The Abdus Salam
International Centre
for Theoretical Physics

School on Synchrotron and Free-Electron-Laser Based Methods: Multidisciplinary Applications and Perspectives | (smr 3202) | 7-18 May 2018

High resolution RIXS: introduction and applications to strongly correlated systems

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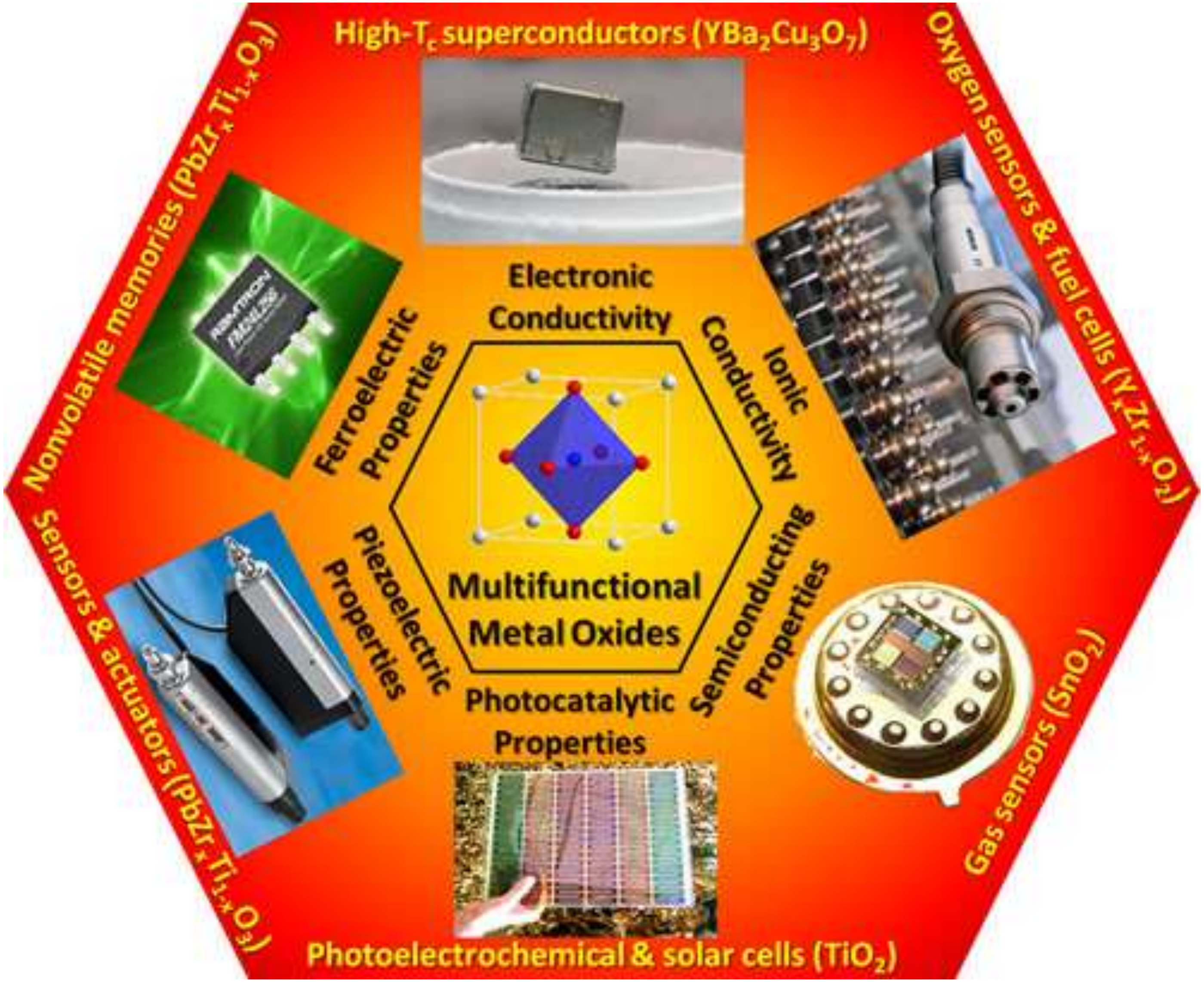
9 May, 2018

Summary

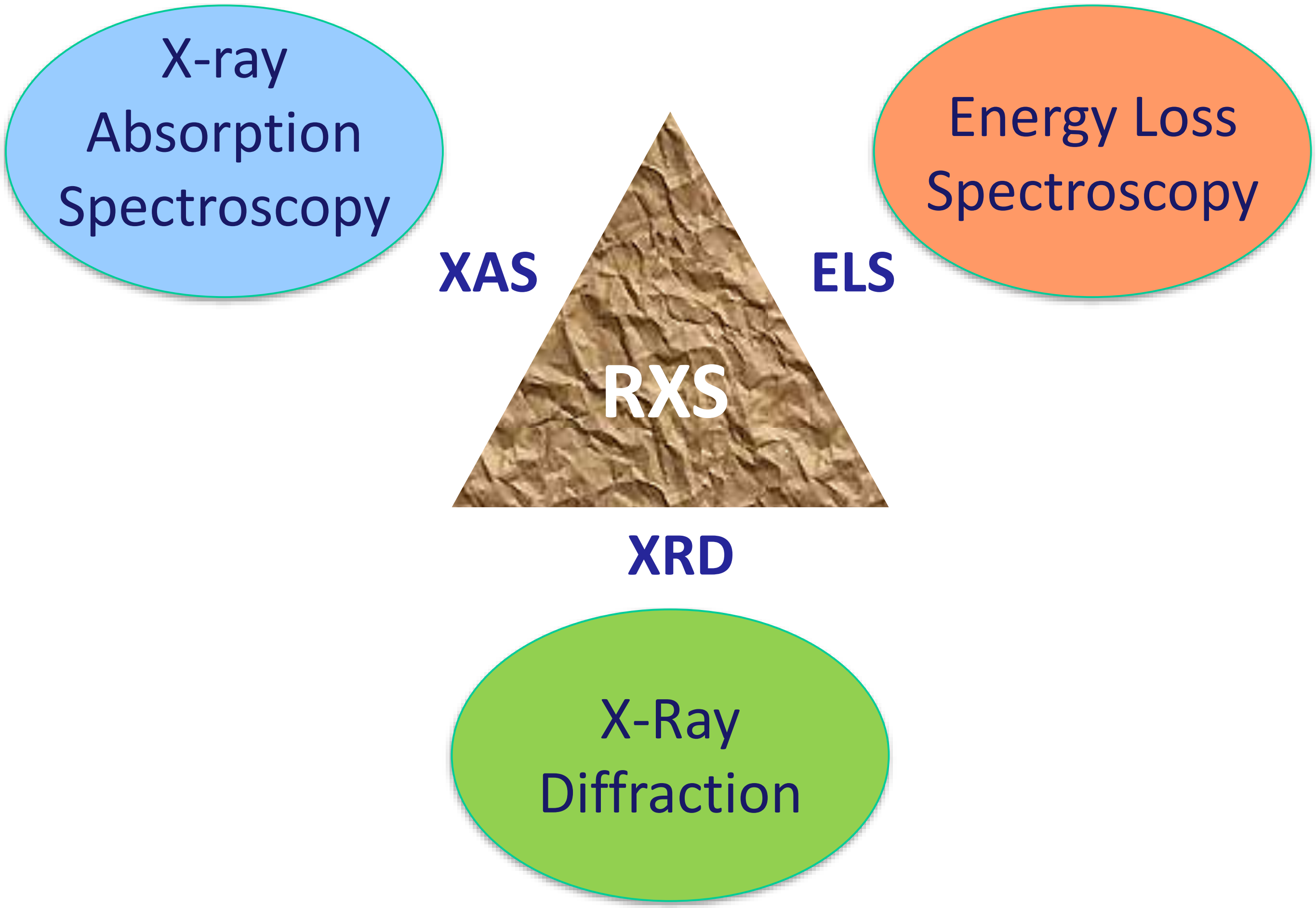
- Introducing resonant x-ray inelastic scattering
- *dd* excitations
- Cu L_3 RIXS and spin excitations in cuprates

Transition metal oxides

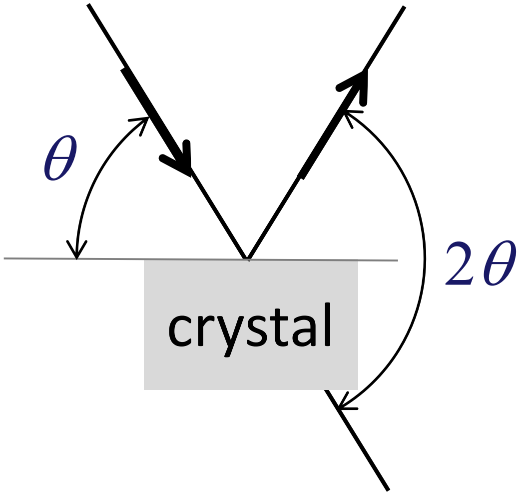


matwww.technion.ac.il

Introduction to Resonant X-ray Scattering



From XRD to X-ray Scattering



X-Ray Diffraction

$|\mathbf{k}'|=|\mathbf{k}|$

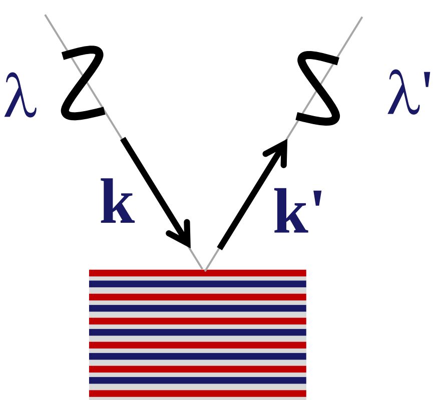
$\lambda = \lambda'$

X-Ray Scattering

$|\mathbf{k}'|=|\mathbf{k}|$

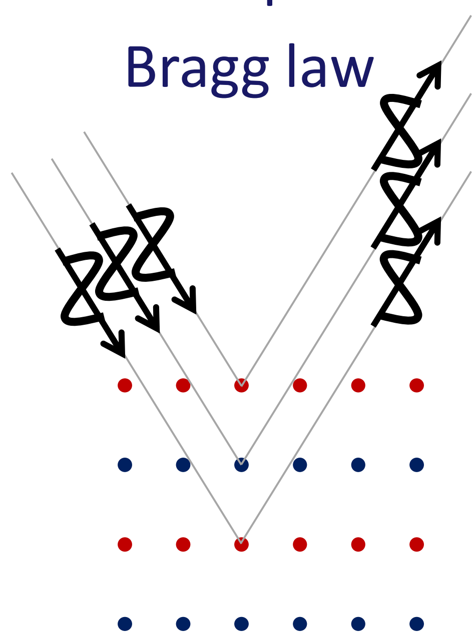
$\lambda = \lambda'$

$\mathbf{q}=\mathbf{G}$



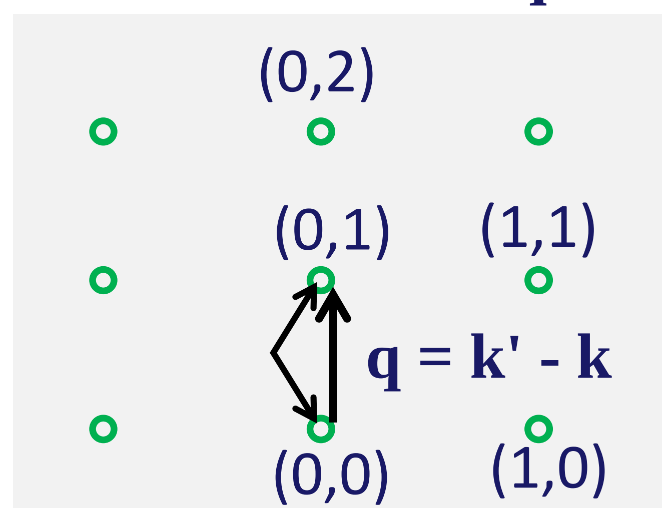
Real space

Bragg law



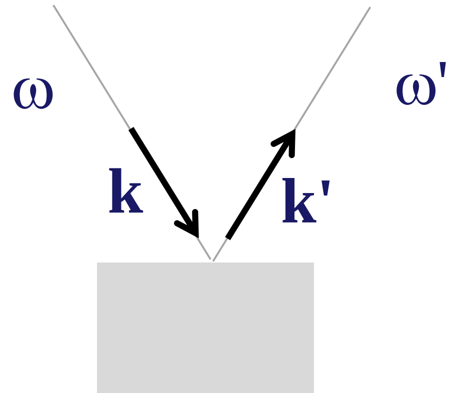
Reciprocal lattice

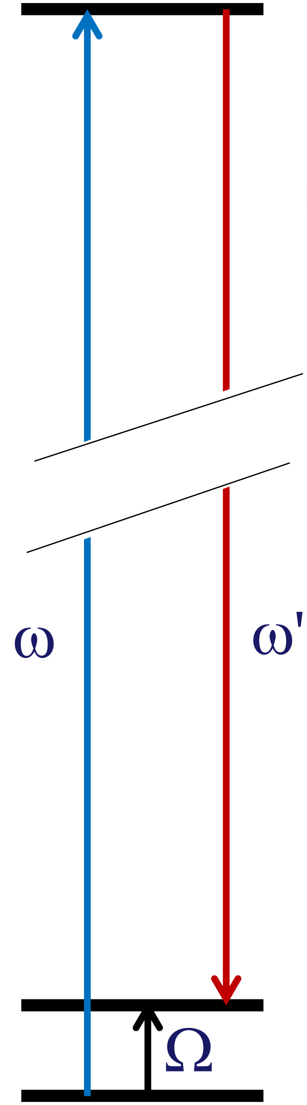
Laue condition: $\mathbf{q}=\mathbf{G}$



ELS: from Raman to Inelastic X-ray Scattering

Energy Loss Spectroscopy

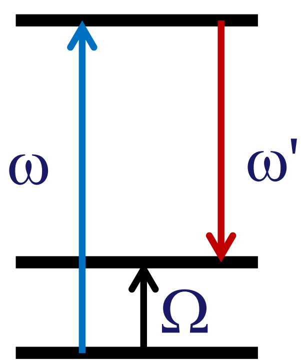




Inelastic X-ray Scattering

Raman light scattering

$k \approx 0$, $q \approx 0$,
$\Omega = \omega - \omega'$



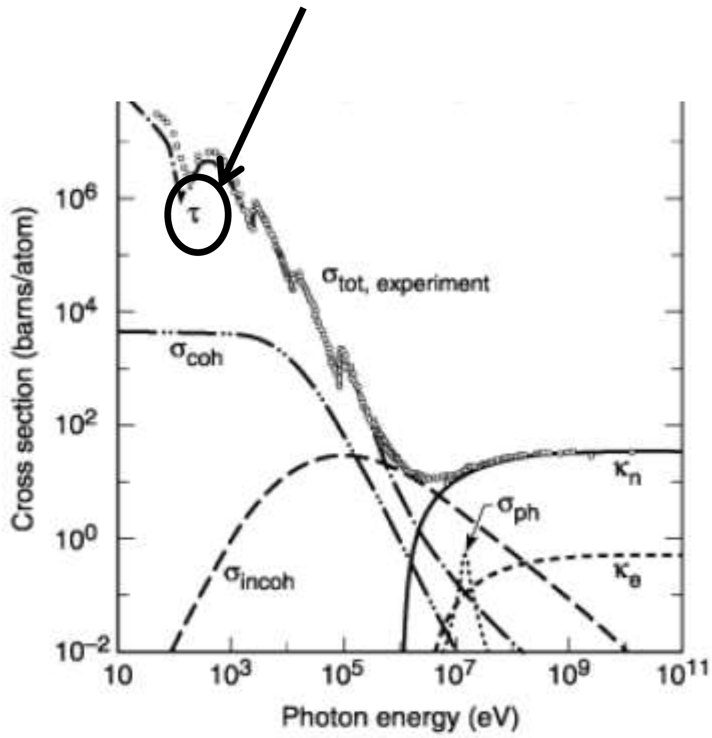
X-ray photons

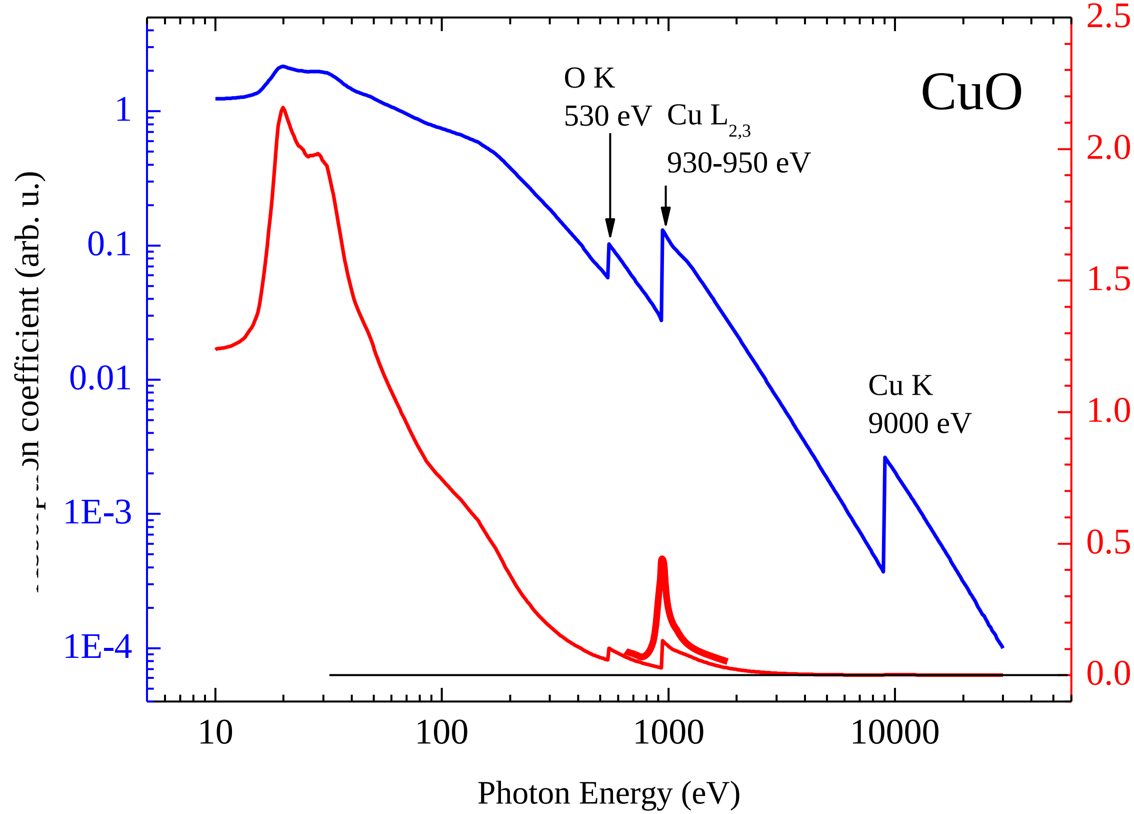
$\Omega = \omega - \omega'$

$\mathbf{q} = \mathbf{k}' - \mathbf{k}$

Resonant X-ray Absorption

Photoelectric effect dominates x-ray absorption below 100,000 eV

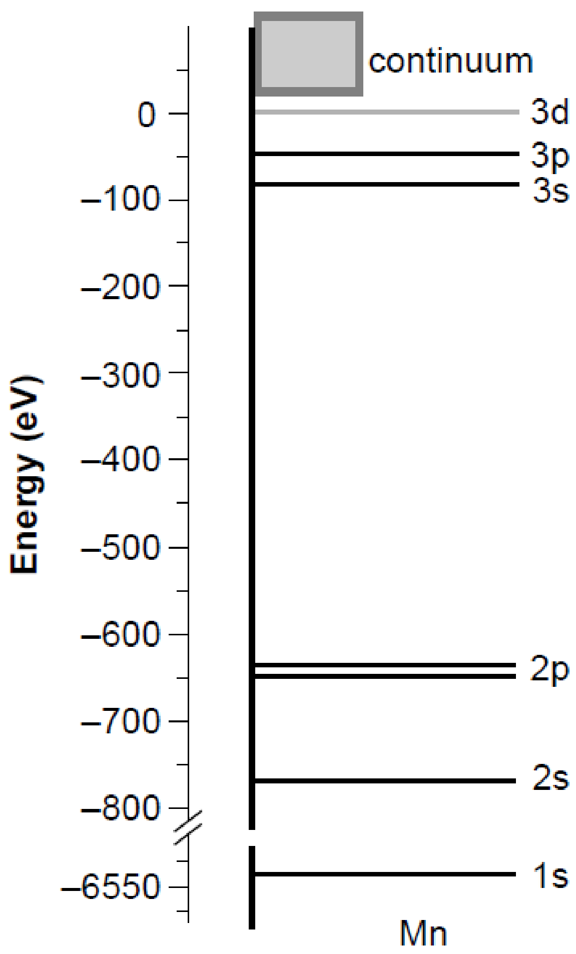


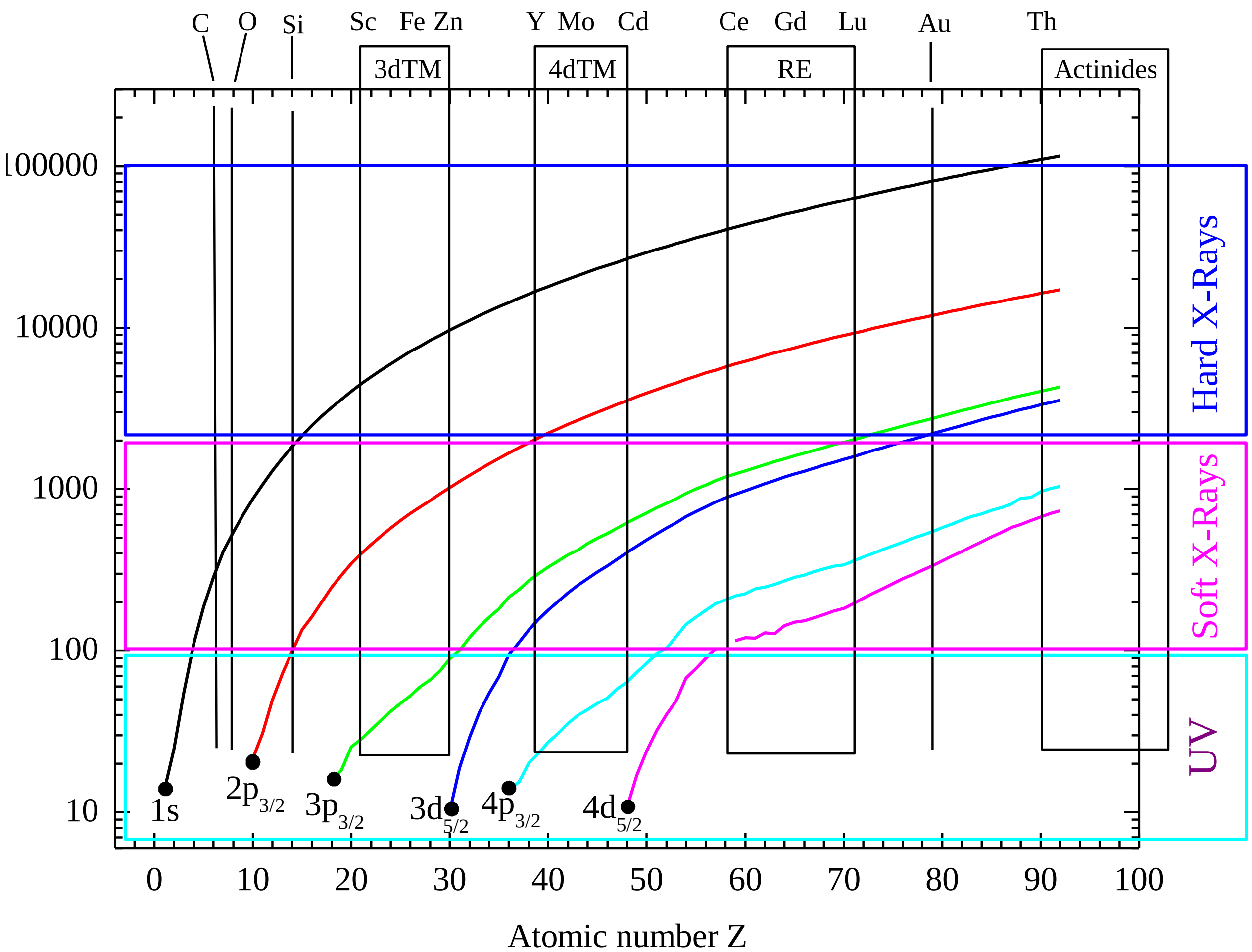


Edges are univocally element specific

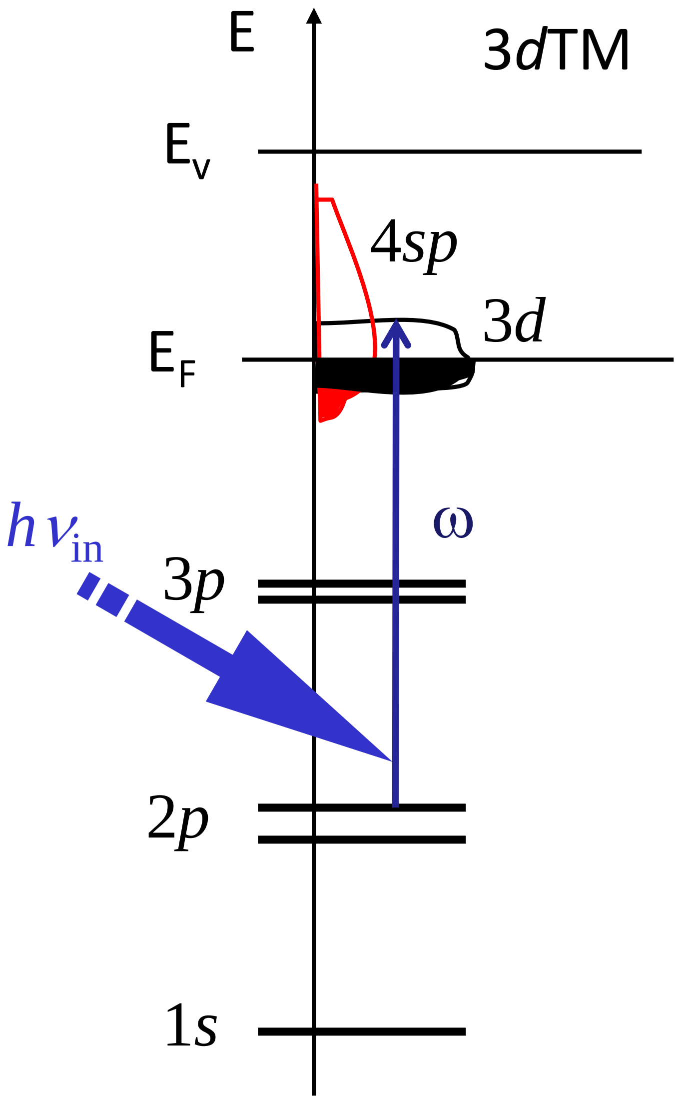
And often are dressed with a strong resonance

