{\mathbf{k}} - E_n + i\Gamma_n}$$

RIXS intensity:

$$I(\omega,\mathbf{k},\mathbf{k}',\boldsymbol{\epsilon},\boldsymbol{\epsilon}') = r_e^2 m^2 \omega_{\mathbf{k}'}^3 \omega_{\mathbf{k}} \sum_{\mathbf{f}} |\mathcal{F}_{fg}(\mathbf{k},\mathbf{k}',\boldsymbol{\epsilon},\boldsymbol{\epsilon}',\omega_{\mathbf{k}},\omega_{\mathbf{k}'})|^2 \times \delta(E_g - E_f + \hbar\omega),$$

This expression is essentially exact (non-relativistic limit)