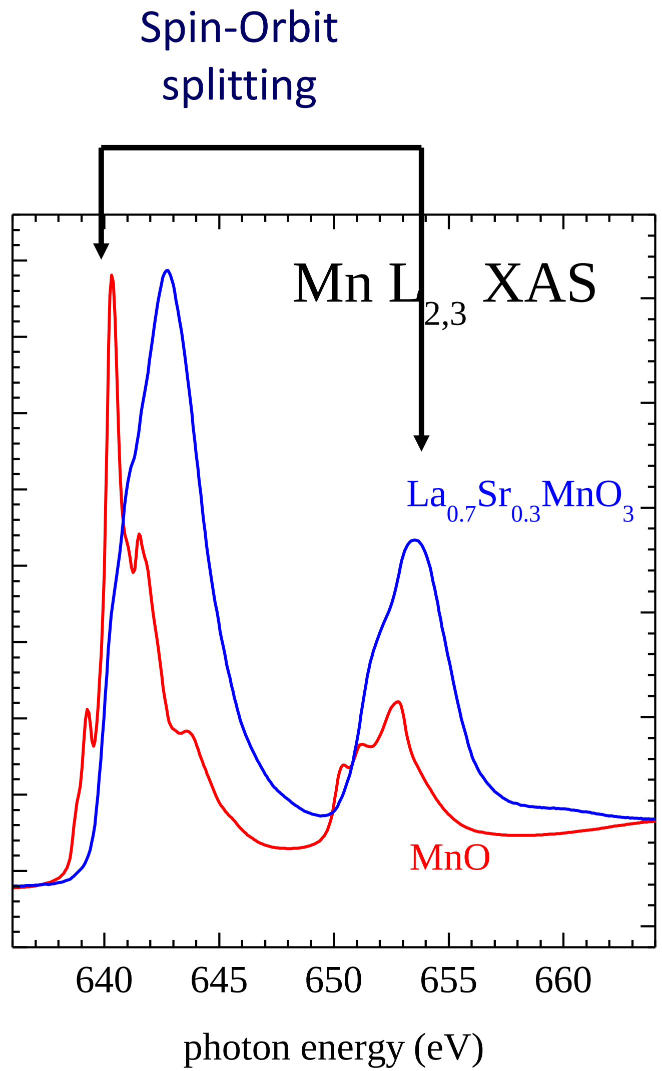
Core level binding energies and edges



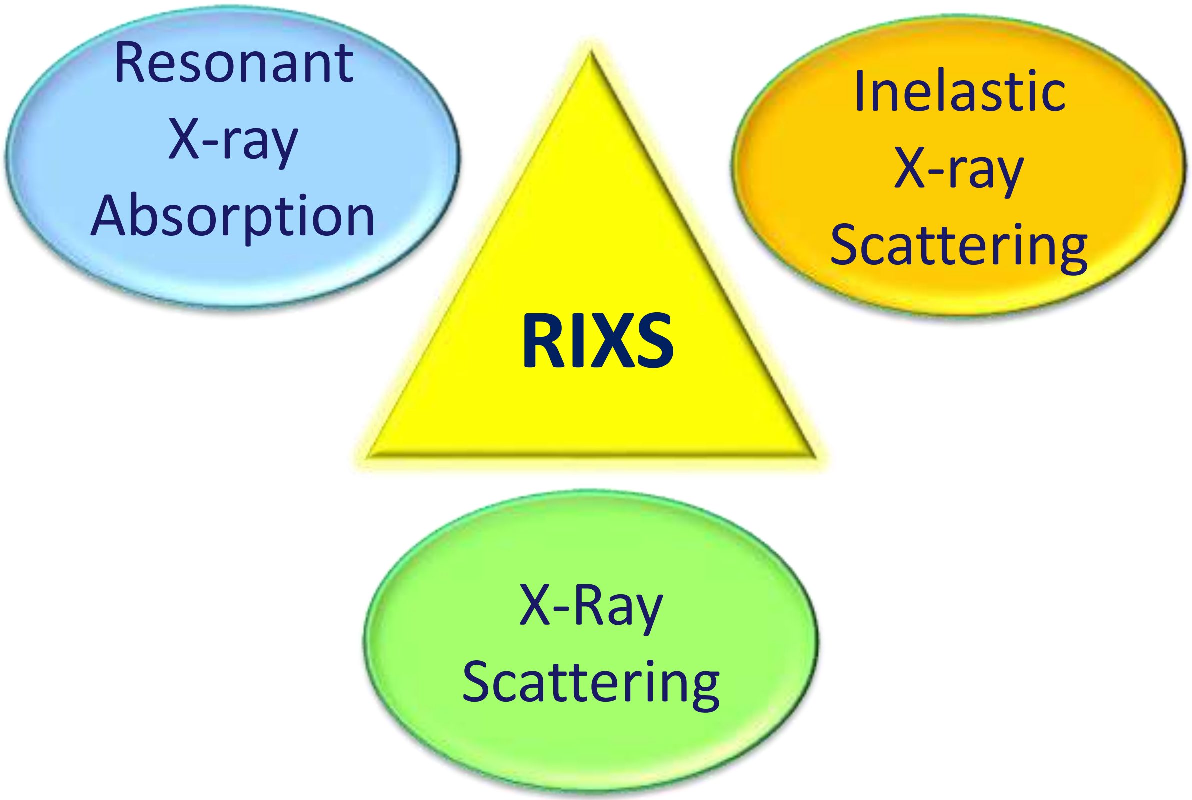


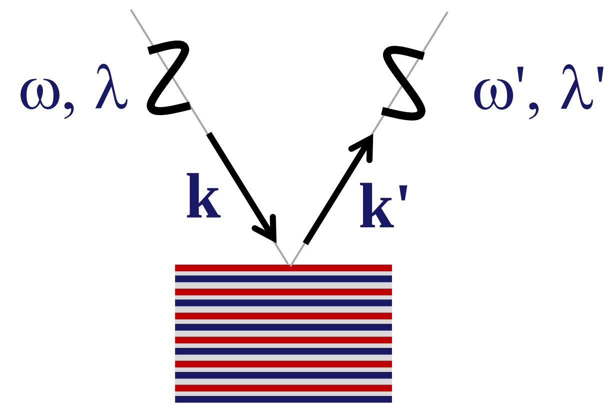
XAS of 3d transition metals





Resonant Inelastic X-ray Scattering



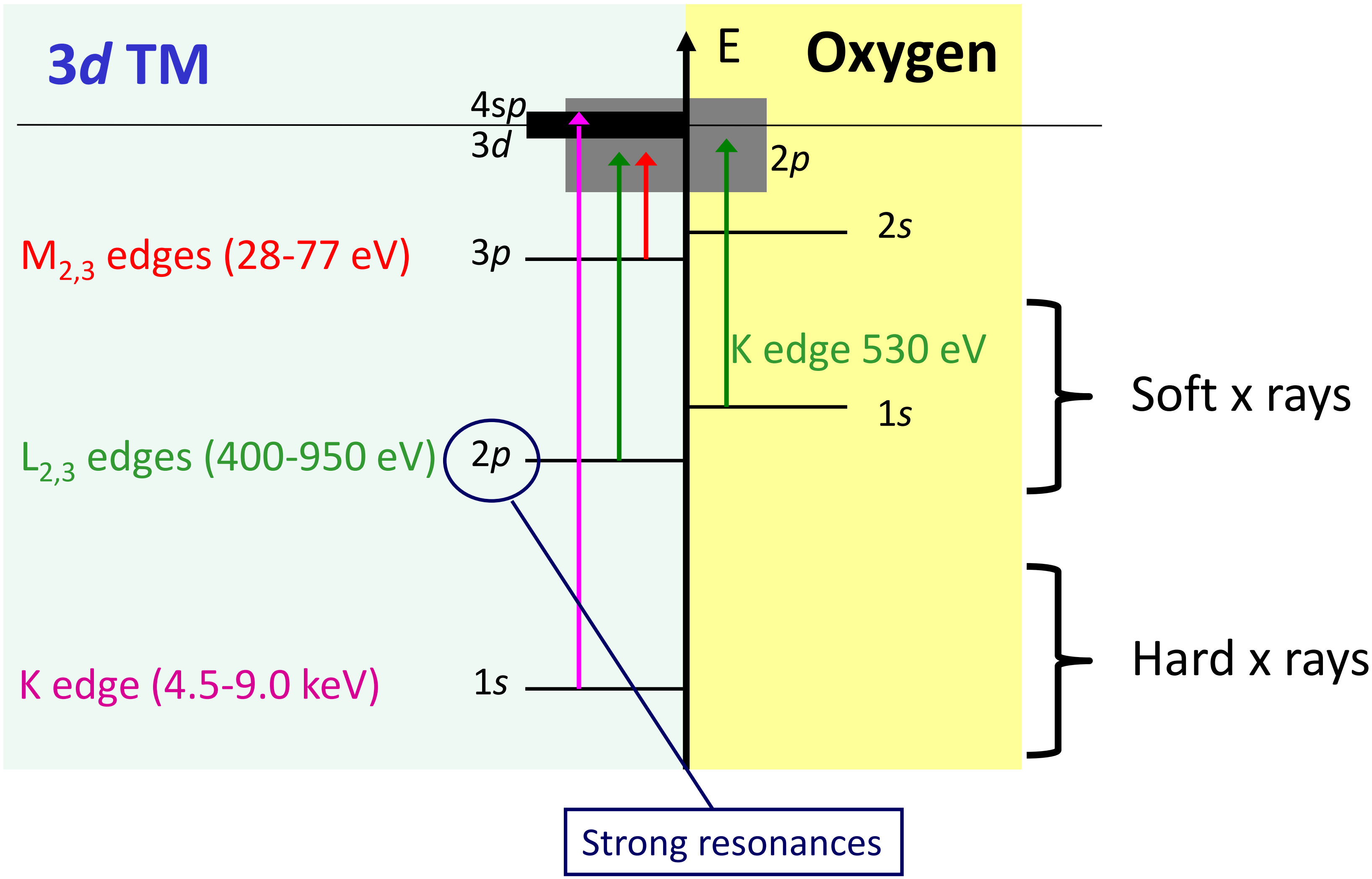


$$\Omega = \omega - \omega'$$

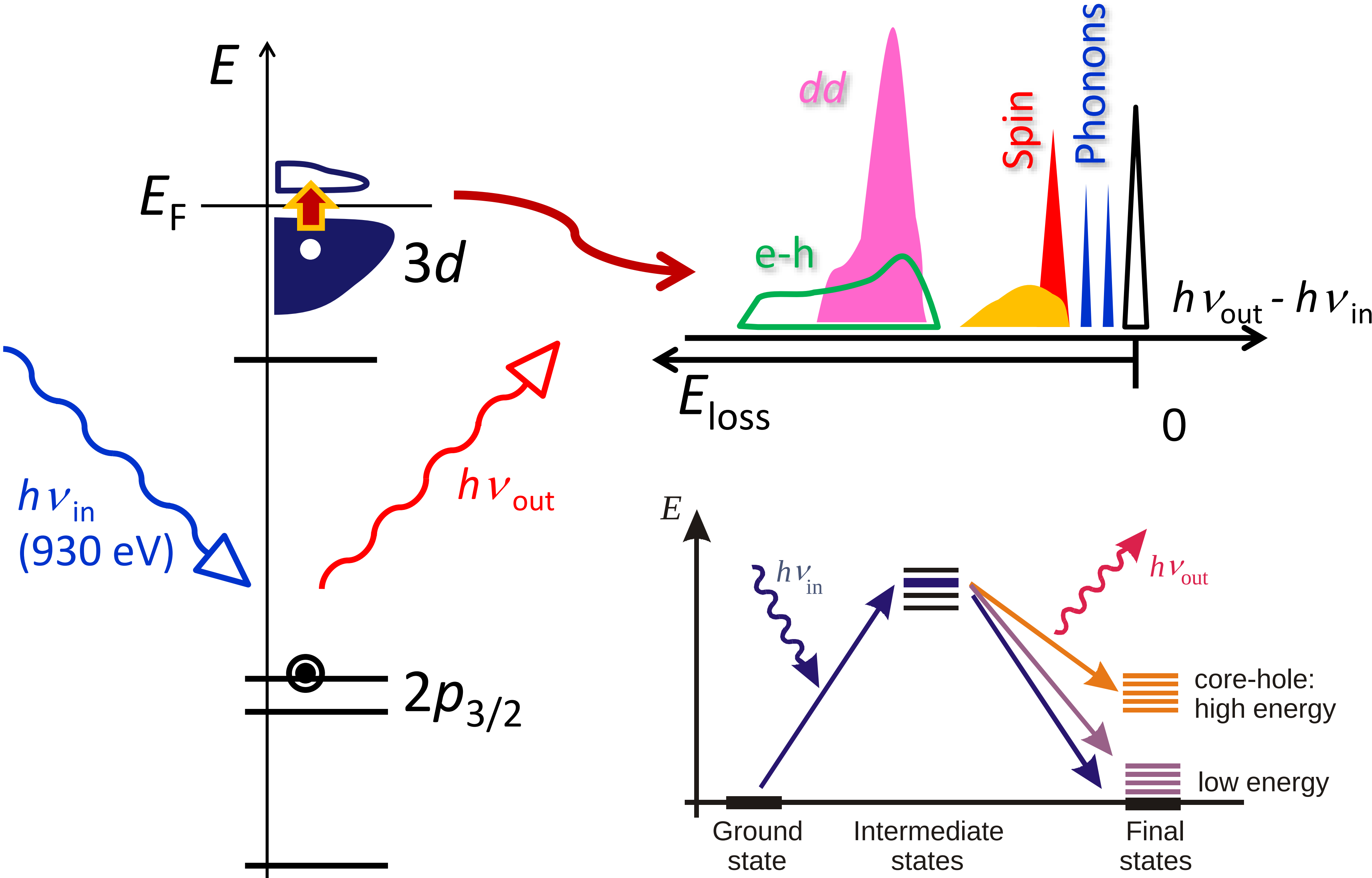
$$\mathbf{q} = \mathbf{k}' - \mathbf{k}$$

The choice of the resonance: 2*p* → 3*d*, L_3 edge

3*d* Transition Metal oxides: a lucky coincidence for soft x-rays

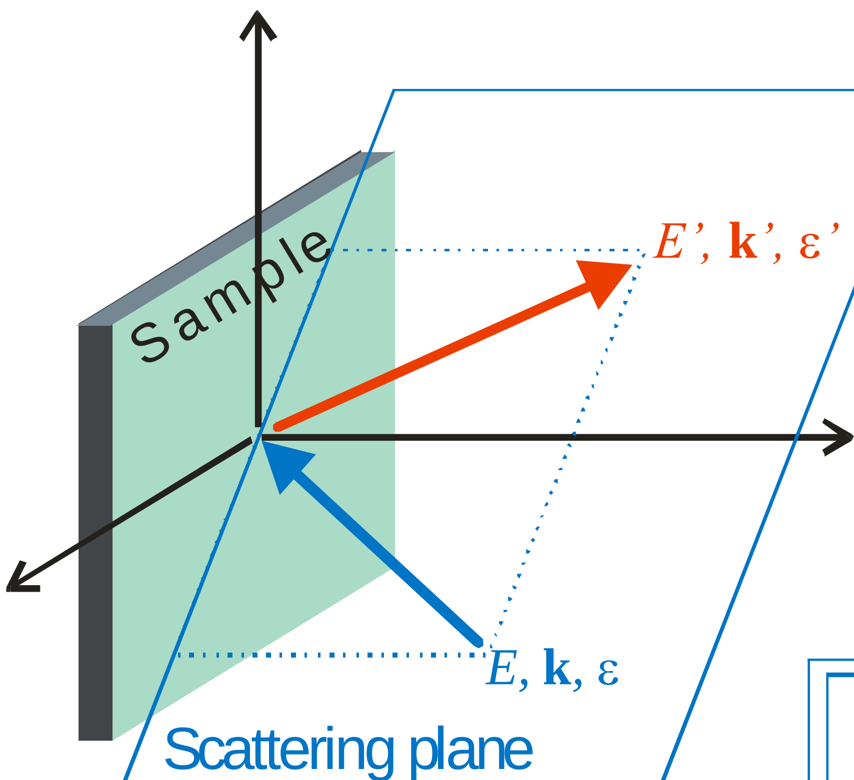


L_3 RIXS



E
E_F
3d
$h\nu_{in}$
(930 eV)
$h\nu_{out}$
$2p_{3/2}$
dd
e-h
Spin
Phonons
$h\nu_{out} - h\nu_{in}$
E_{loss}
0
E
$h\nu_{in}$
$h\nu_{out}$
core-hole:
high energy
low energy
Ground
state
Intermediate
states
Final
states

L edge RIXS : energy and momentum transfer

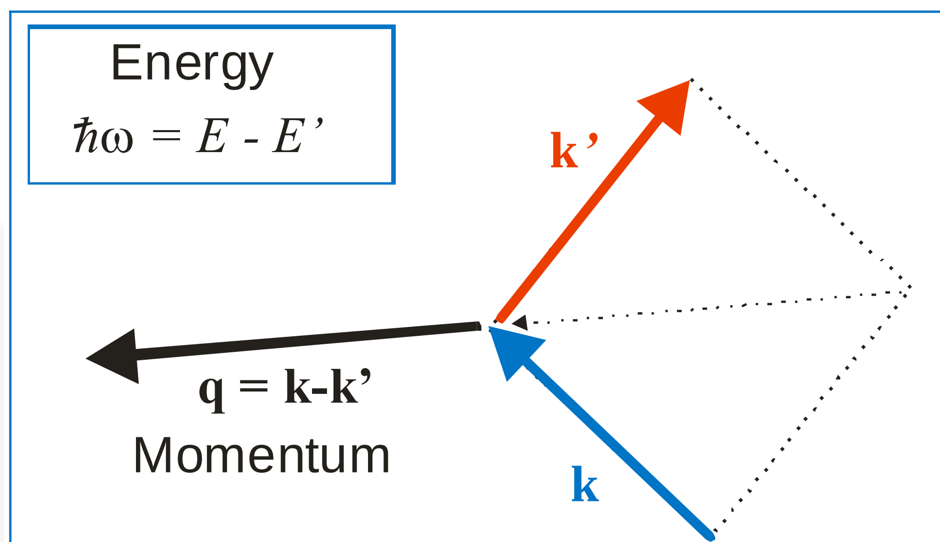


Resonant Inelastic X-ray Scattering:

- an energy loss experiment
- made with photons of high energy
- at a core absorption resonance

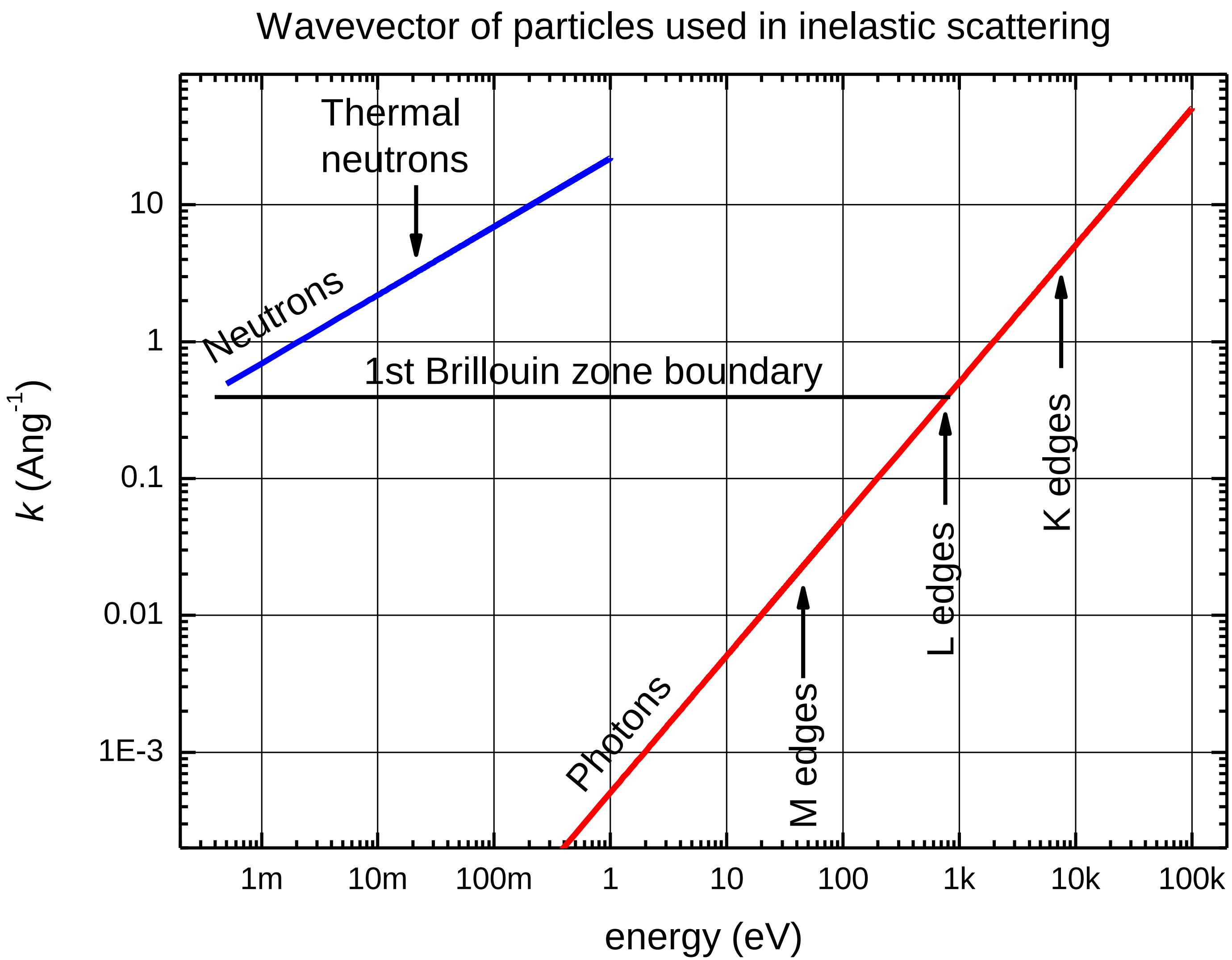
Conservation laws:

- Energy
- Momentum
- "Angular momentum"

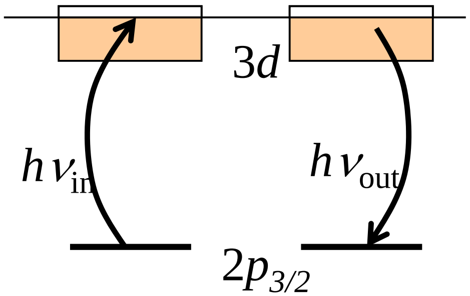


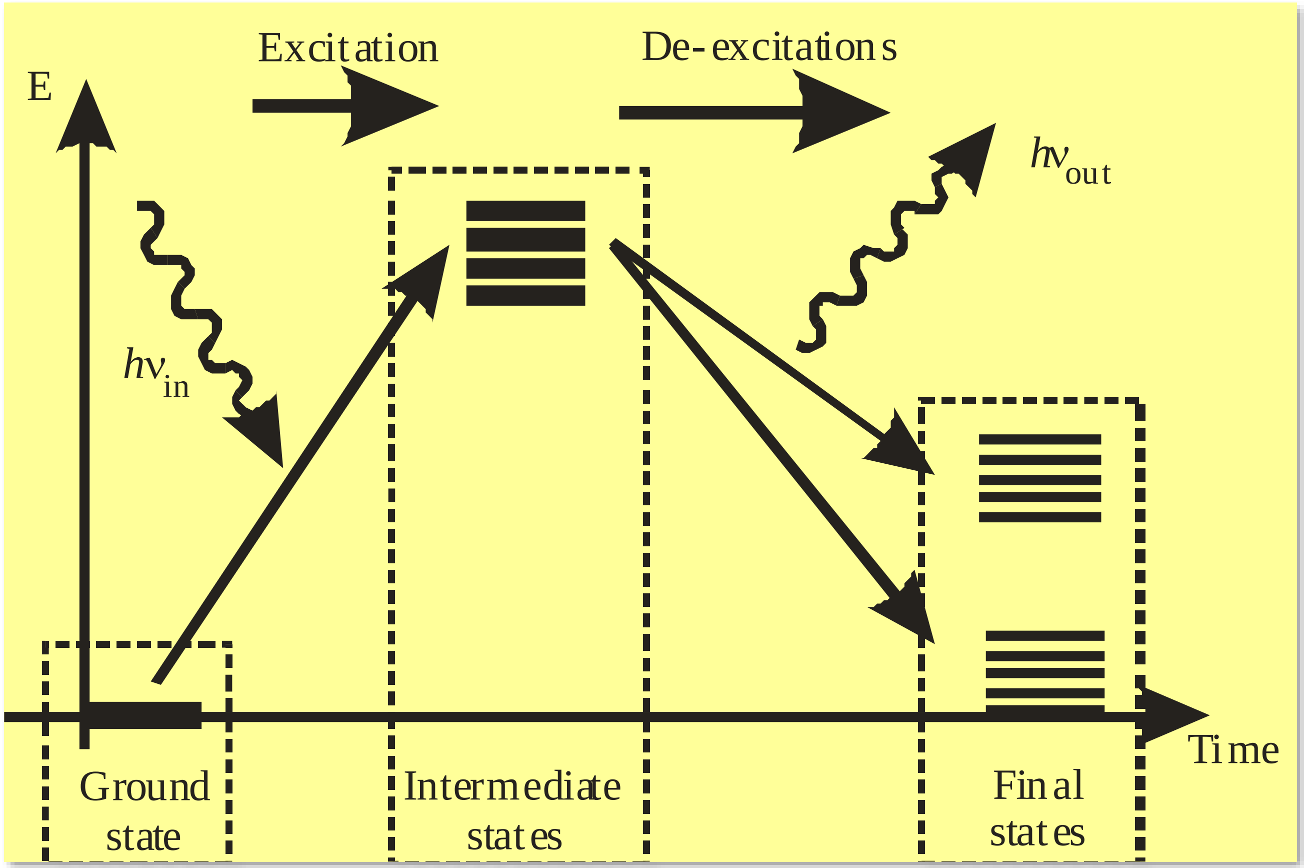
Photon momentum and kinematics

Photons vs Neutrons: energy and momentum



$L_{2,3}$ edge RIXS: intermediate and final states





$$3d^n \longrightarrow 2p^5 3d^{n+1} \longrightarrow 3d^n$$

The potential of soft RIXS (for 3dTM systems)

Site selective,
***q* resolved probe of**
elementary excitations

- charge excitations across the gap
- *dd* excitations
- magnetic excitations
- phonons

$3d^n \longrightarrow 2p^5 3d^{n+1} \longrightarrow$

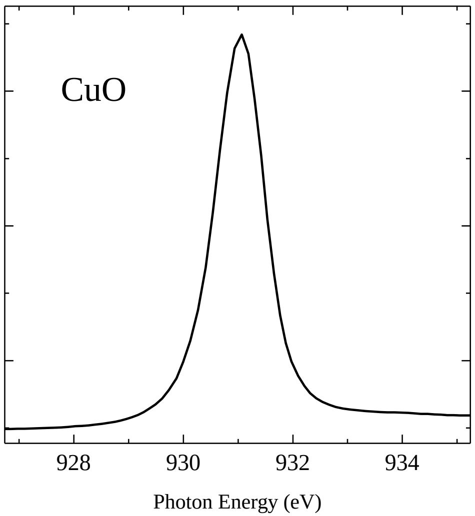
- $3d^n$: elastic, magnetic and phonons
- $3d^{n*}$: *dd* excitations
- $3d^n\underline{L}$: *Charge Transfer excitations*

Cuprates: the "easy" case

In cuprates Cu is divalent: $Cu^{2+} \Longleftrightarrow 3d^9$

This makes XAS almost trivial: 1 peak only

$$3d^9 \longrightarrow (2p_{3/2})^3 3d^{10}$$

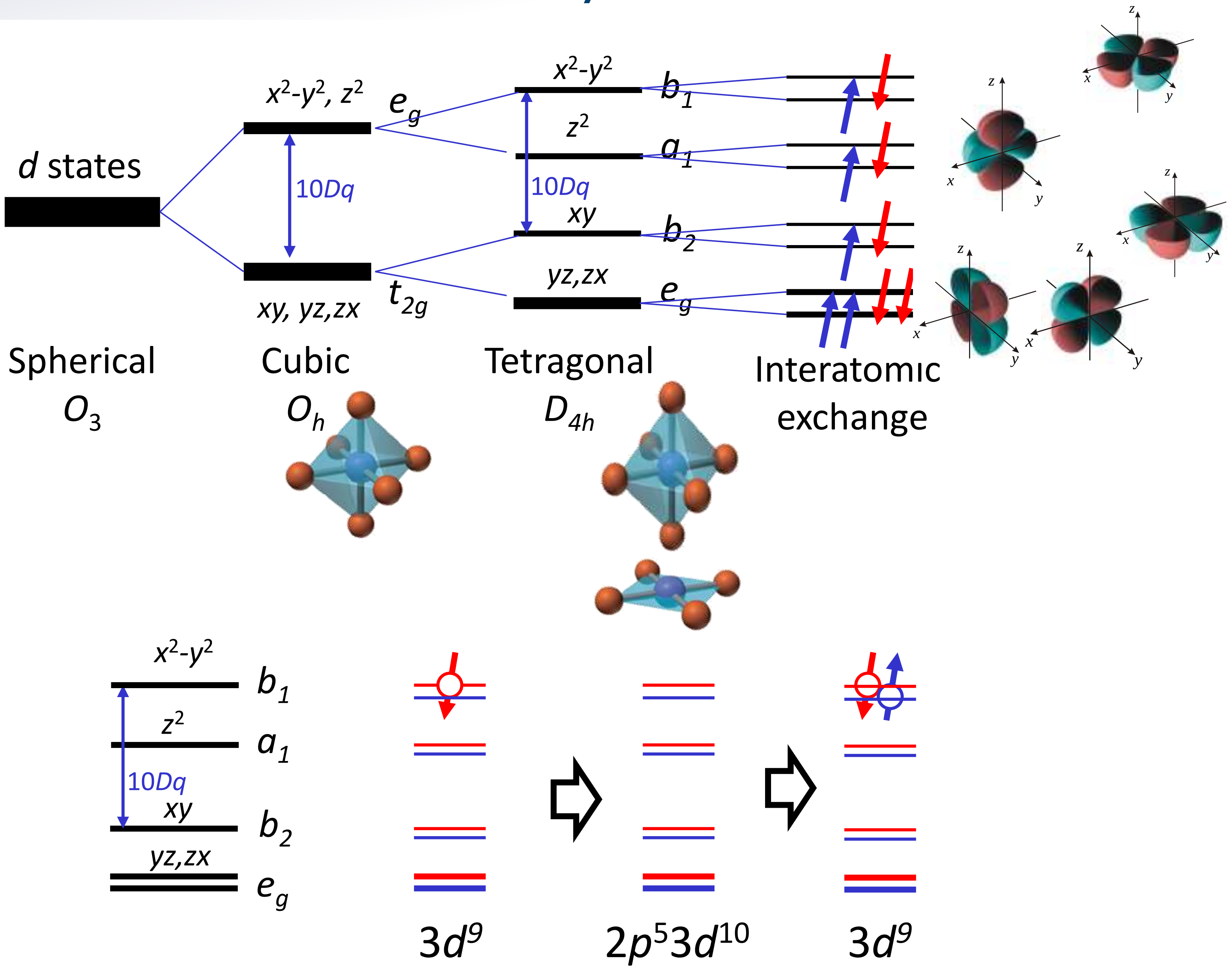


RIXS can be calculated even by hand:

$$3d^9 \longrightarrow (2p_{3/2})^3 3d^{10} \longrightarrow (3d^9)^*$$

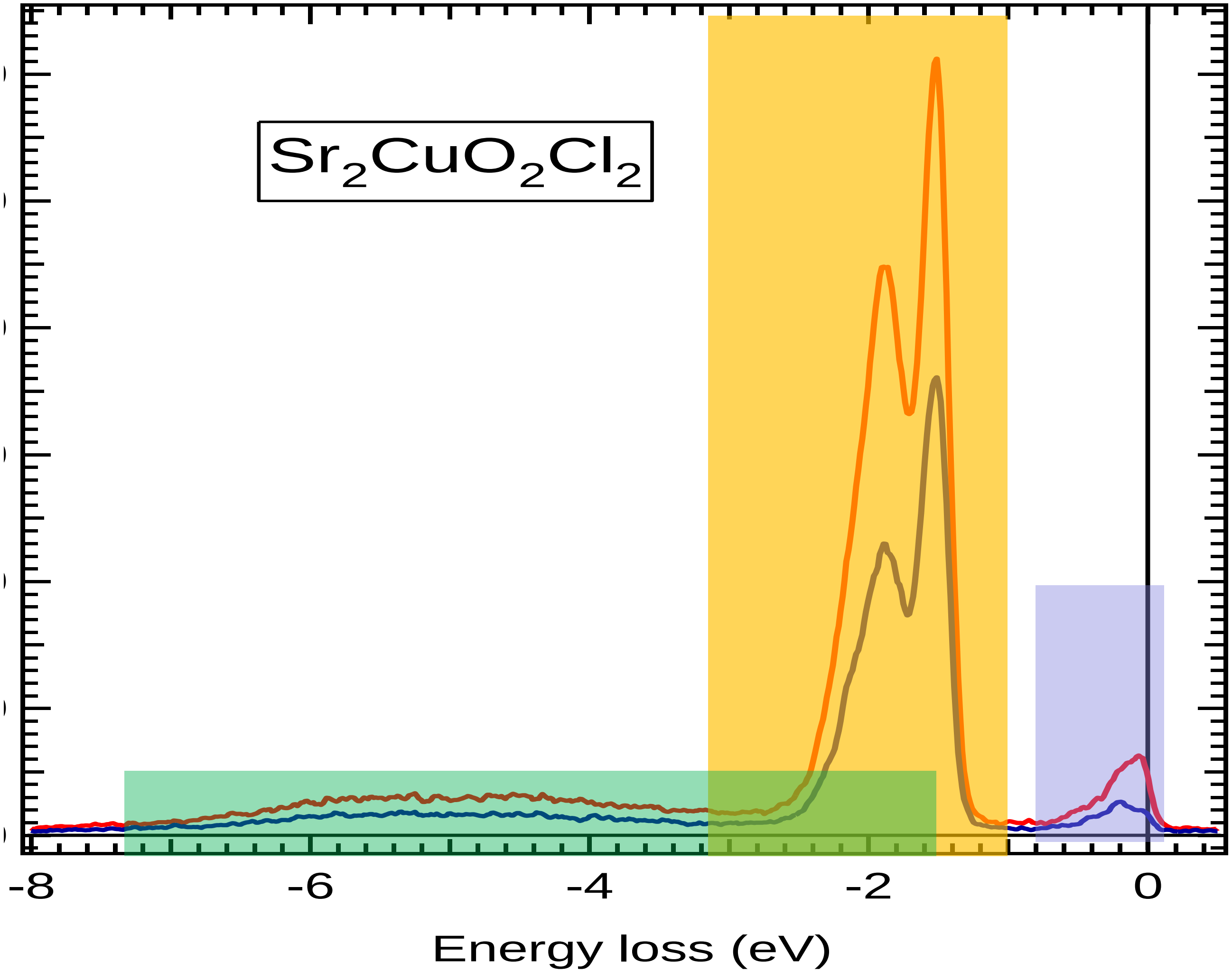
Even for magnetic excitations (spin waves), because fast collision approximation is a very good approximation

dd excitations in Cu^{2+} systems



Cu L_3 RIXS of cuprates: mainly *dd* excitations

$3d^9 \rightarrow 2p^5 3d^{10} \rightarrow$
- $3d^9$: elastic, magnetic and phonons
- $3d^{9*}$: *dd* excitations
- $3d^{10}\underline{L}$: *Charge Transfer excitations*

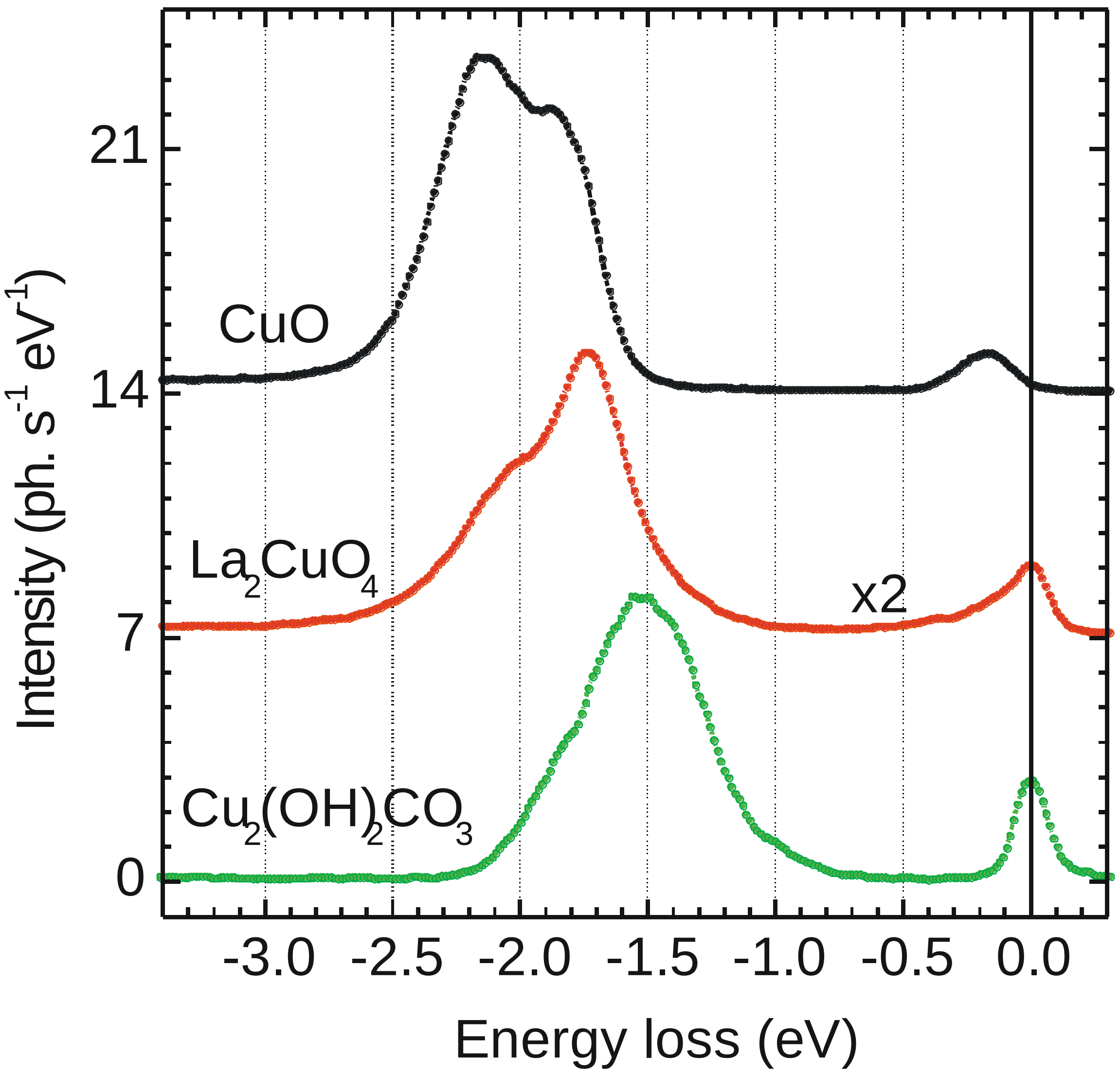


All final states are reached via 2 electric dipole allowed transitions!

Photons get coupled to electrons spin thanks to 2*p* spin-orbit interaction

At L_3 edge elastic peak is very small (not the case at K)

Cu L_3 edge: CuO, La_2CuO_4, Malachite



Cu^{2+} in square approximately planar coordination

Cu-O distances:
CuO 1.7 – 2-2 Ang
LCO 1.9 – 2.4 Ang
Malachite 1.9 – 2.6 Ang

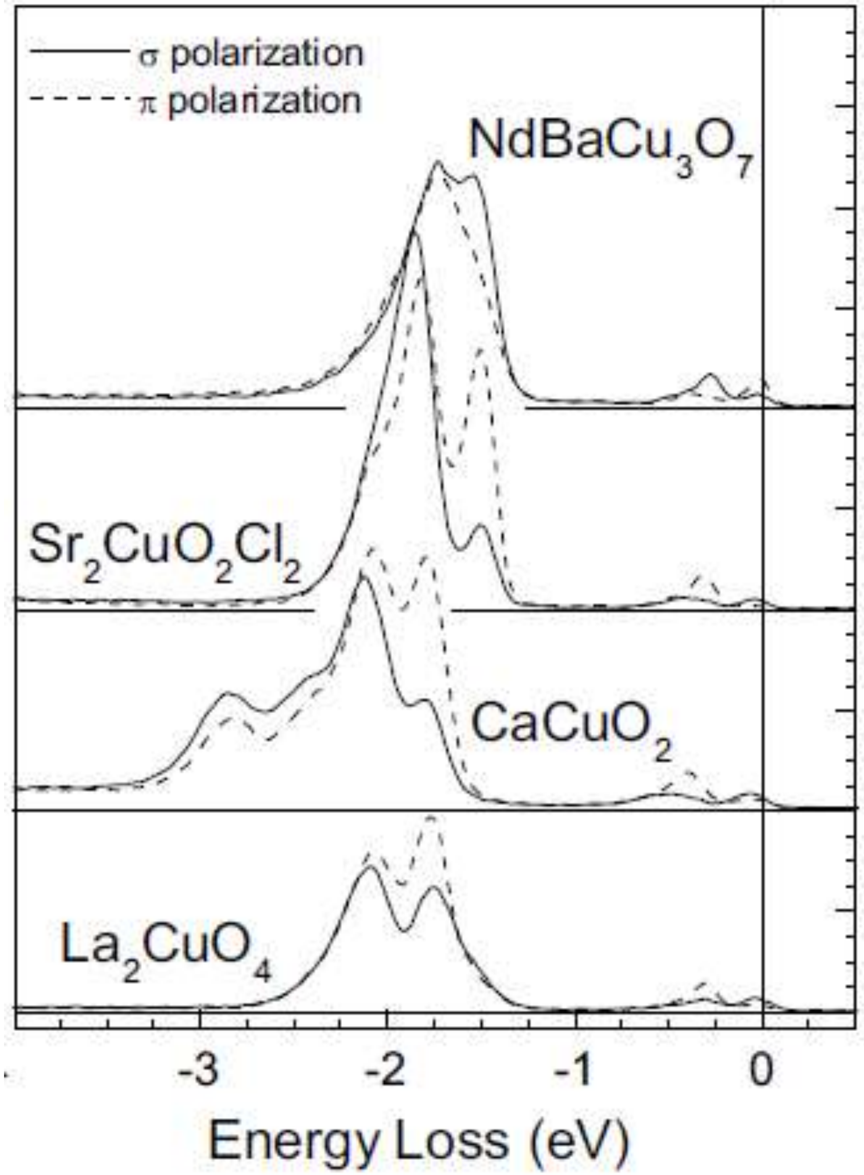
Different Cu^{2+} coordination, symmetry, hybridization

⇓

Different *dd* excitations

G. Ghiringhelli, A. Piazzalunga, X. Wang, A. Bendounan, H. Berger, F. Bottegoni, N. Christensen, C. Dallera, M. Grioni, J.-C. Grivel, M. Moretti Sala, L. Patthey, J. Schlappa, T. Schmitt, V. Strocov , and L. Braicovich, Eur.Phys. J. Special topics **169**, 199 (2009)

This is a very direct way of measuring the *dd*-excitation energies

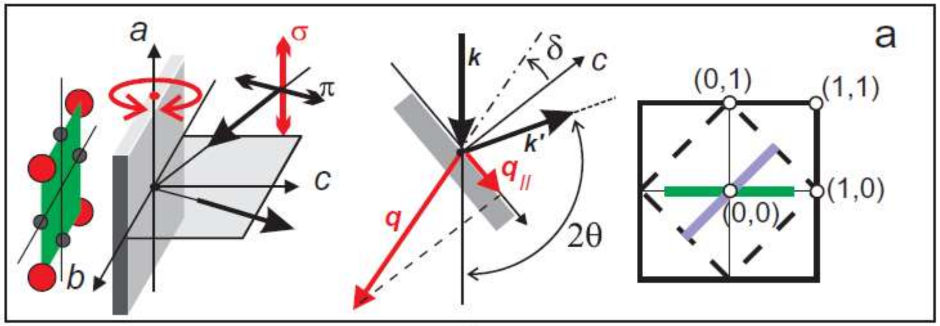


New Journal of Physics

The open-access journal for physics

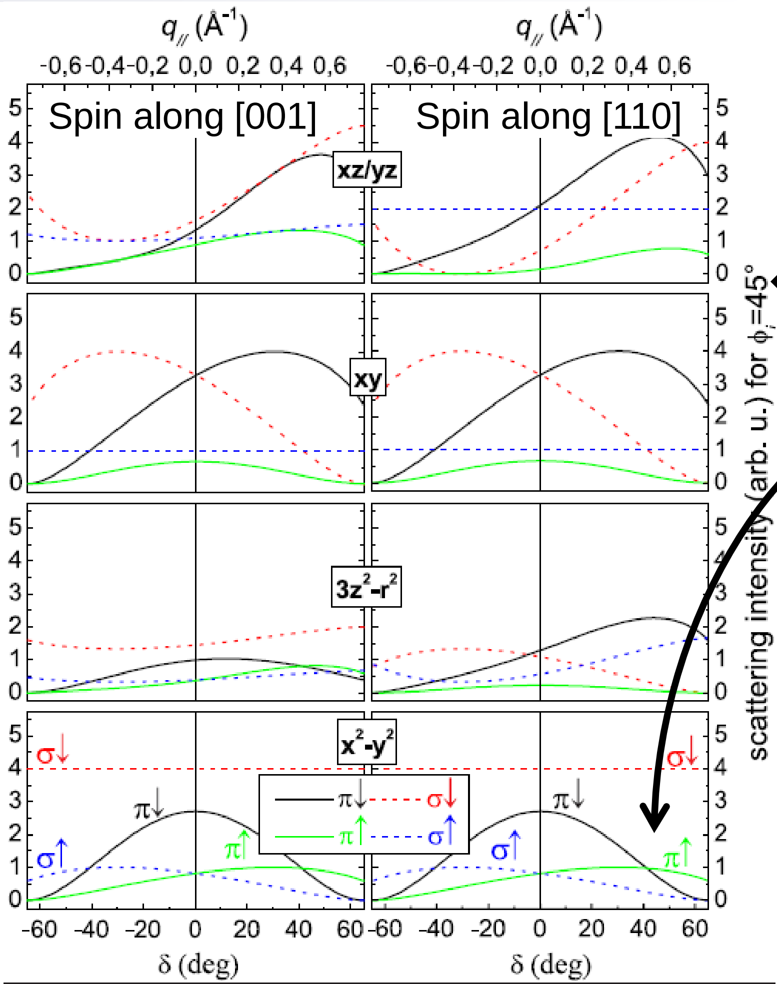
Energy and symmetry of dd excitations in undoped layered cuprates measured by Cu L_3 resonant inelastic x-ray scattering

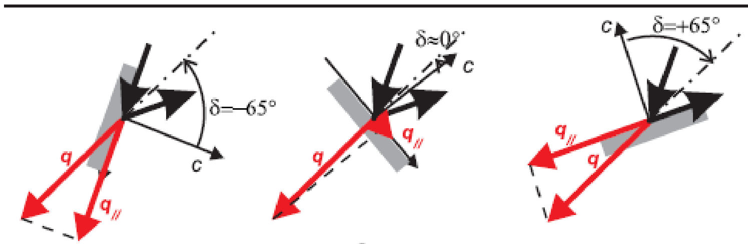
M Moretti Sala[1,8,9], V Bisogni[2,10], C Aruta[3], G Balestrino[4], H Berger[5], N B Brookes[2], G M de Luca[3], D Di Castro[4], M Grioni[5], M Guarise[5], P G Medaglia[4], F Miletto Granozio[3], M Minola[1], P Perna[3], M Radovic[3,11], M Salluzzo[3], T Schmitt[6], K J Zhou[6], L Braicovich[7] and G Ghiringhelli[7]



M. Moretti Sala, et al New J. Phys. **13**, 043026 (2011)

dd-excitation energies from fitting using atomic cross sections





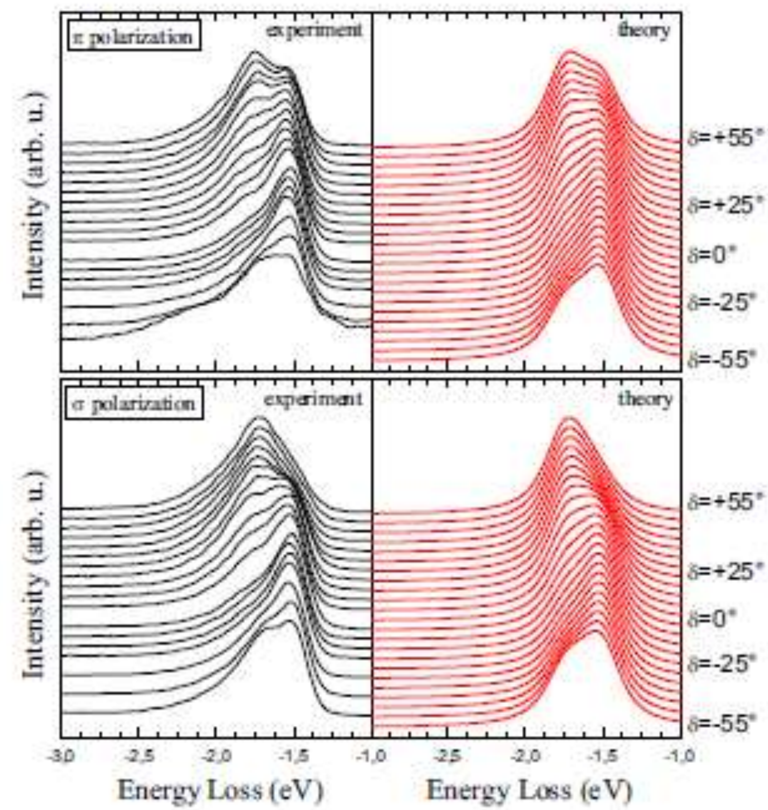
2θ=130°

Ground state: $\underline{3d}^{\downarrow}{}_{x^2-y^2}$

Spin flip: $\underline{3d}^{\uparrow}{}_{x^2-y^2}$

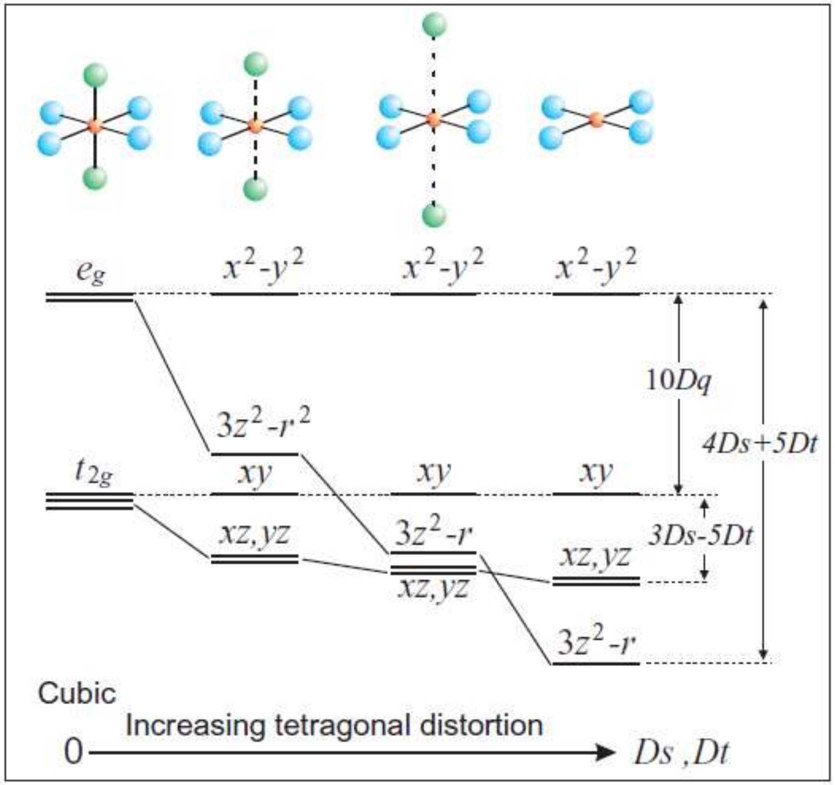
$F(\theta_{in},\phi_{in},\theta_{out},\phi_{out},\theta_{spin},\phi_{spin},\varepsilon_{in},\varepsilon_{out})$

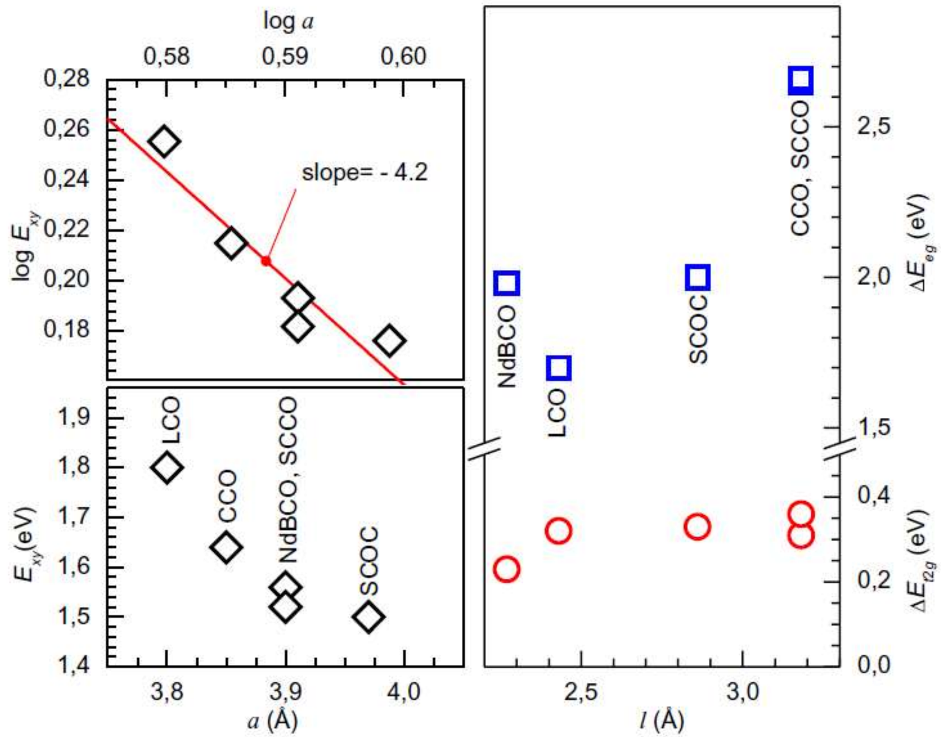
NdBCO



M. Moretti Sala, et al New J. Phys. **13**, 043026 (2011)

Crystal field trends in cuprates





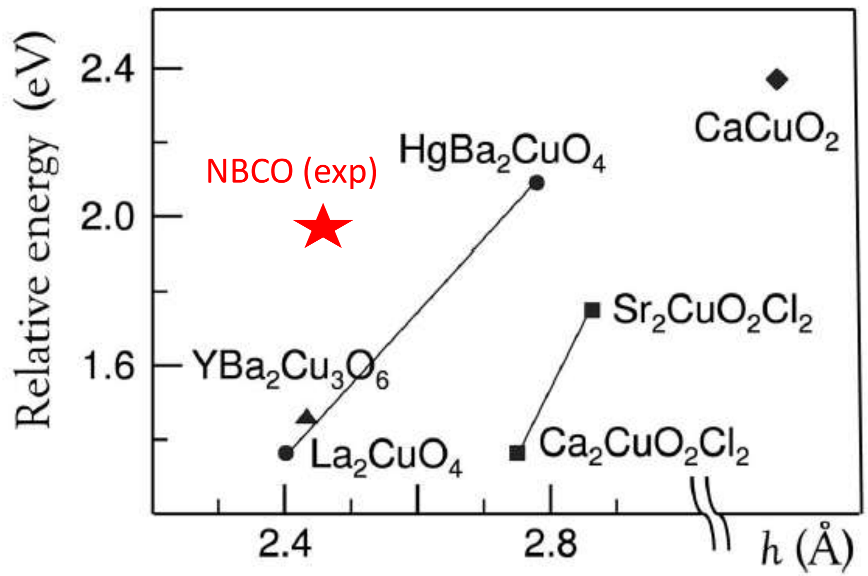
	La_2CuO_4	$Sr_2CuO_2Cl_2$	$CaCuO_2$
J [meV]	130[34,35]	130[35]	130[35]
$E_{3z^2-r^2}$ $(\Gamma_{3z^2-r^2})$ [eV]	1.70 (.14)	1.97 (.10)	2.72 (.12)
E_{xy} (Γ_{xy}) [eV]	1.80 (.10)	1.50 (.08)	1.75 (.09)
$E_{xz/yz}$ $(\Gamma_{xz/yz})$ [eV]	2.12 (.14)	1.84 (.10)	2.10 (.18)

M. Moretti Sala, et al New J. Phys. **13**, 043026 (2011)

CASSCF: complete-active-space self-consistent-field method

Table 1 | CASSCF+SDCI versus RIXS results for the Cu *d*-level splittings in La_2CuO_4, $Sr_2CuO_2Cl_2$, and $CaCuO_2$ (eV). The ground-state Cu $t_{2g}^6 d_{z^2}^2 d_{x^2-y^2}^1$ configuration is taken as reference. A 2*J* term was here substracted from each of the RIXS values reported in Ref. [14], see text.

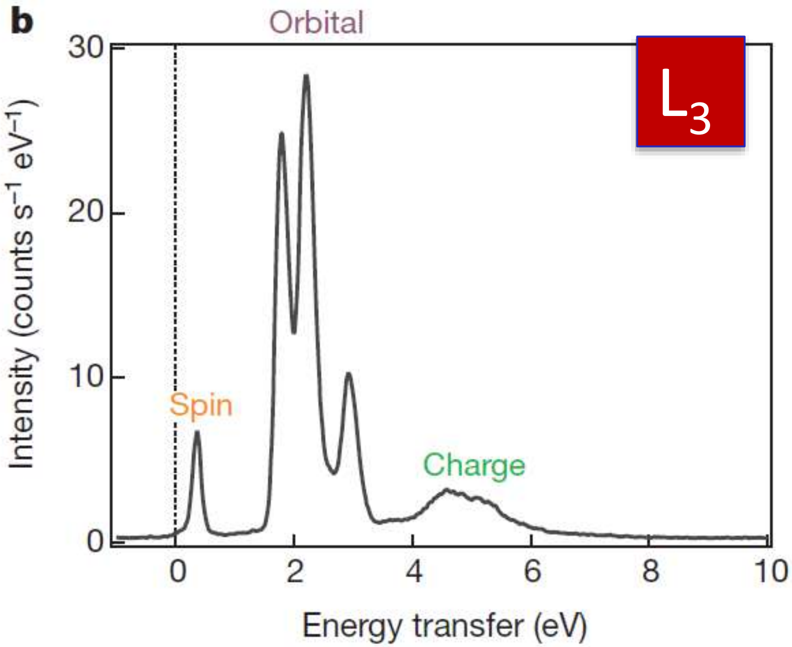
Hole orbital	La_2CuO_4 SDCI/RIXS	$Sr_2CuO_2Cl_2$ SDCI/RIXS	$CaCuO_2$ SDCI/RIXS
x^2-y^2	0	0	0
z^2	1.37/1.44	1.75/1.71	2.38/2.39
xy	1.43/1.54	1.16/1.24	1.36/1.38
xz,yz	1.78/1.86	1.69/1.58	2.02/1.69

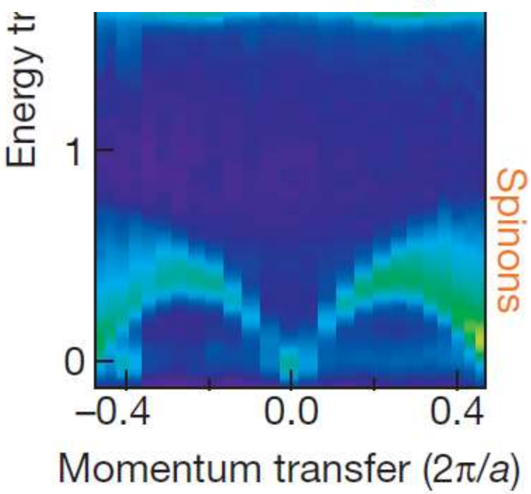


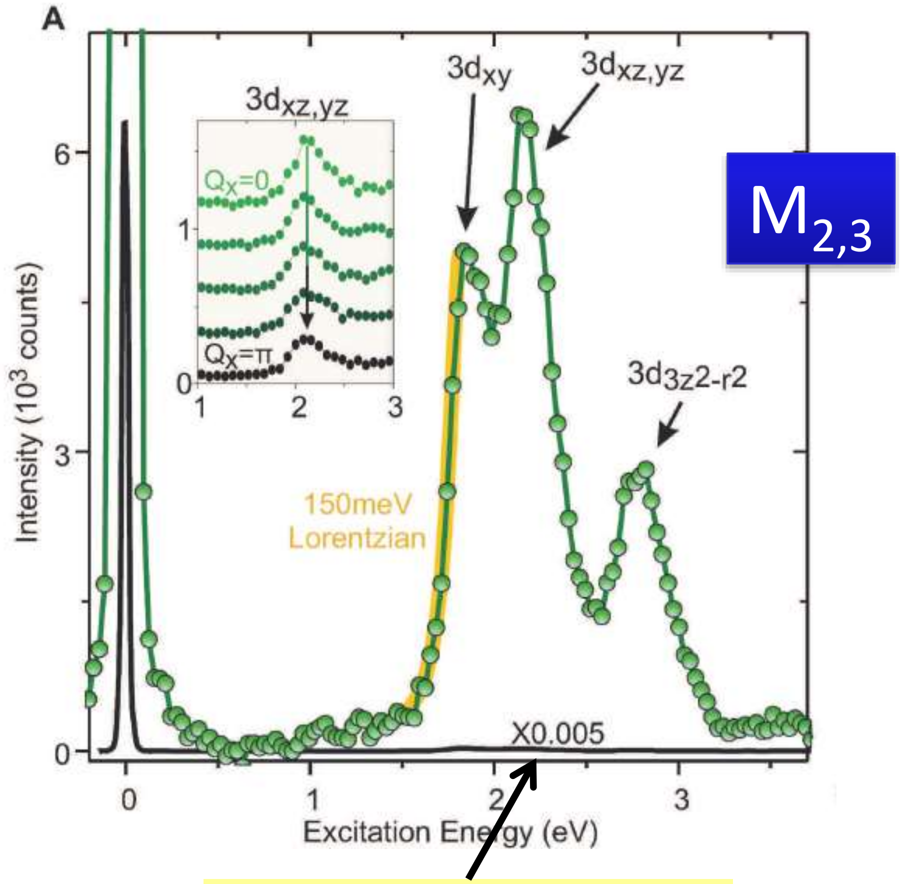
Liviu Hozoi, Liudmila Siurakshina, Peter Fulde & Jeroen van den Brink,
SCIENTIFIC REPORTS 1 : 65 (2011)

[14] M. Moretti Sala, et al New J. Phys. **13**, 043026 (2011)

dd excitations: Cu L_3 vs $M_{2,3}$ edges







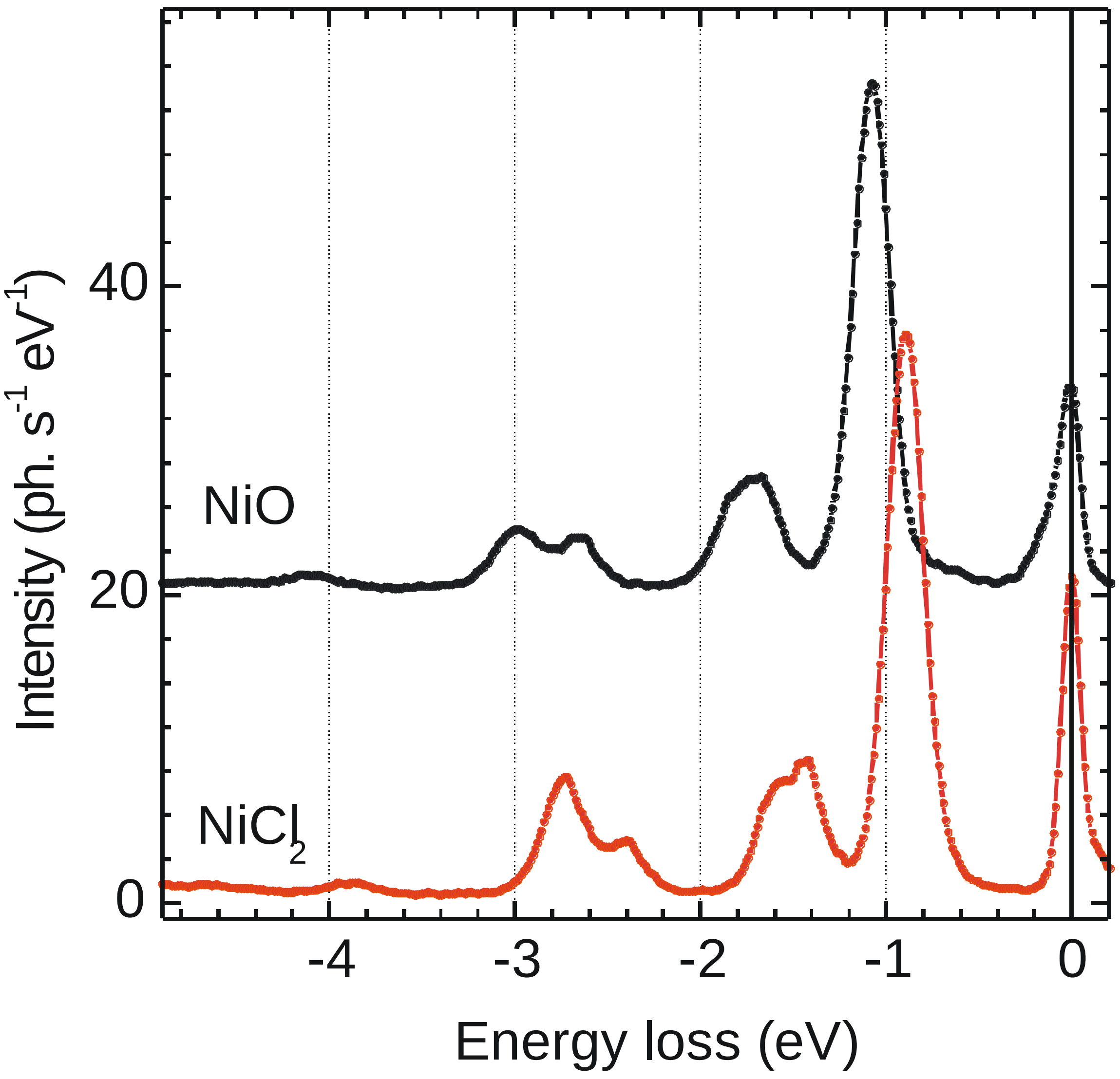
No spin excitations at Gamma point

Very weak signal with respect to the elastic peak

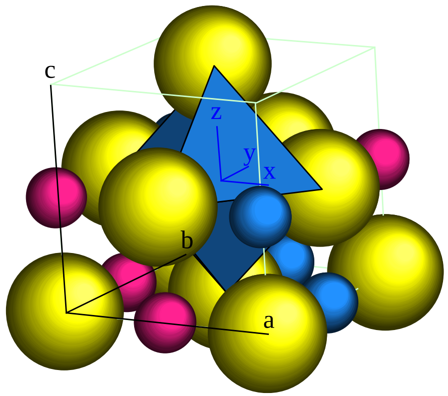
J. Schlappa et al NATURE 485, 82 (2012)

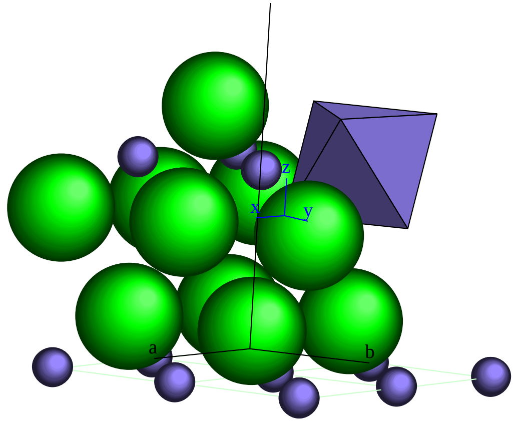
L.A. Wray et al arXiv:1203.2397v1

Ni L_3 edge: NiO, $NiCl_2$

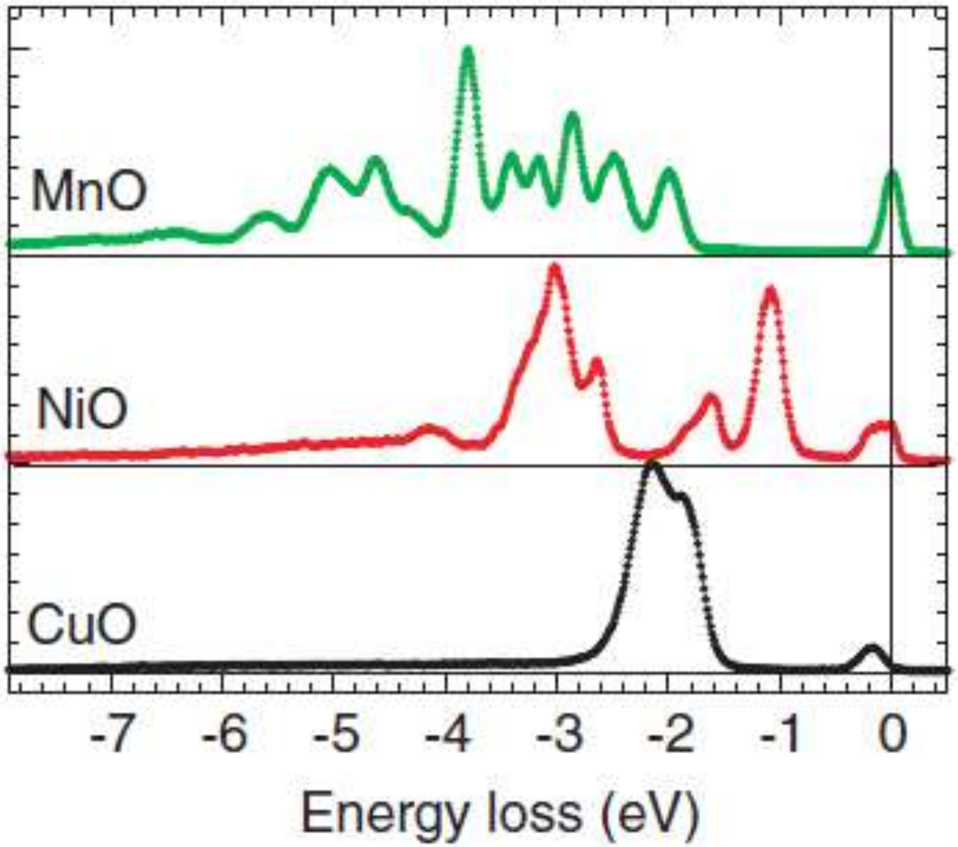


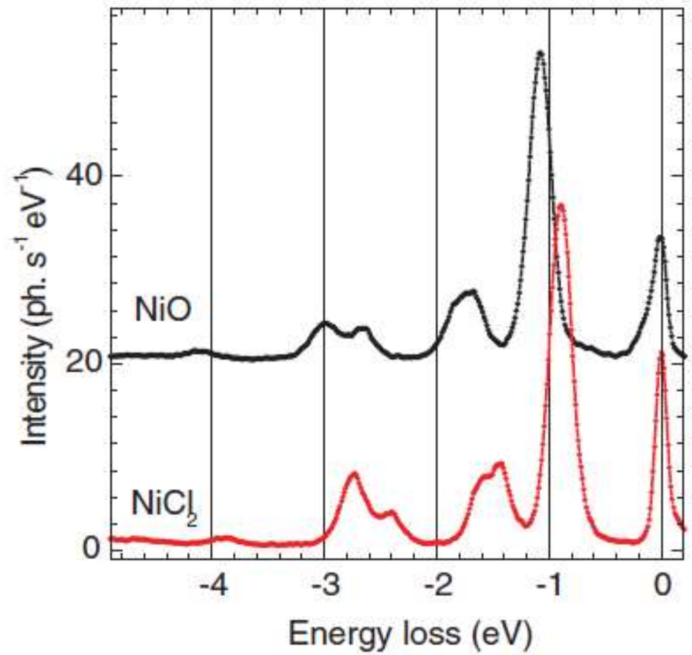
Ni^{2+} ($3d^8$) in octahedral coordination

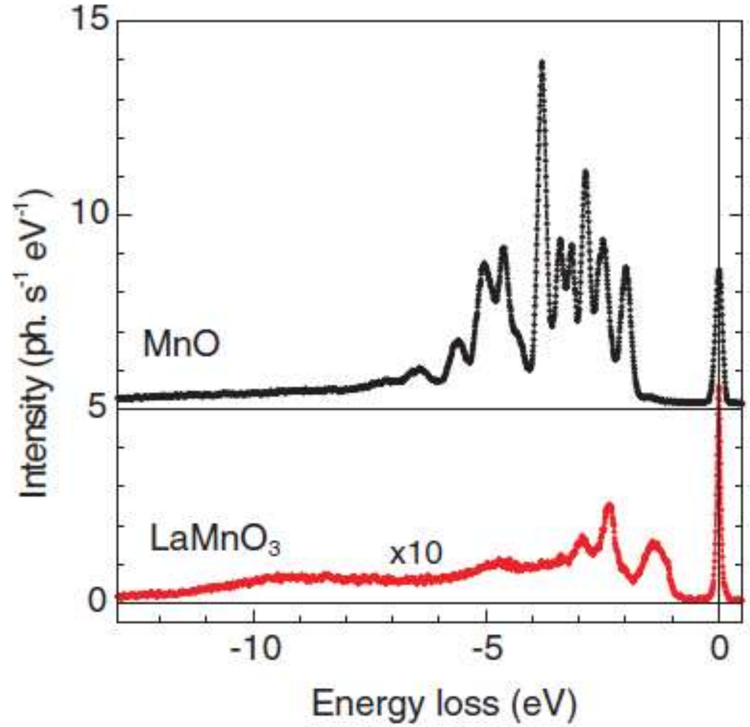




dd and CT excitations in simple oxides

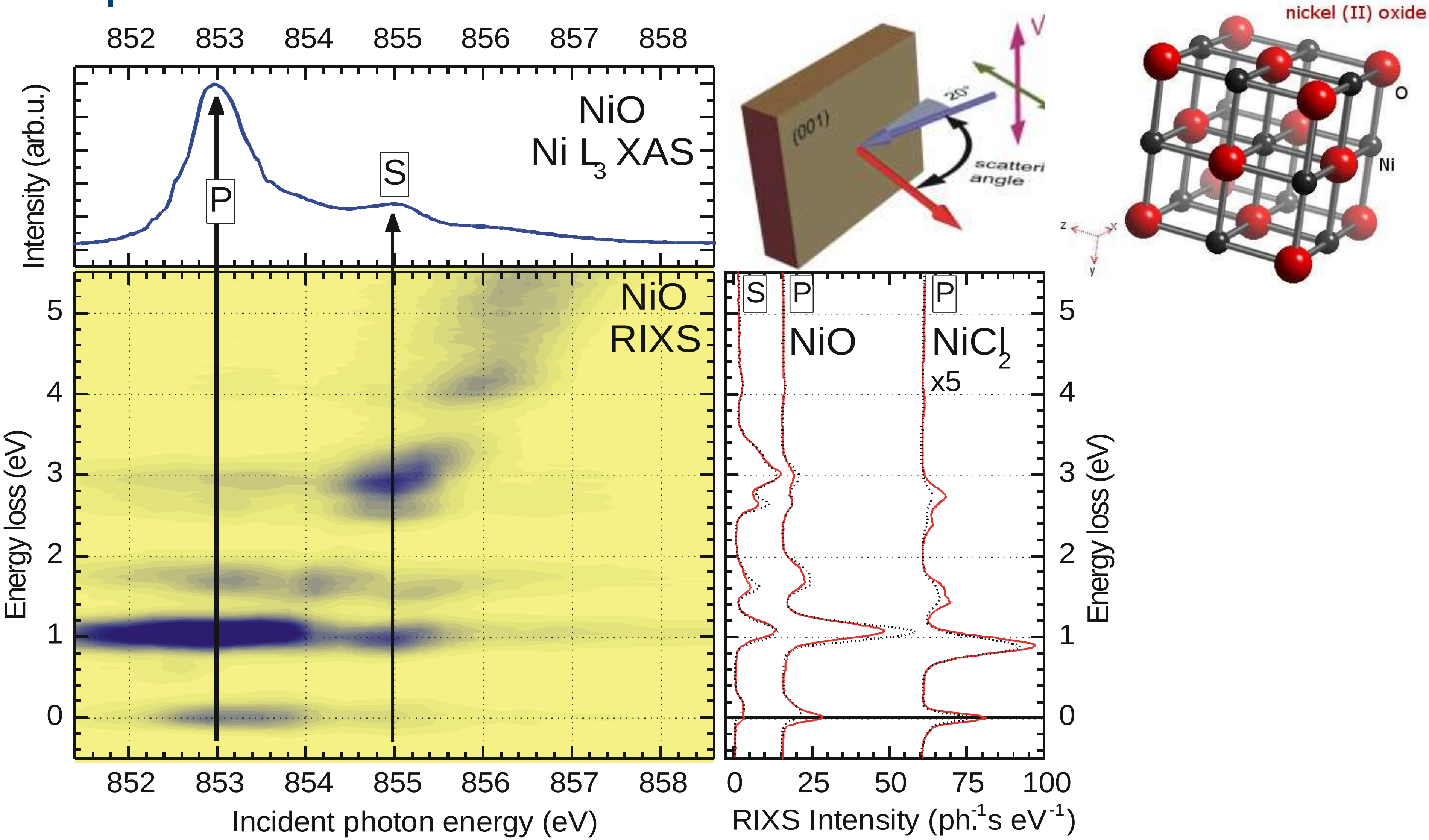






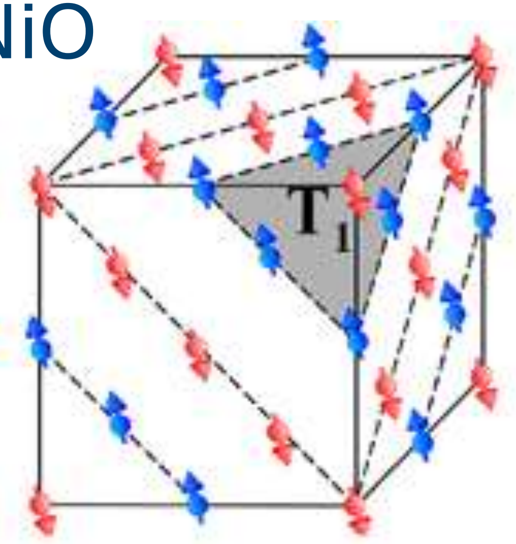
G. Ghiringhelli, A. Piazzalunga, X. Wang, A. Bendounan, H. Berger, F. Bottegoni, N. Christensen, C. Dallera, M. Grioni, J.-C. Grivel, M. Moretti Sala, L. Patthey, J. Schlappa, T. Schmitt, V. Strocov , and L. Braicovich, Eur.Phys. J. Special topics **169**, 199 (2009)

RIXS of NiO: incident photon energy depencence …

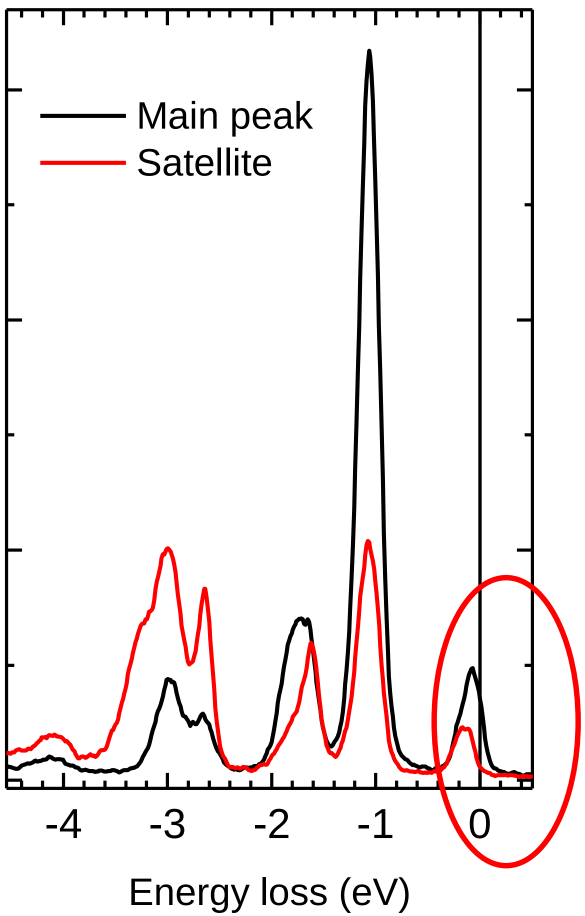


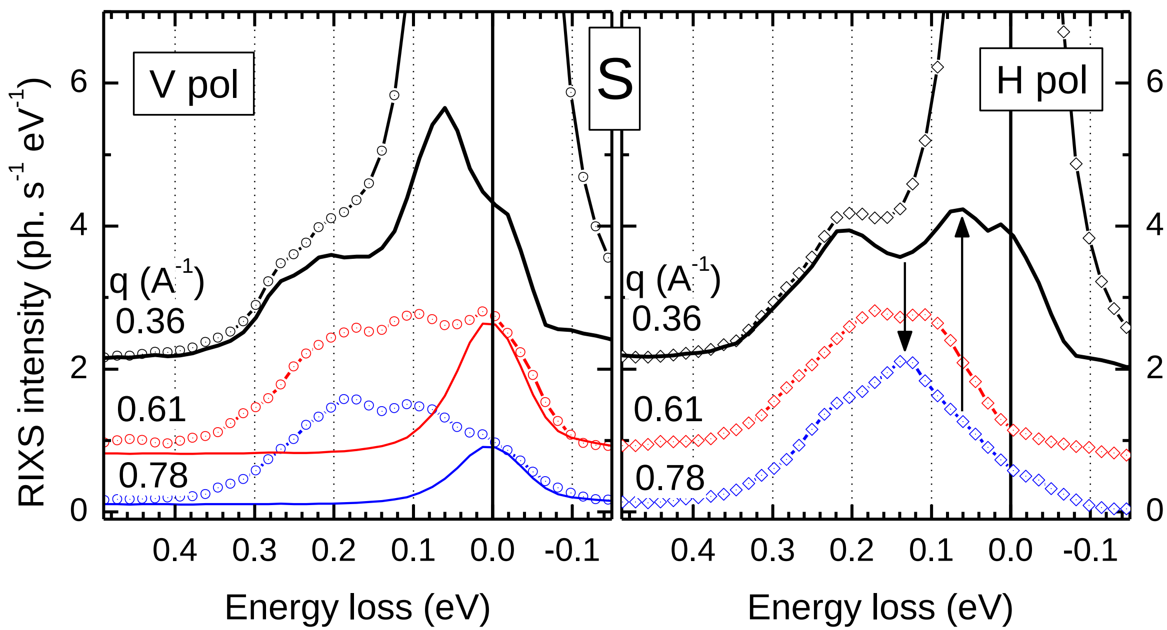
G. Ghiringhelli A. Piazzalunga, C. Dallera, L. Braicovich, T. Schmitt, V.N. Strocov, J. Schlappa, L. Patthey, X. Wang, H. Berger, and M. Grioni, PRL **102**, 027401 (2009)

... and magnetic excitations in NiO



Interatomic exchange splitting : ~115 meV



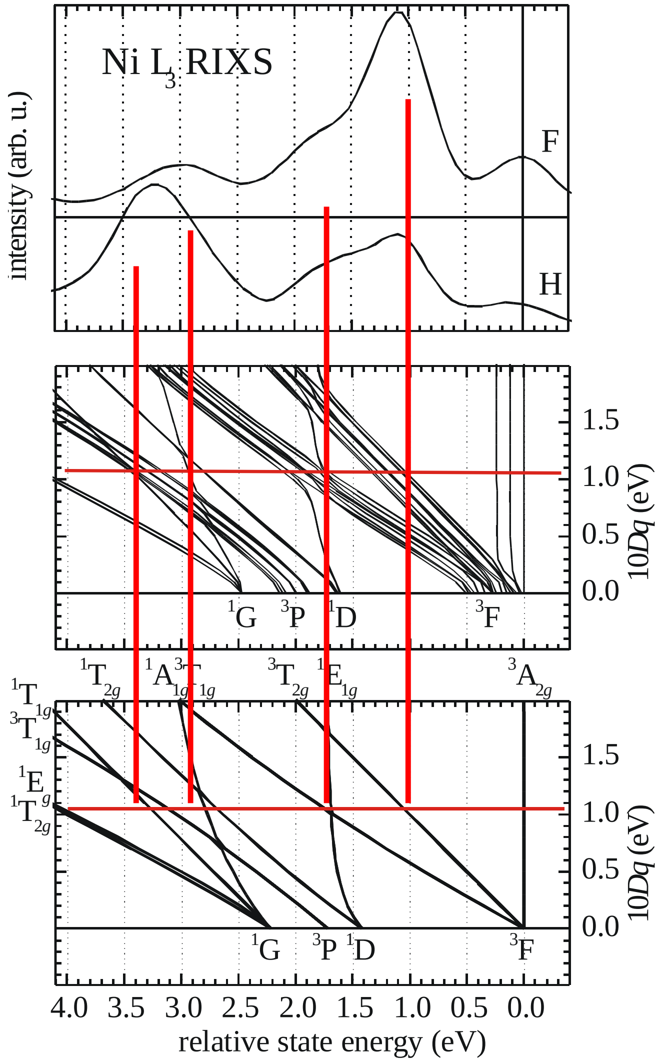


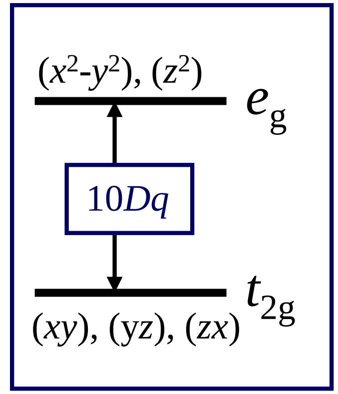
No evident dispersion of these magnetic excitations

G. Ghiringhelli A. Piazzalunga, C. Dallera, L. Braicovich, T. Schmitt, V.N. Strocov, J. Schlappa, L. Patthey, X. Wang, H. Berger, and M. Grioni, PRL **102**, 027401 (2009)

Many excited states

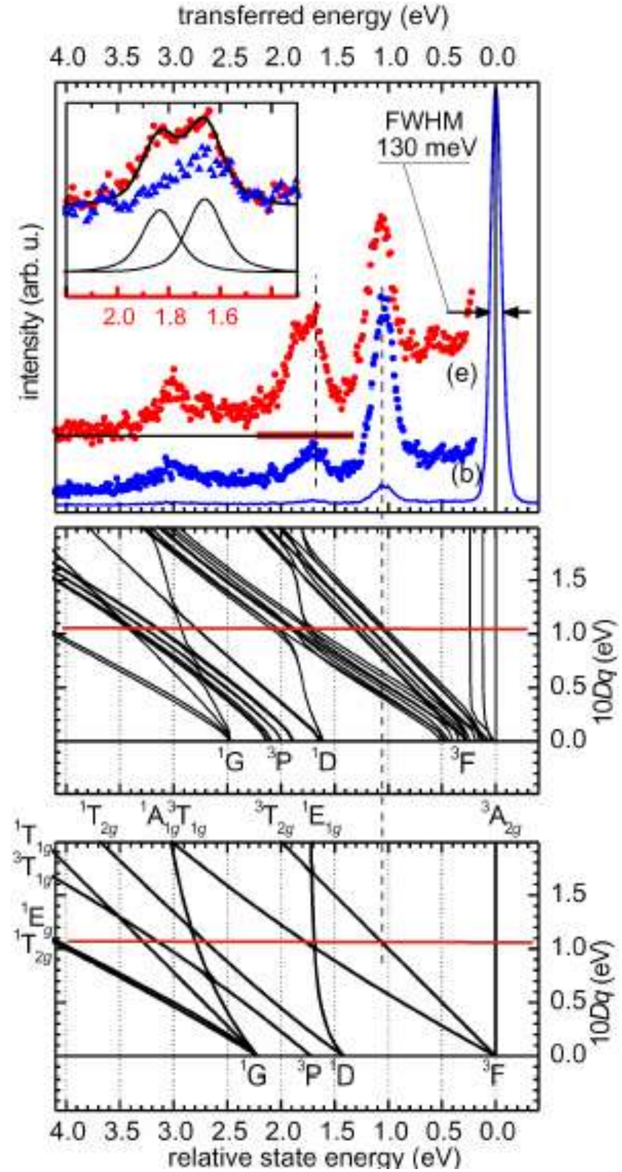
Crystal field model: Sugano-Tanabe diagrams





Single ion
Octahedral C.F.
3d spin-orbit
Exchange

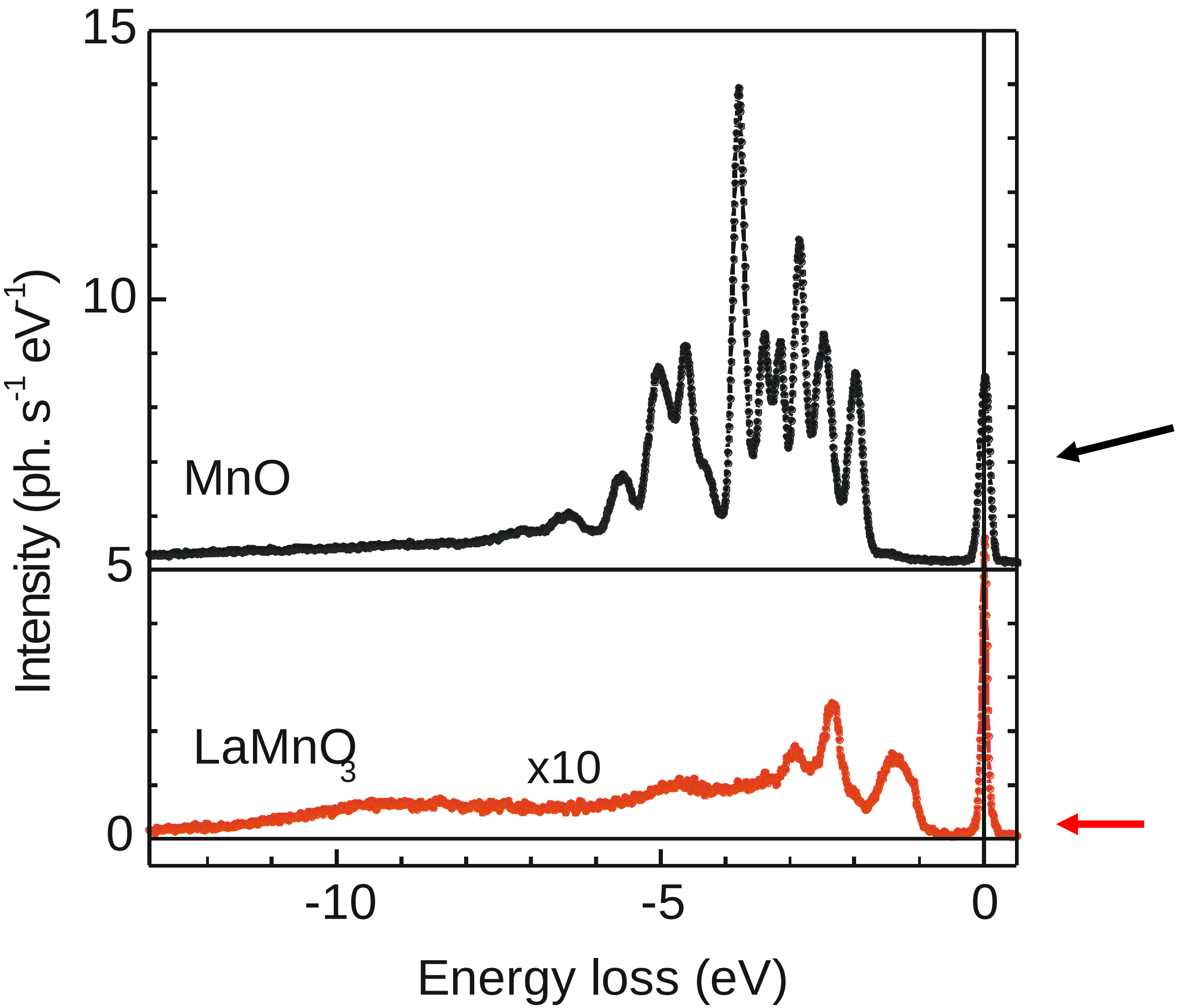
Single ion
Octahedral C.F.



G. Ghiringhelli *et al*, J. Phys. Cond. Mat. **17**, 5397 (2005)

S.G.Chiuzbaian, G. Ghiringhelli *et al*, Phys. Rev. Lett. **95**, 197402 (2005)

Mn L_3 edge: MnO, $LaMnO_3$

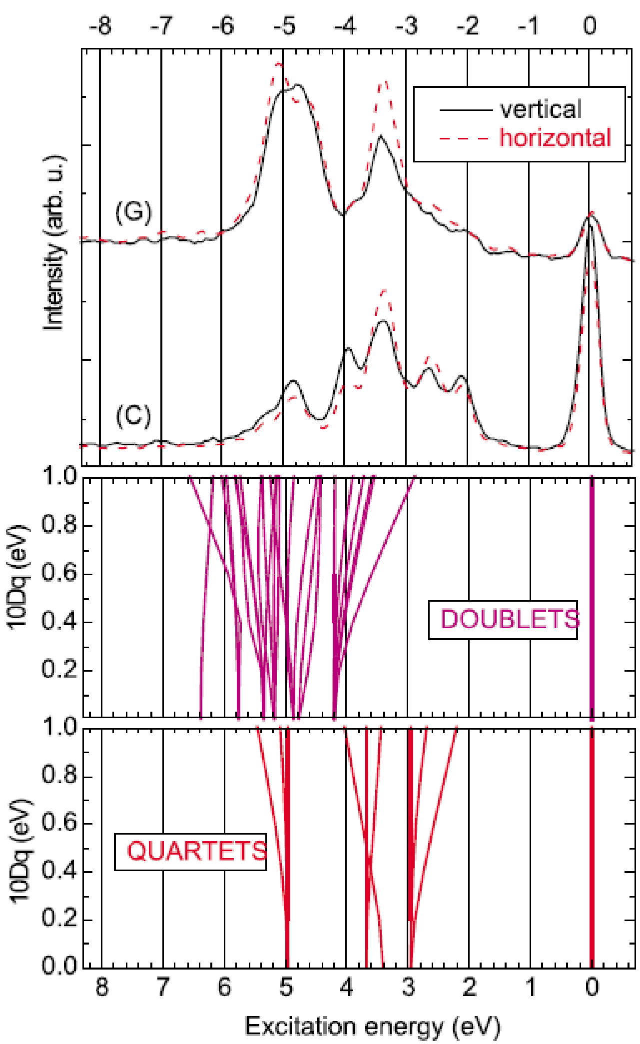


Mn^{2+} and Mn^{3+} in octahedral coordination

Mn^{2+}: $3d^5$

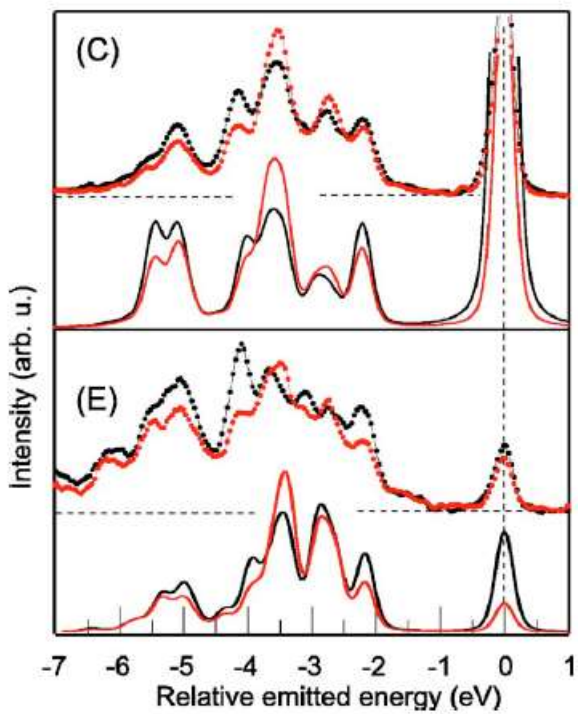
Mn^{3+}: $3d^4$

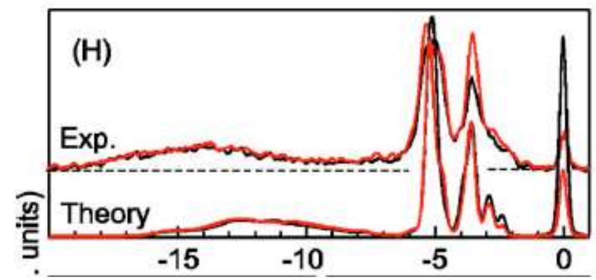
dd of Mn^{2+}: Sugano-Tanabe, Single ion, Single Ion Impurity Model

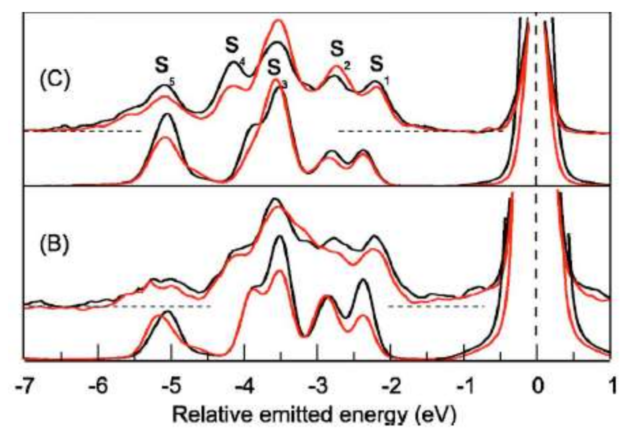


G. Ghiringhelli et al, PRB **78**, 117102 (2008)

Ground state: $3d^5$, high spin (^6S for $10Dq$=0)



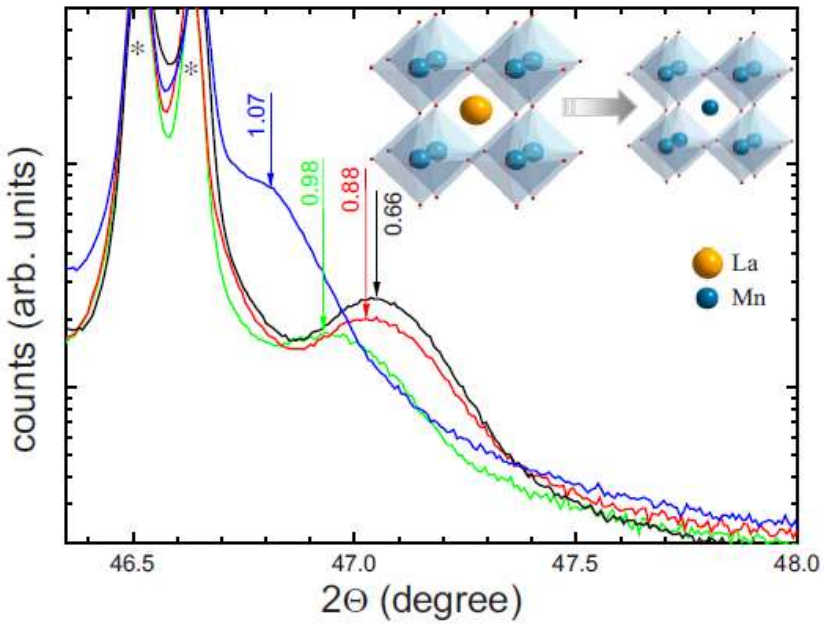


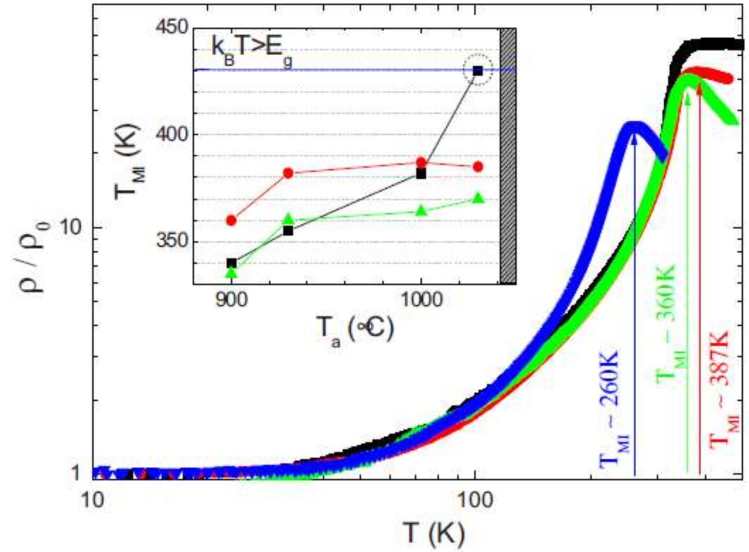


G. Ghiringhelli et al, PRB **73**, 035111 (2006)

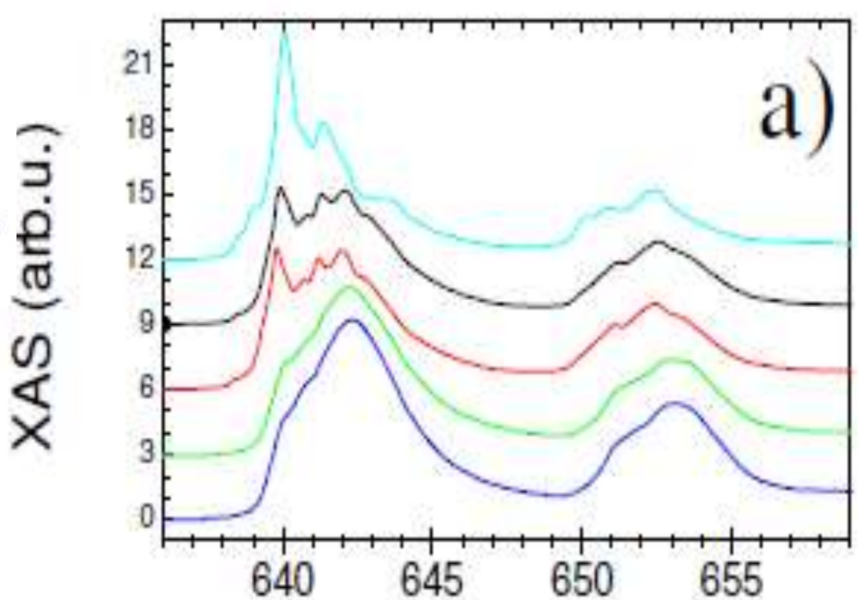
An application to thin film: Mn^{2+} in La_xMnO_3

$La_xMnO_{3-\delta}$/STO films
x=La/Mn ratio
for x<1 becomes FM (self doping)





XAS reveals the presence of Mn^{2+} for x<1

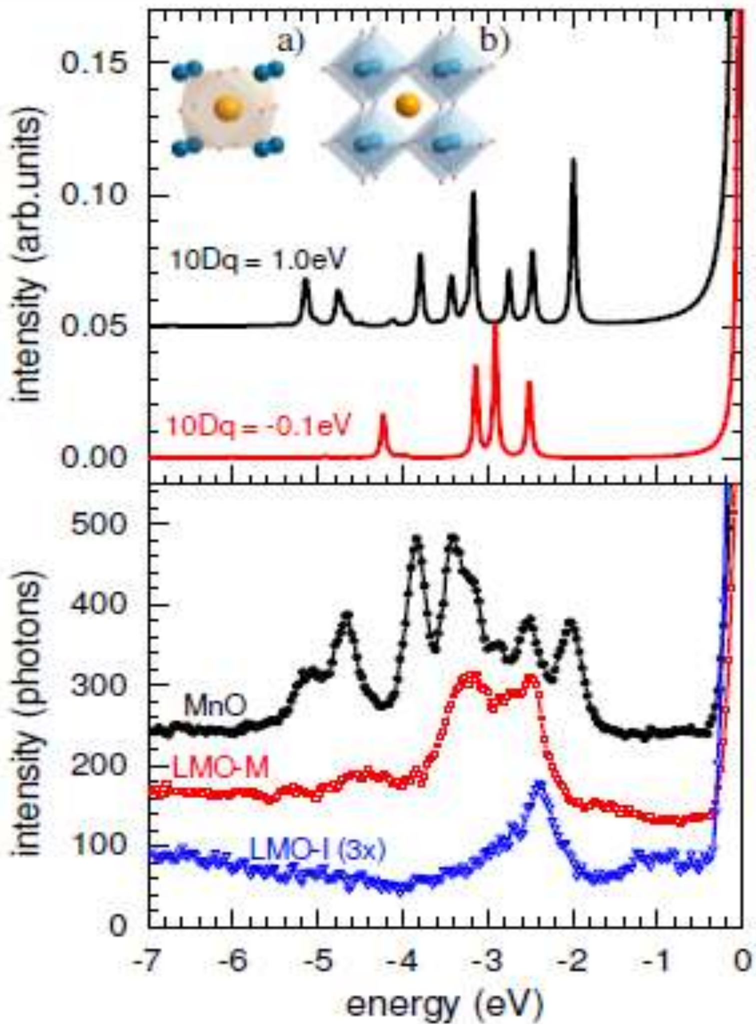


MnO
x=0.66
x=0.88
x=0.98
x=1.07

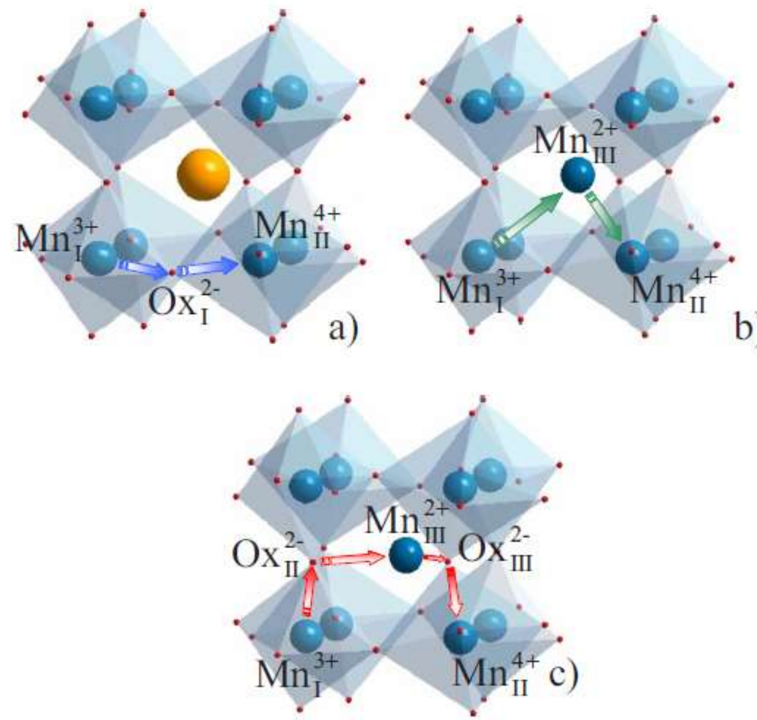
P. Orgiani, A. Galdi, C. Aruta, V. Cataudella, G. De Filippis, C.A. Perroni, V. Marigliano Ramaglia, R. Ciancio, N.B. Brookes, M. Moretti Sala, G. Ghiringhelli, and L. Maritato, Phys. Rev. B **82**, 205122 (2010)

An application to thin film: Mn^{2+} in La_xMnO_3

RIXS shows that Mn^{2+} is at site A, ie, it replaces La^{3+}



The Mn^{2+} in site A allows new Double Exchange paths, increasing T_{MI}



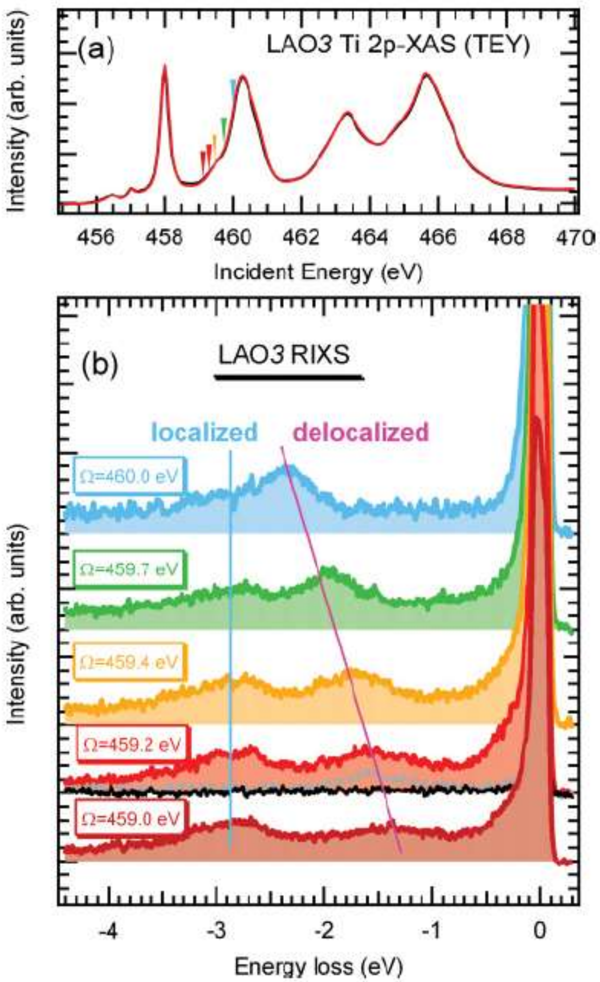
P. Orgiani, A. Galdi, C. Aruta, V. Cataudella, G. De Filippis, C.A. Perroni, V. Marigliano Ramaglia, R. Ciancio, N.B. Brookes, M. Moretti Sala, G. Ghiringhelli, and L. Maritato, Phys. Rev. B **82**, 205122 (2010)

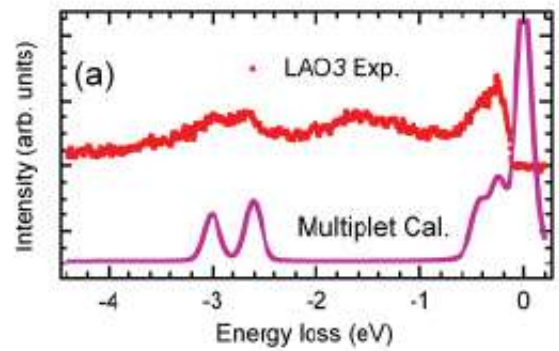
STO/LAO superlattice: RIXS at Ti L_3

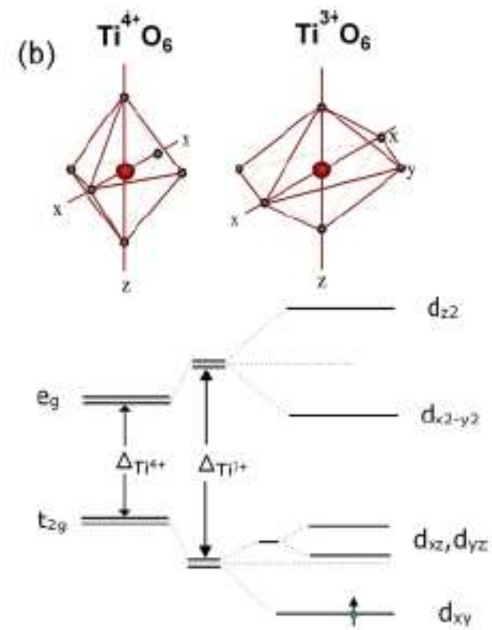
PHYSICAL REVIEW B 83, 201402(R) (2011)

Localized and delocalized Ti 3*d* carriers in $LaAlO_3/SrTiO_3$ superlattices revealed by resonant inelastic x-ray scattering

Ke-Jin Zhou,[1] Milan Radovic,[2,1] Justine Schlappa,[1,*] Vladimir Strocov,[1] Ruggero Frison,[3] Joel Mesot,[1,2]
Luc Patthey,[1] and Thorsten Schmitt[1,†]





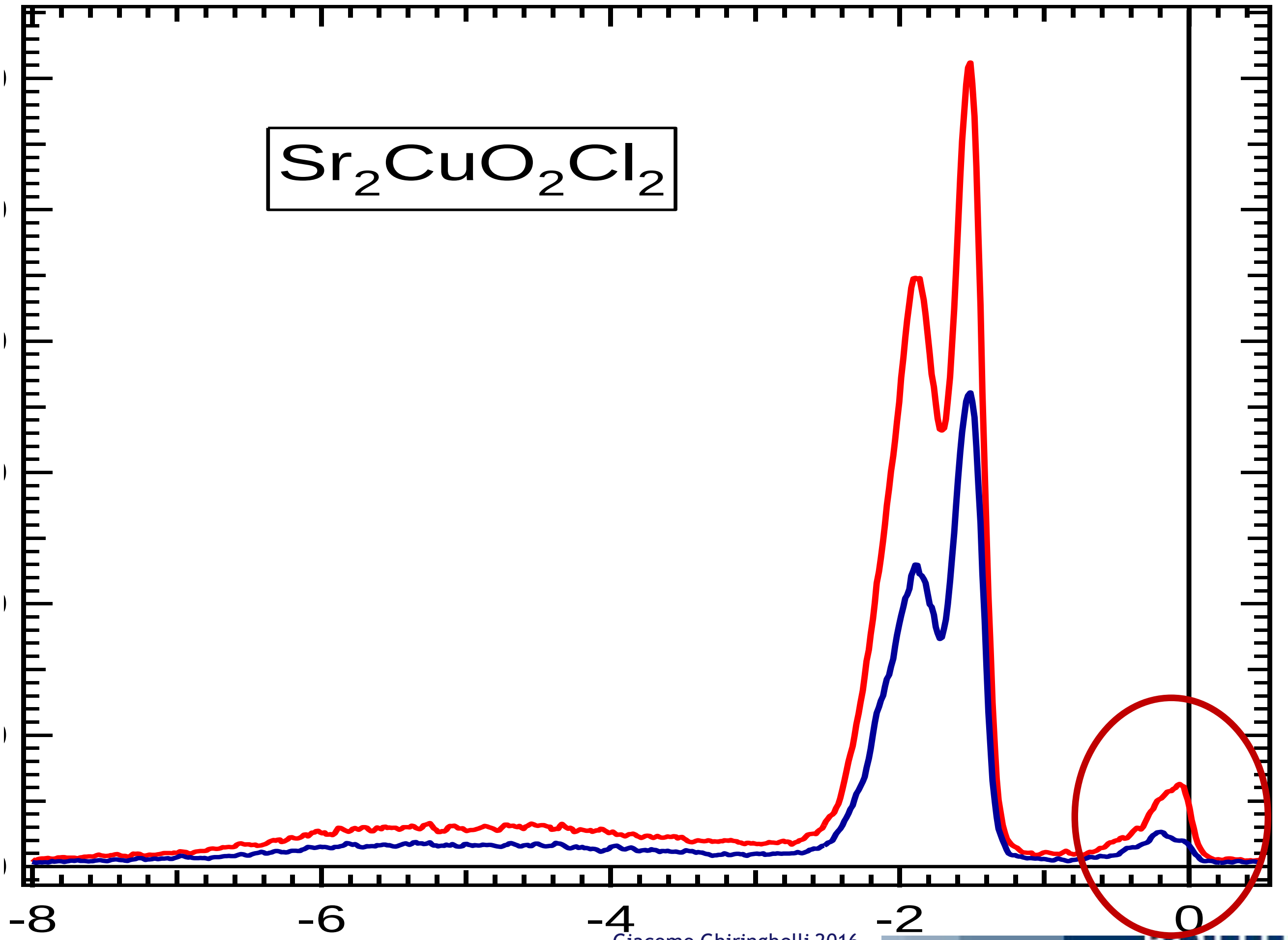


What about the “quasi-elastic” spectral features?

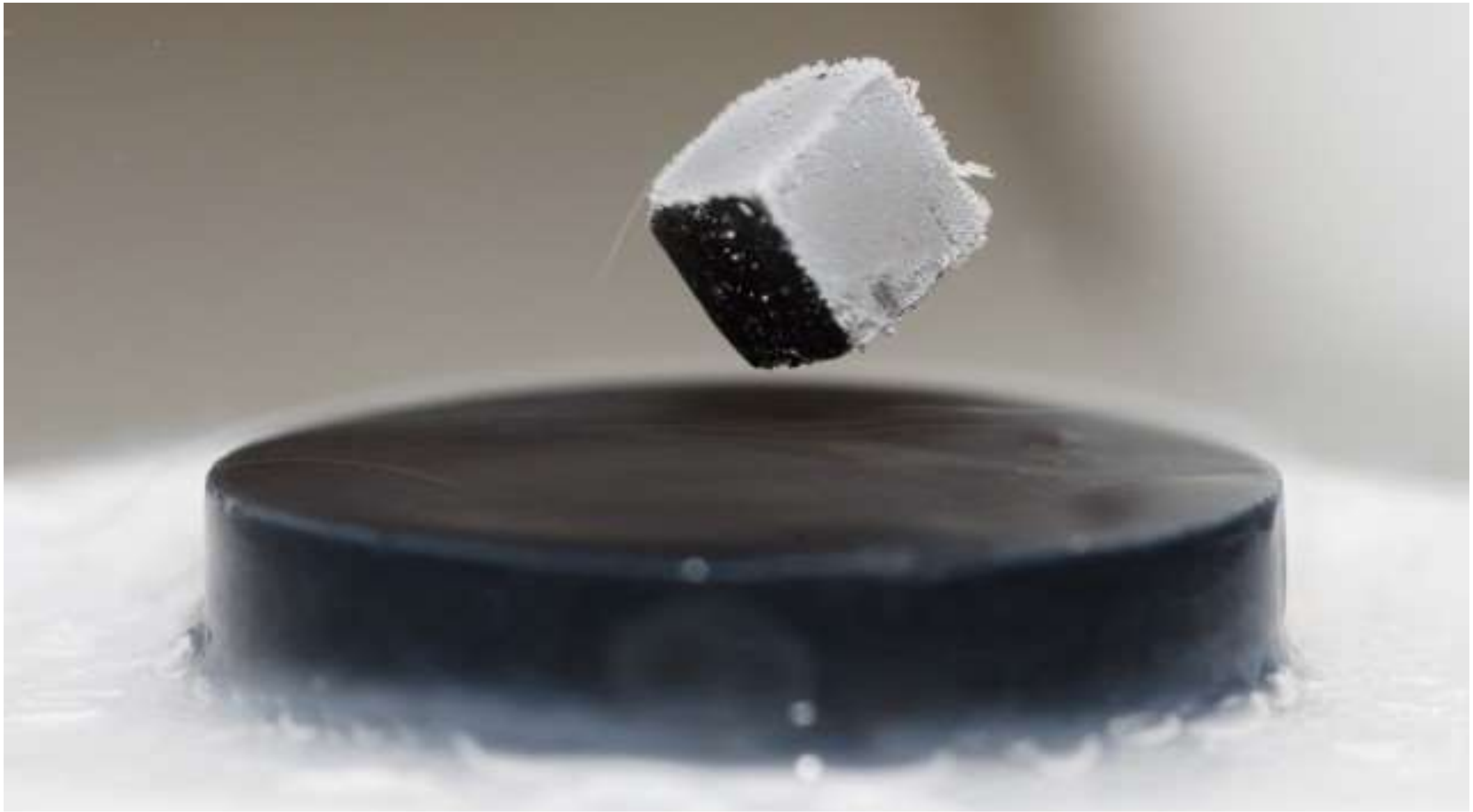
Phonons: up to 90meV

Magnons (2*J* at BZB): up to 300 meV ($J_{eff} \approx 140$ meV)

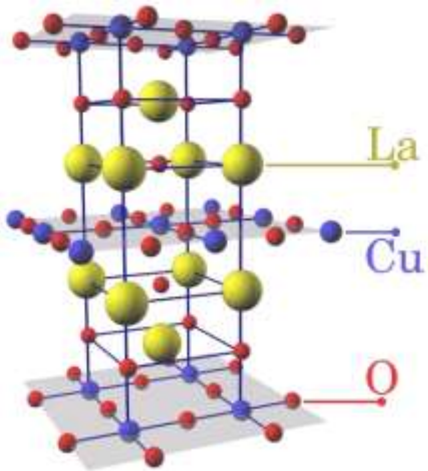
Multi mangons…



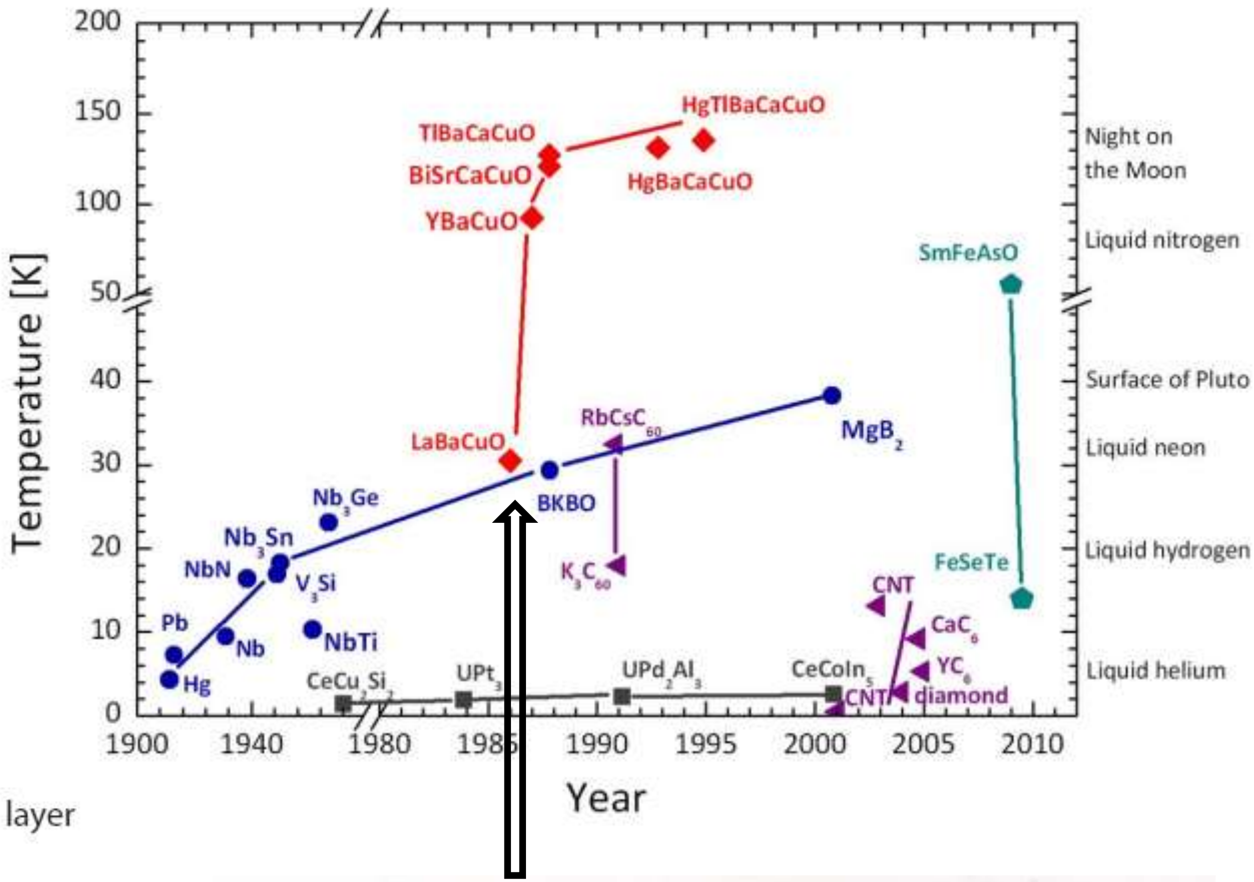
High Tc superconductors

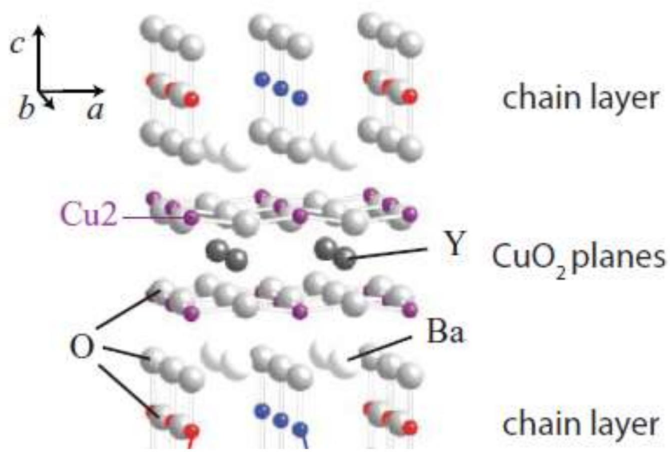


High Tc superconducting cuprates

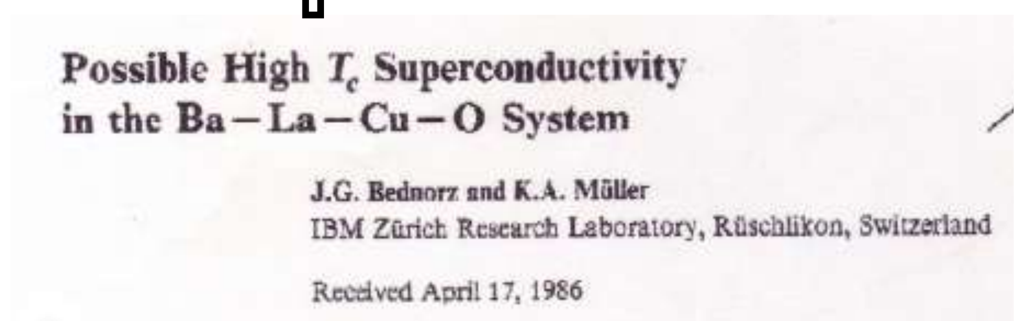


$La_{2-x}Sr_xCuO_4$ (LSCO)





$YBa_2Cu_3O_{6+\delta}$ (YBCO)

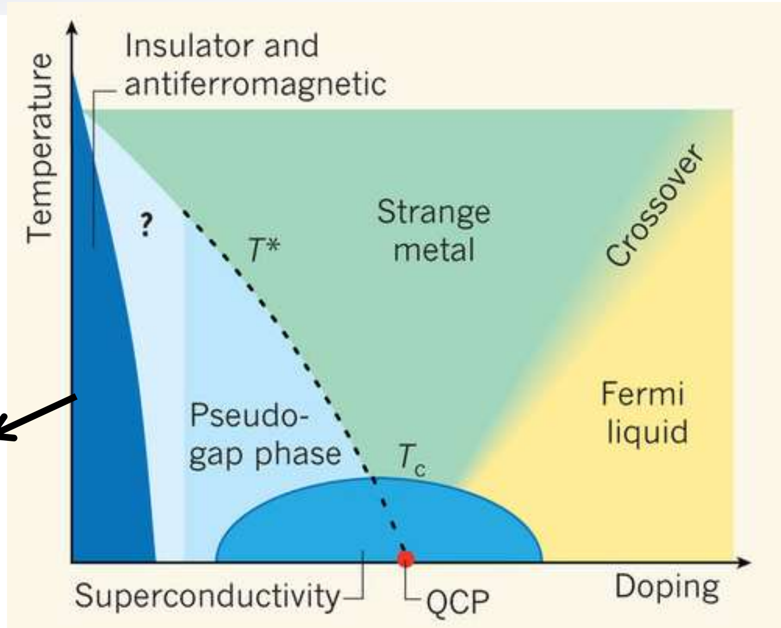


Possible High T_c Superconductivity in the Ba–La–Cu–O System

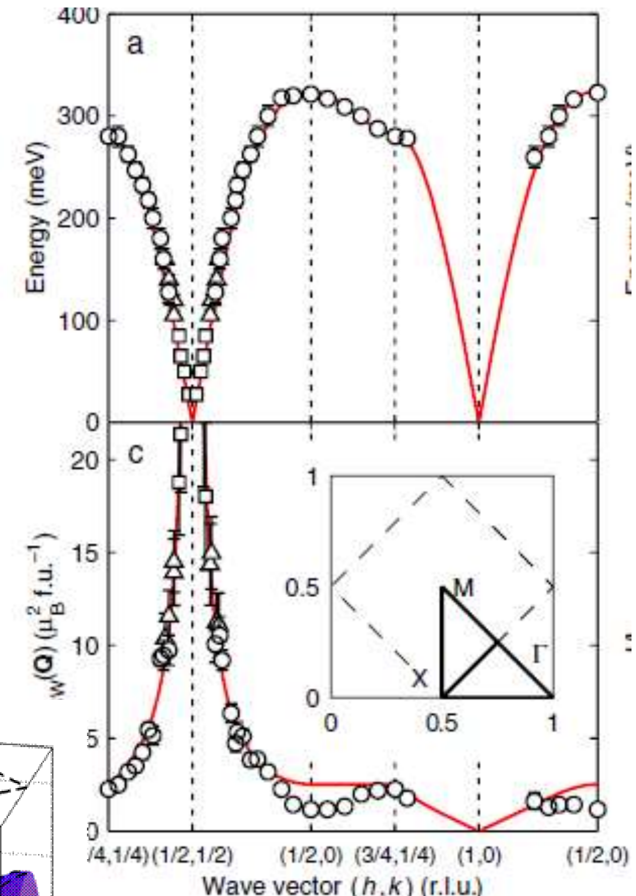
J.G. Bednorz and K.A. Müller
IBM Zürich Research Laboratory, Rüschlikon, Switzerland

Received April 17, 1986

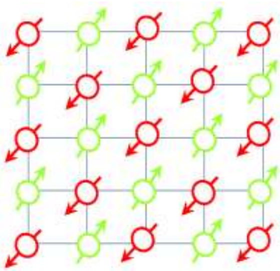
Spin excitations in HTcS: undoped AF



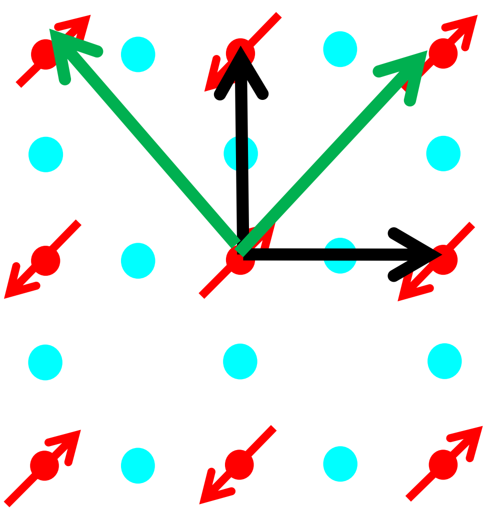
INS: La_2CuO_4



2D AF

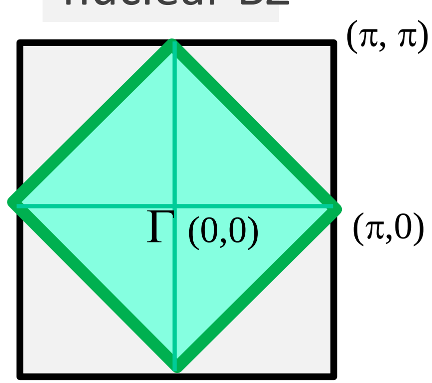


DIRECT SPACE

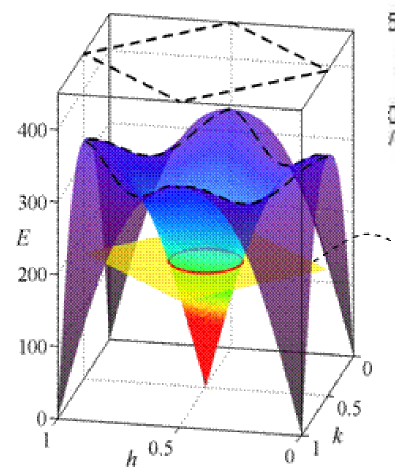


RECIPROCAL SPACE

nuclear BZ

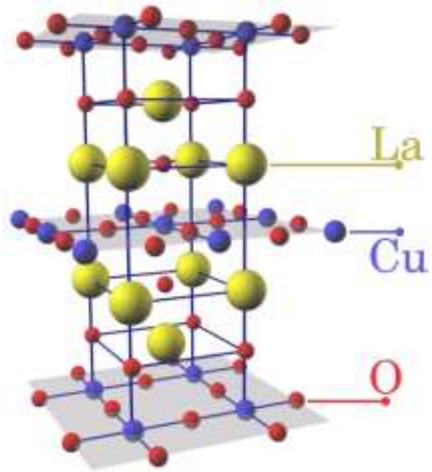


magnetic BZ

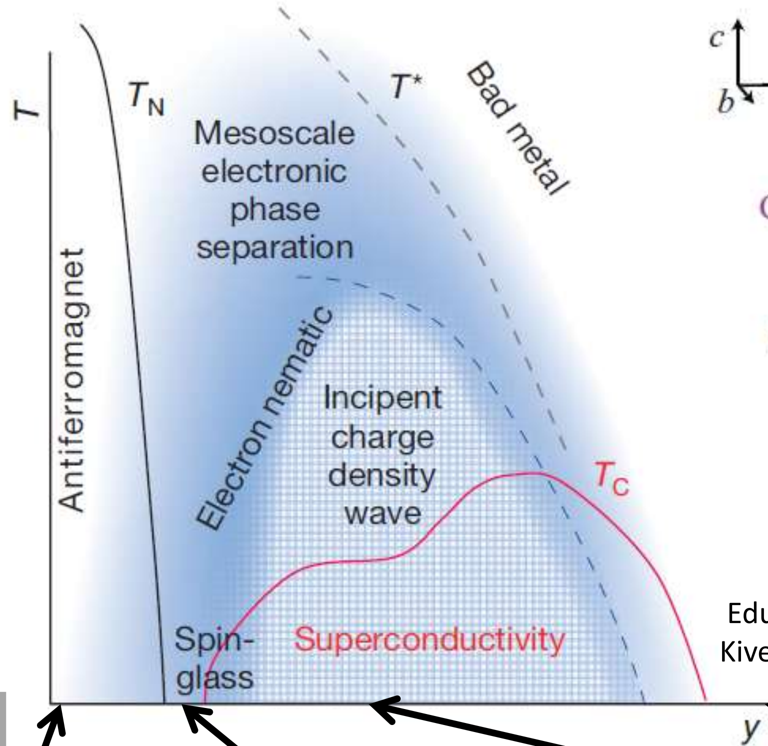


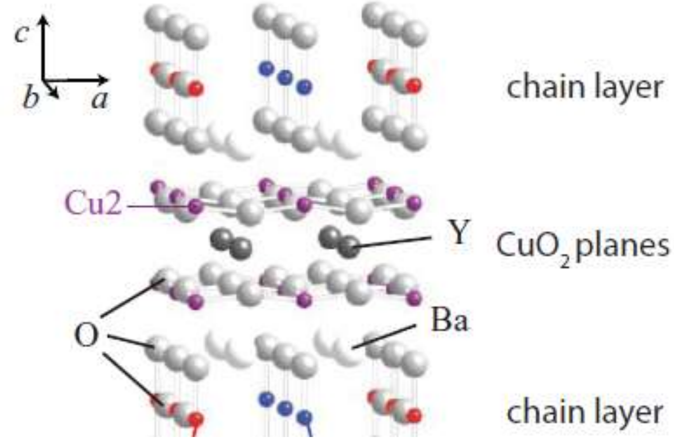
N. S. Headings, S. M. Hayden, R. Coldea, and T. G. Perring, Phys Rev Lett. **105** 247001 (2011)

The mysteries of HT_cS



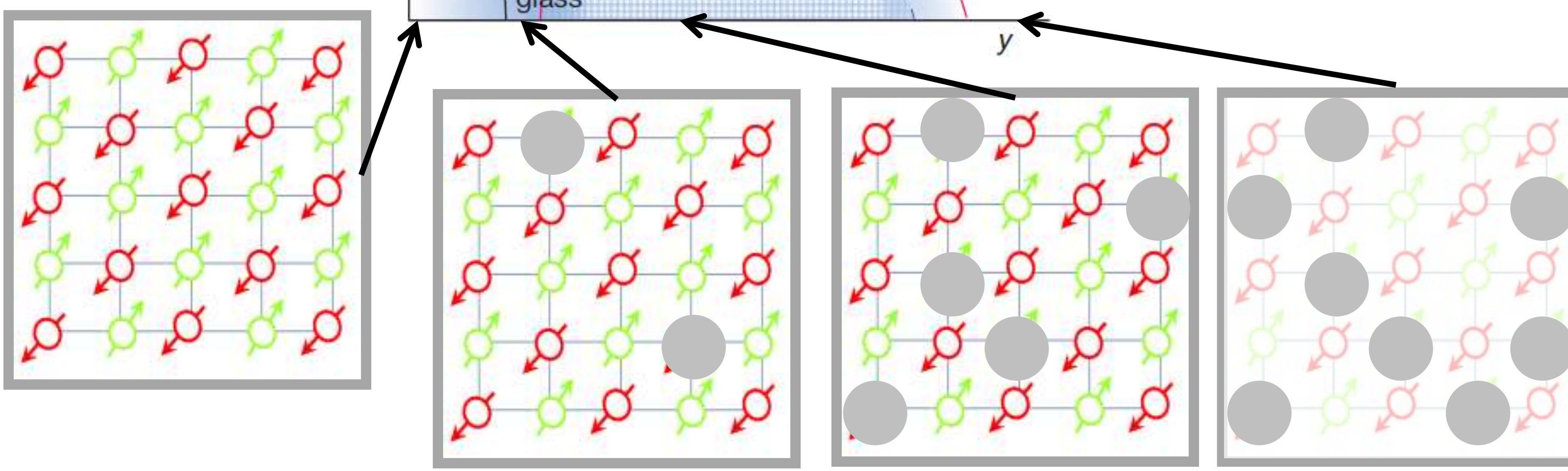
$La_{2-x}Sr_xCuO_4$ (LSCO)



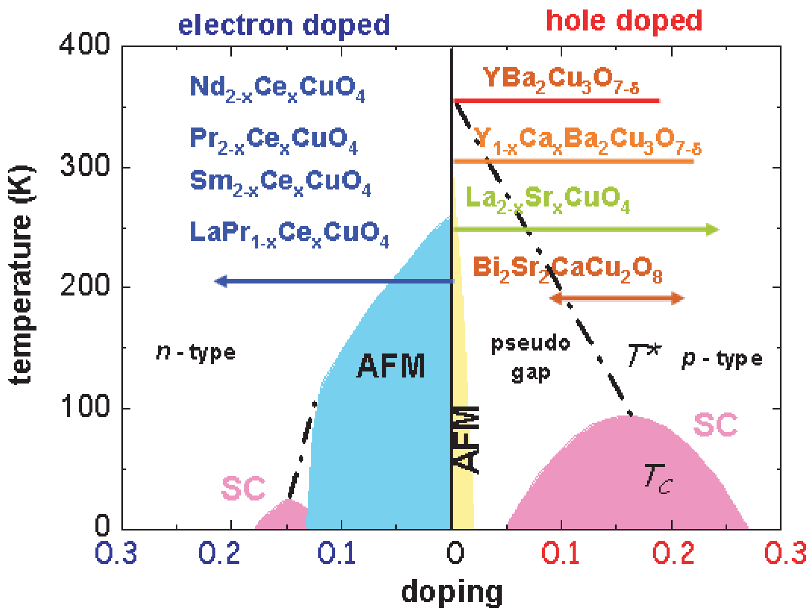


$YBa_2Cu_3O_{6+\delta}$ (YBCO)

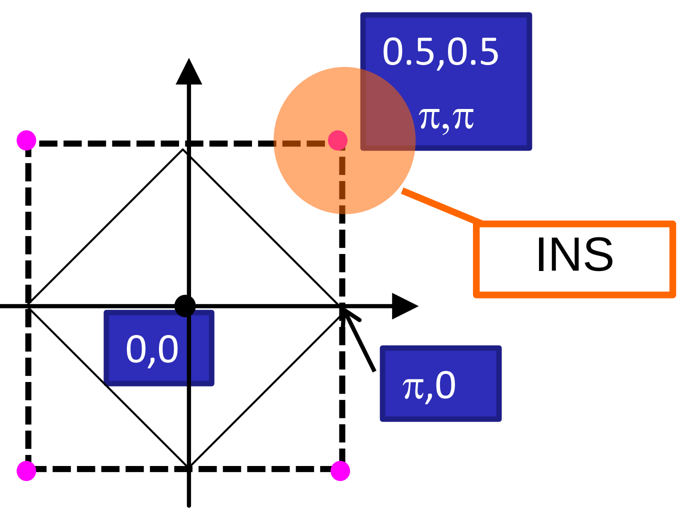
Eduardo Fradkin and Steven A. Kivelson, Nature Physics, 8, 864 (2012)

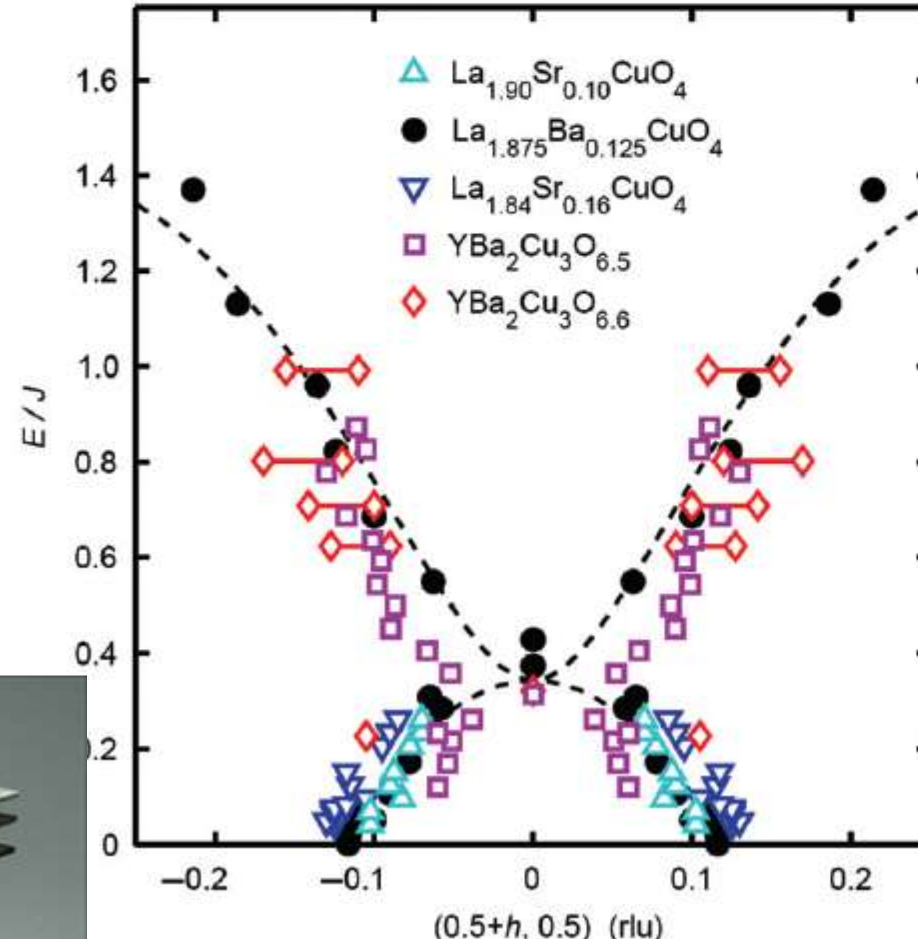


Spin excitations in HTcS: doped SC



http://for538.wmi.badw.de/projects/P4_crystal_growth/index.htm

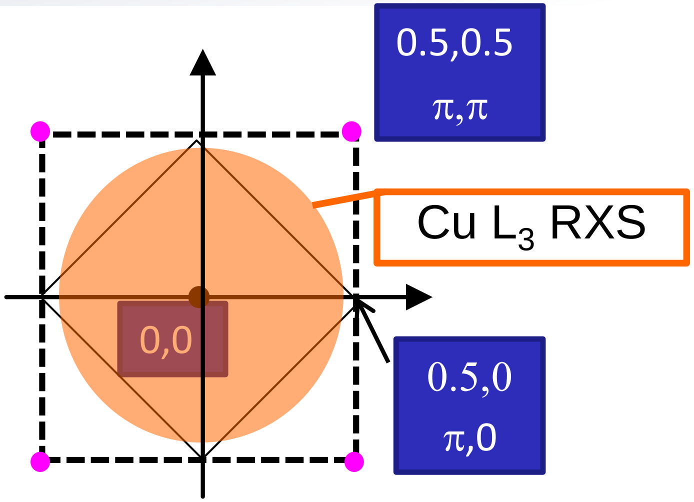




J.M. Tranquada, in *Handbook of High-Temperature Superconductivity: Theory and Experiment*, J.R. Schrieffer and J.S. Brooks, eds., Springer, 2007,

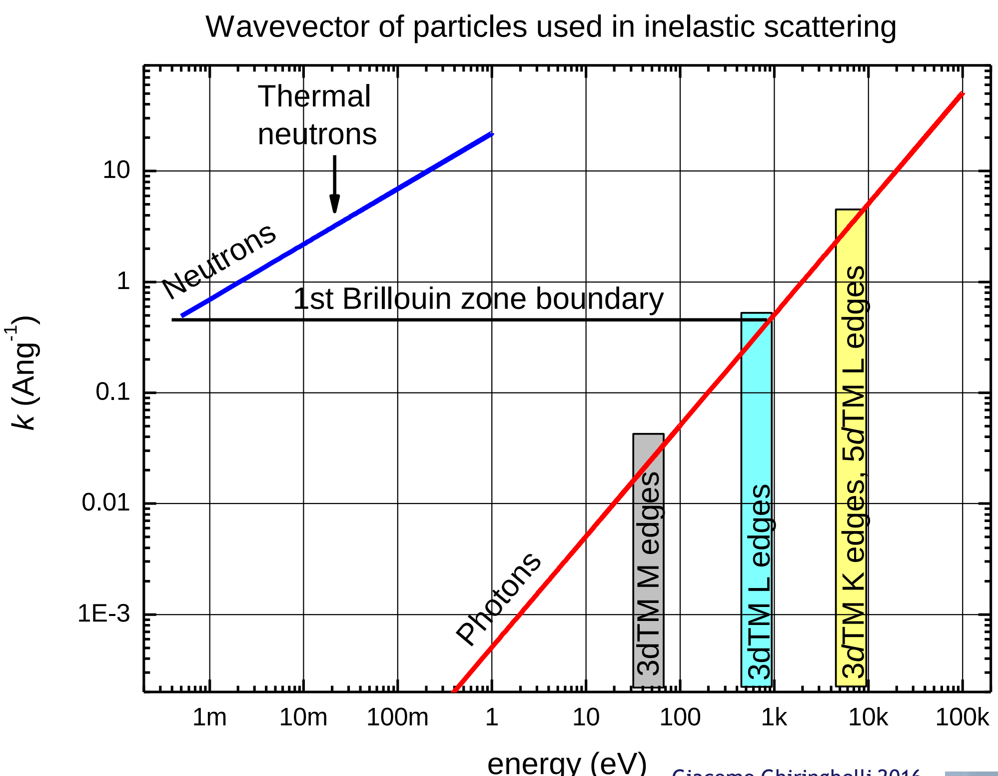
V. Hinkov et al, Eur. Phys. J. Special Topics 188, 113–129 (2010)

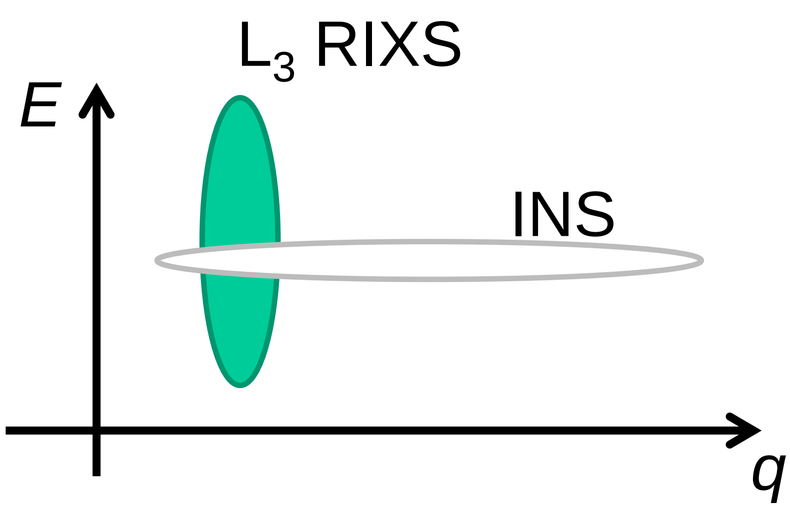
RIXS: Experimental conditions



Cu L_3 resonance:

- E_0 = 930 eV
- q_{max} = 0.86 Ang^{-1}
- confined inside a region around Γ
- 2p core hole: spin-orbit interaction
- E resolution: 120-240 meV
- *q* resolution: 0.005 rlu
- ½ - 1 hour per spectrum





spin-flip excitations and the 2p S-O coupling

3d	E_g	$d_{3z^2-r^2}$	$Y_{2,0}$
		$d_{x^2-y^2}$	$\frac{Y_{2,2}+Y_{2,-2}}{\sqrt{2}}$
	T_{2g}	d_{xy}	$-i\frac{Y_{2,2}-Y_{2,-2}}{\sqrt{2}}$
		d_{yz}	$i\frac{Y_{2,1}+Y_{2,-1}}{\sqrt{2}}$
		d_{zx}	$-\frac{Y_{2,1}-Y_{2,-1}}{\sqrt{2}}$

The 3d spin must not be pure UP or DOWN

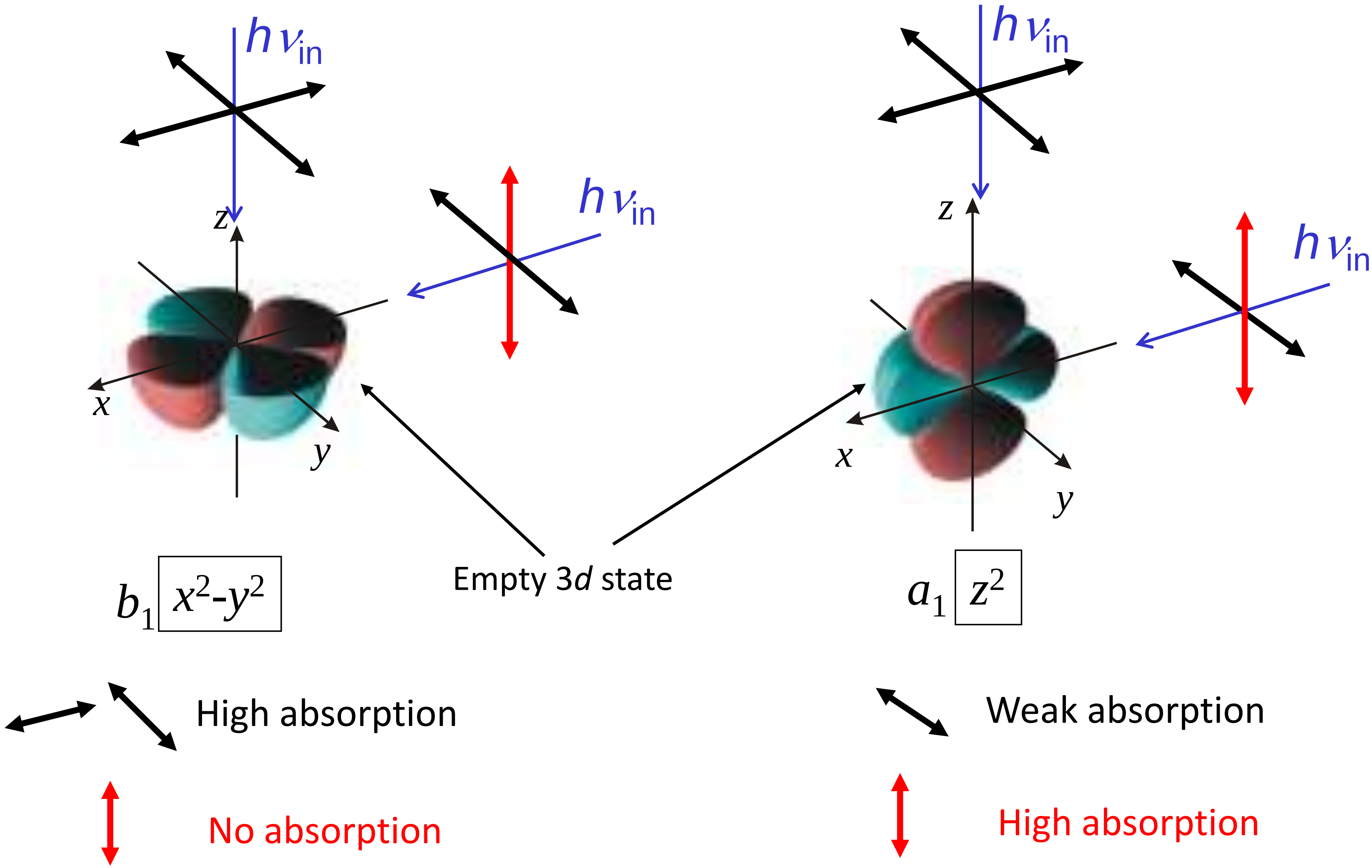
$2p_{3/2}$

$\left\|\frac{3}{2},\frac{3}{2}\right\rangle$	$\left\|\frac{3}{2},\frac{1}{2}\right\rangle$	$\left\|\frac{3}{2},-\frac{1}{2}\right\rangle$	$\left\|\frac{3}{2},-\frac{3}{2}\right\rangle$
$Y^{\uparrow}{}_{1,1}$	$\frac{\sqrt{2}}{\sqrt{3}}Y^{\uparrow}_{1,0}+\frac{1}{\sqrt{3}}Y^{\downarrow}_{1,1}$	$\frac{1}{\sqrt{3}}Y^{\uparrow}_{1,-1}+\frac{\sqrt{2}}{\sqrt{3}}Y^{\downarrow}_{1,0}$	$Y^{\downarrow}_{1,-1}$

$2p_{1/2}$

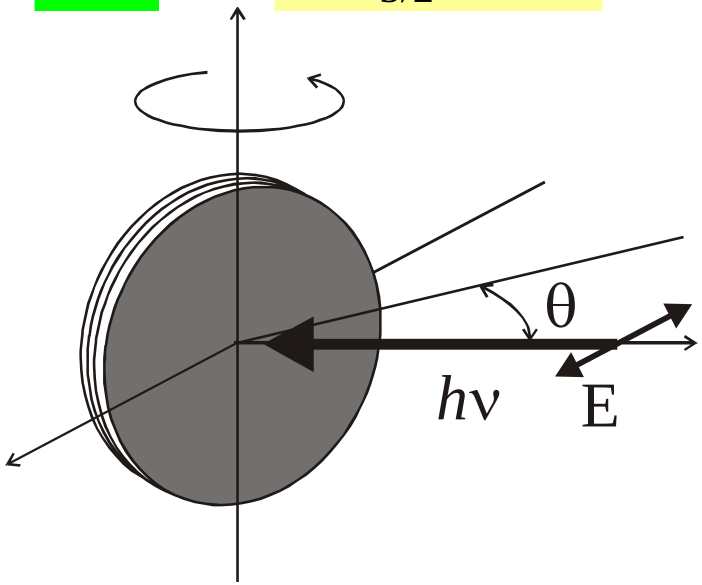
$\left\|\frac{1}{2},\frac{1}{2}\right\rangle$	$\left\|\frac{1}{2},-\frac{1}{2}\right\rangle$
$-\frac{1}{\sqrt{3}}Y^{\uparrow}_{1,0}+\frac{\sqrt{2}}{\sqrt{3}}Y^{\downarrow}_{1,1}$	$-\frac{\sqrt{2}}{\sqrt{3}}Y^{\uparrow}_{1,-1}+\frac{1}{\sqrt{3}}Y^{\downarrow}_{1,0}$

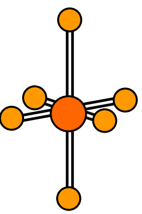
Linear polarization of x-rays and orbital orientation



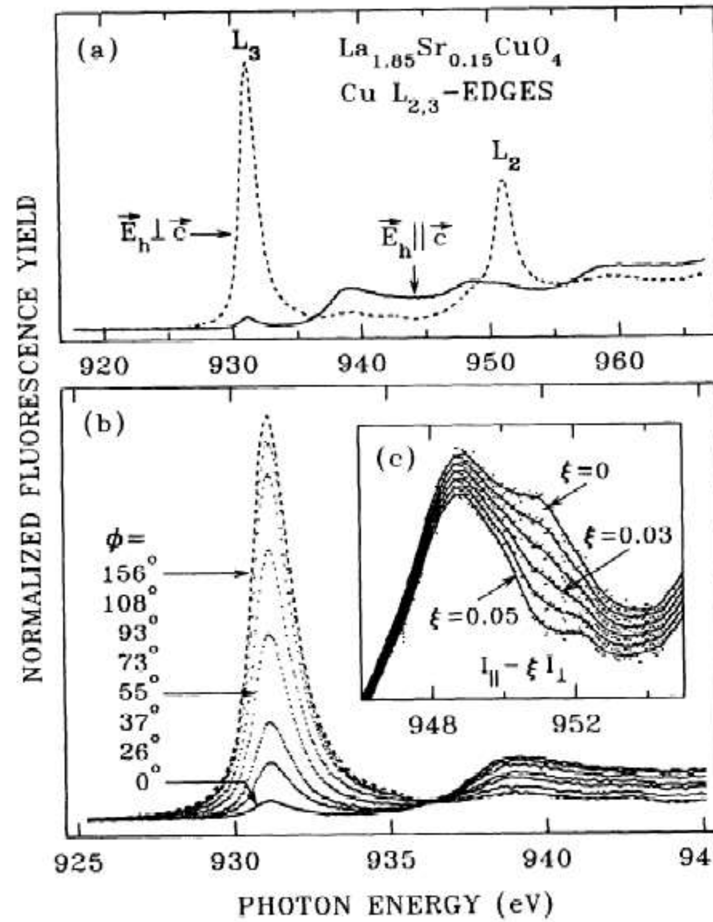
3*d* hole symmetry in cuprates

$3d^9 \rightarrow (2p_{3/2})^3 3d^{10}$





Result: the hole in Cu^{2+} has 100% x^2-y^2 symmetry



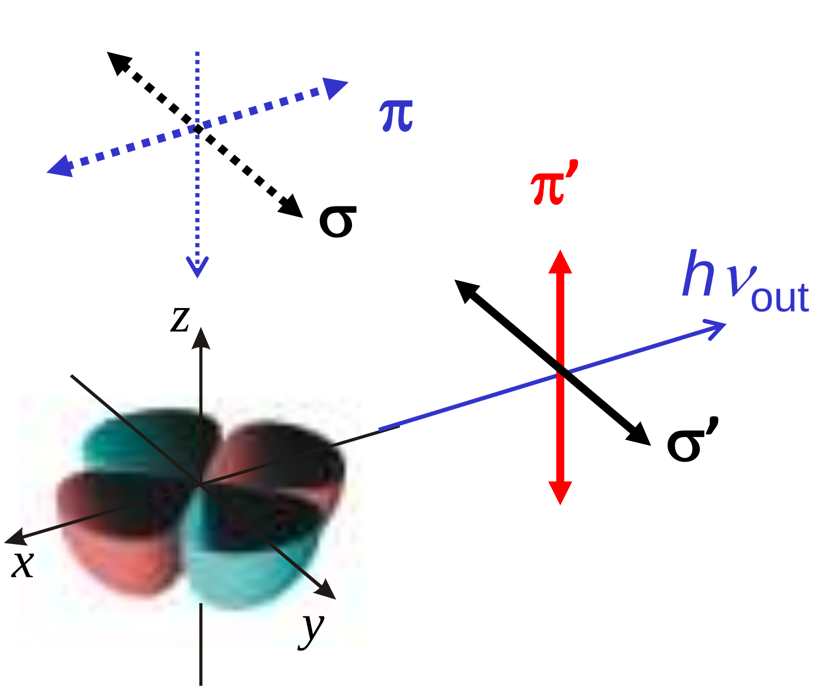
VOLUME 68, NUMBER 16 PHYSICAL REVIEW LETTERS 20 APRIL 1992

Out-of-Plane Orbital Characters of Intrinsic and Doped Holes in $La_{2-x}Sr_xCuO_4$

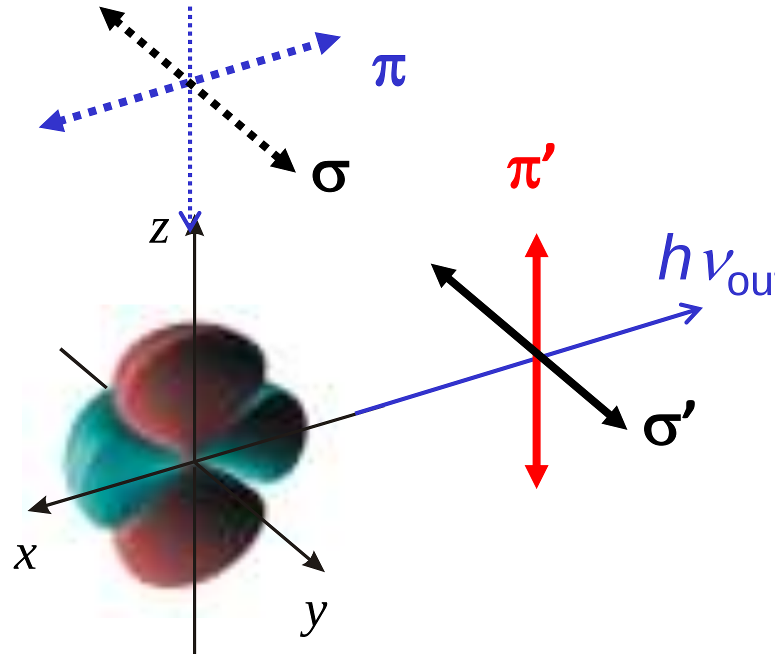
C. T. Chen, L. H. Tjeng, J. Kwo, H. L. Kao, P. Rudolf, F. Sette, and R. M. Fleming

Linear polarization of x-rays and orbital orientation (2)

The same rules hold for emission (radiative de-excitation)



b_1 x^2-y^2

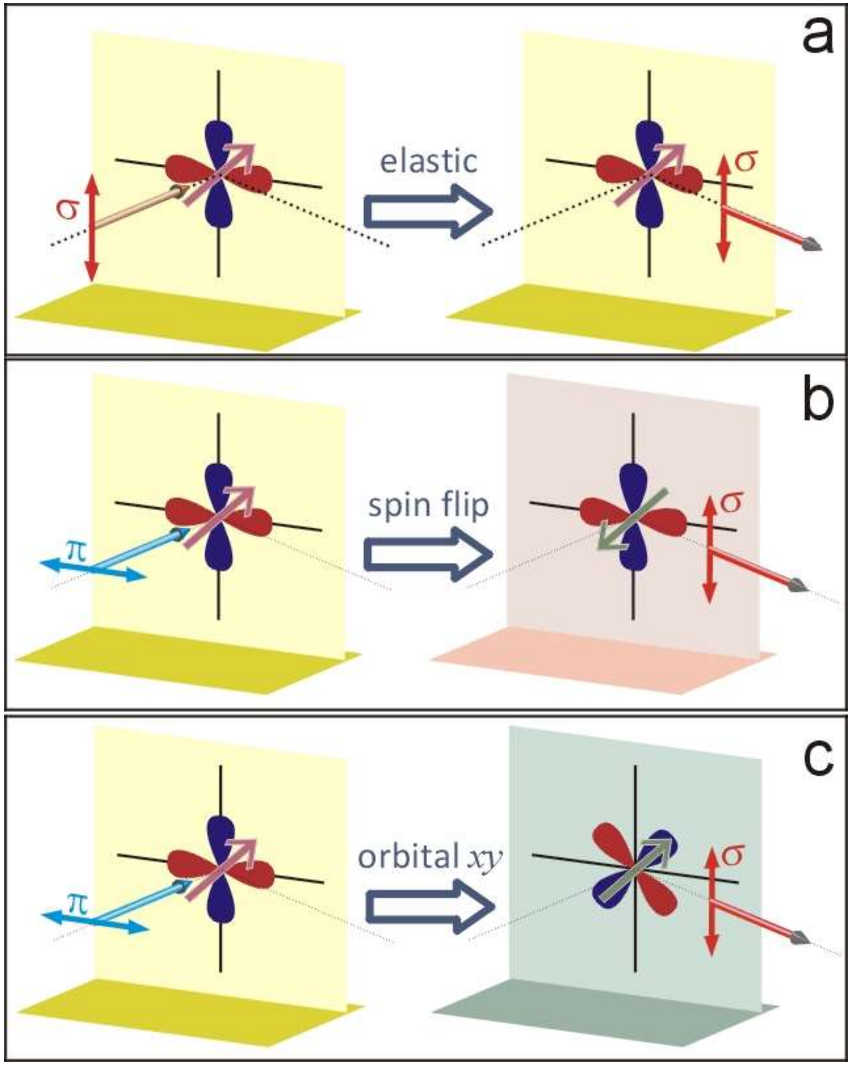


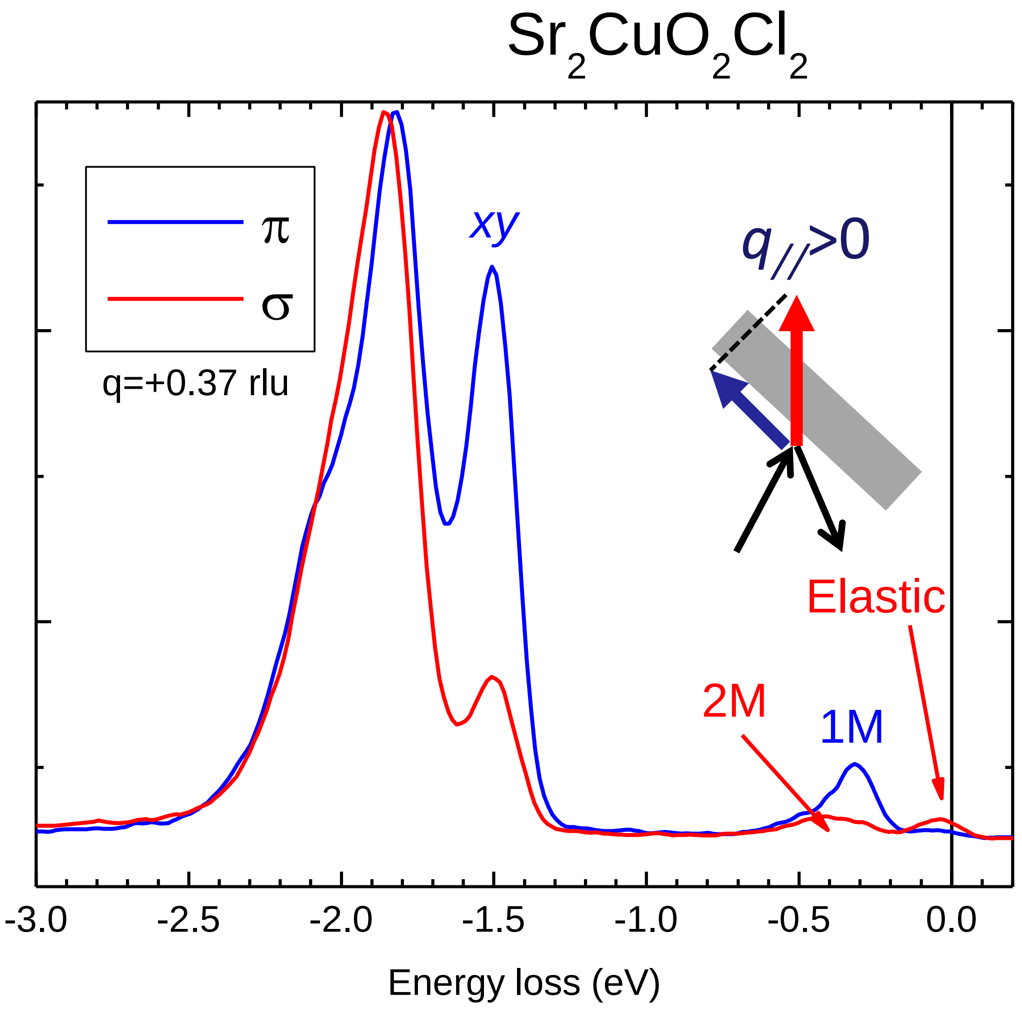
a_1 z^2

↖↘	High emission
↕	No emission

↖↘	Weak emission
↕	High emission

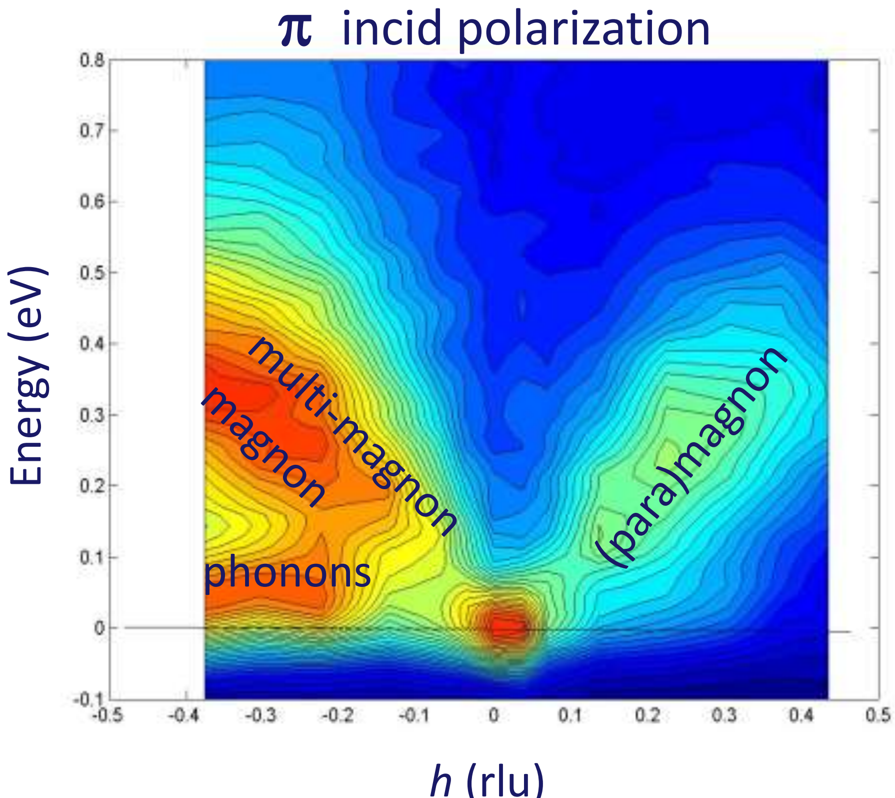
Polarization dep. of Cu L_3 RIXS intensity

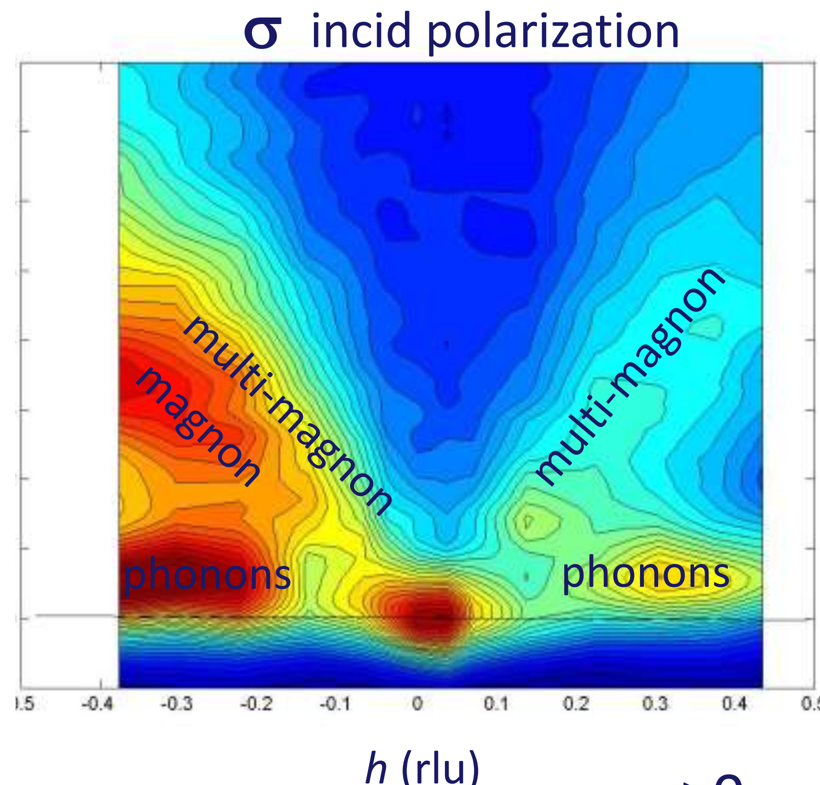


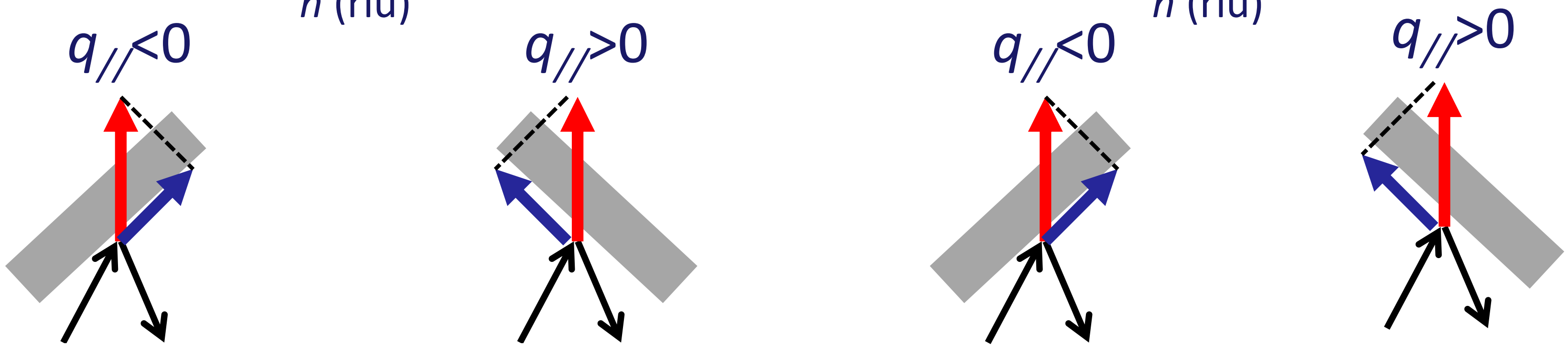


Polarization dependent cross-sections

LSCO, opt. doping

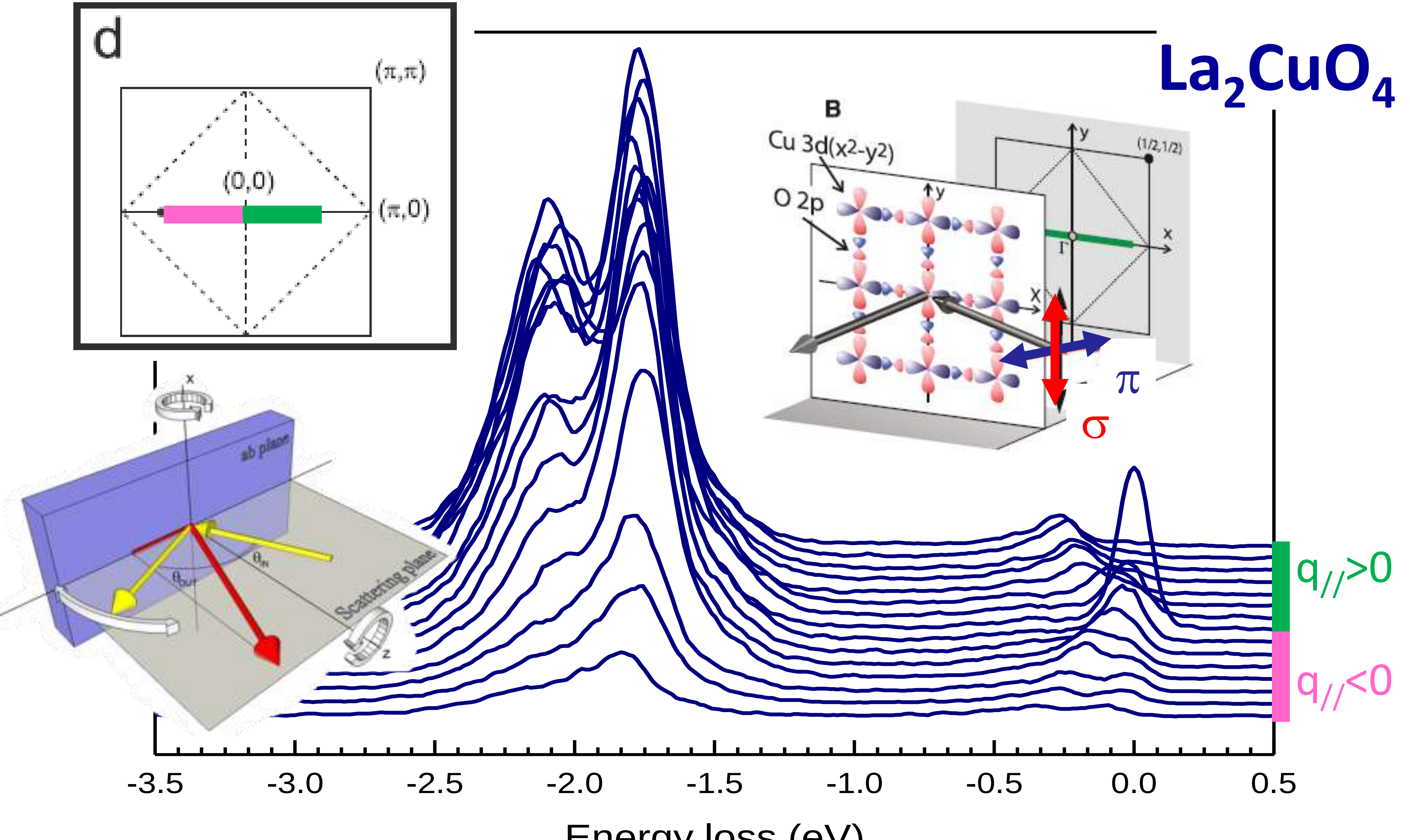






M. Hashimoto, L.Braicovich, M. Minola, GG et al. unpublished

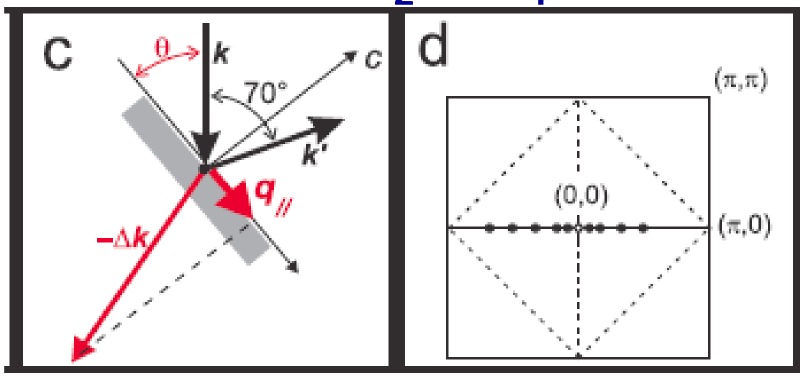
First demonstration: La_2CuO_4

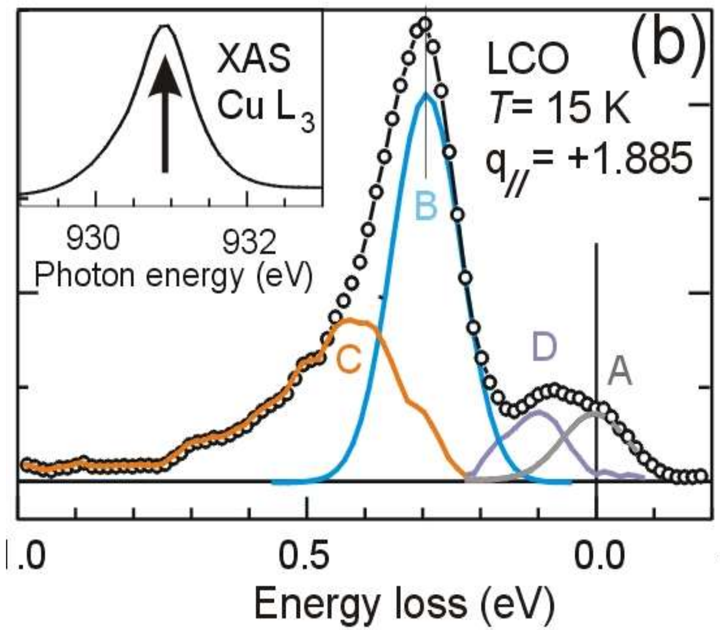


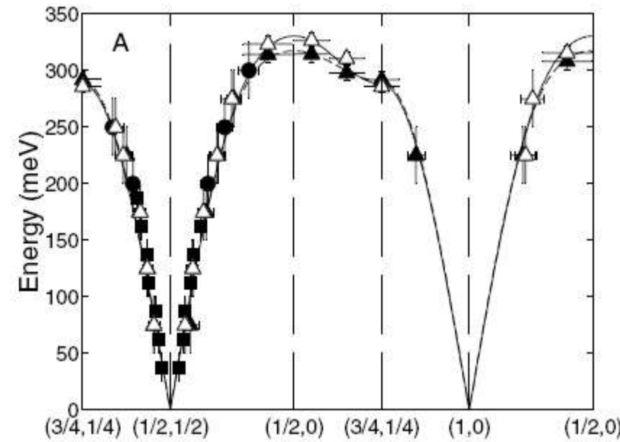
L. Braicovich, J. van den Brink, V. Bisogni, M. Moretti Sala, L. Ament, N.B. Brookes, G.M. de Luca, M. Salluzzo, T. Schmitt, and G. Ghiringhelli PRL **104** 077002 (2010)

La_2CuO_4, RIXS vs INS

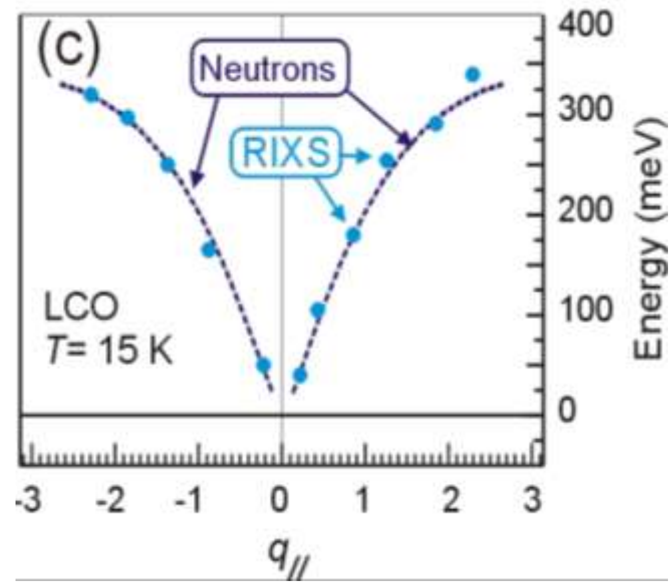
La_2CuO_4







R. Coldea et al, Phys. Rev. Lett. **86**, 5377 (2001).



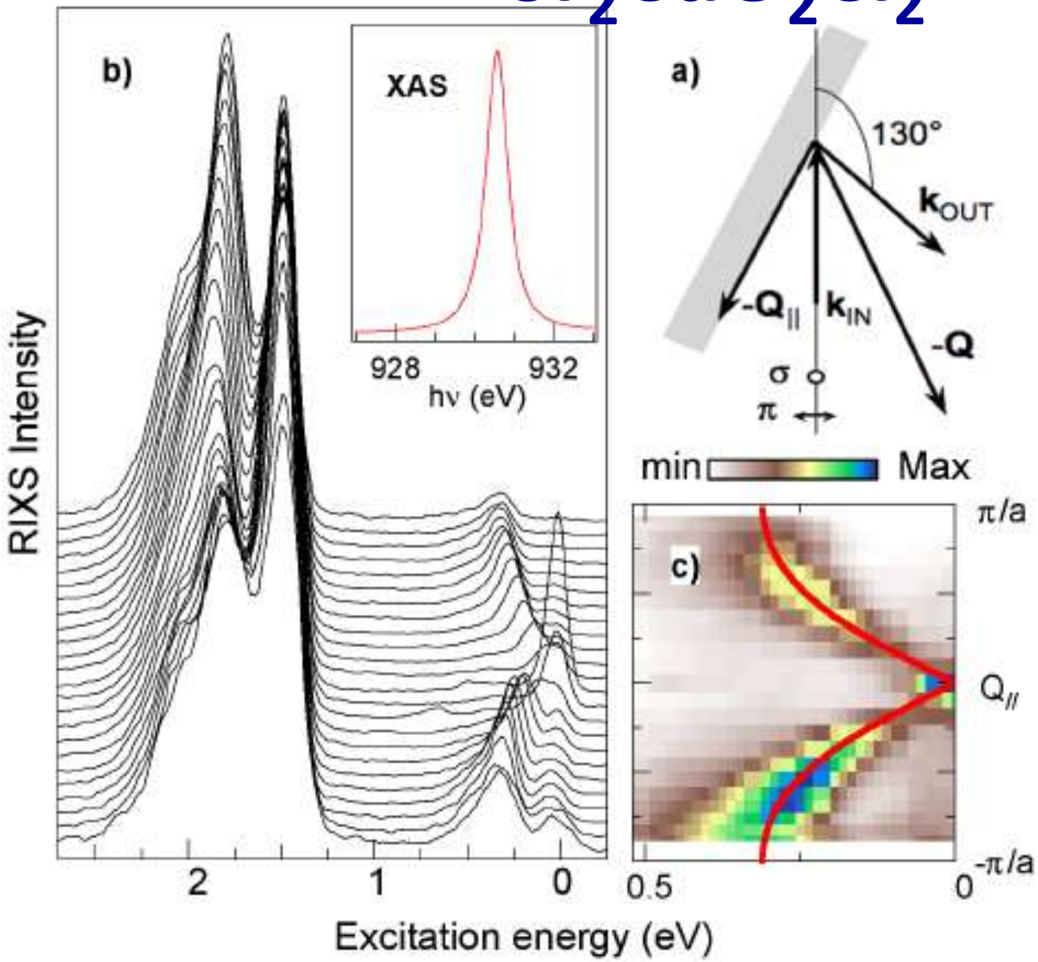
L. Braicovich, J. van den Brink, V. Bisogni, M. Moretti Sala, L. Ament, N.B. Brookes, G.M. de Luca, M. Salluzzo, T. Schmitt, and G. Ghiringhelli PRL **104** 077002 (2010)

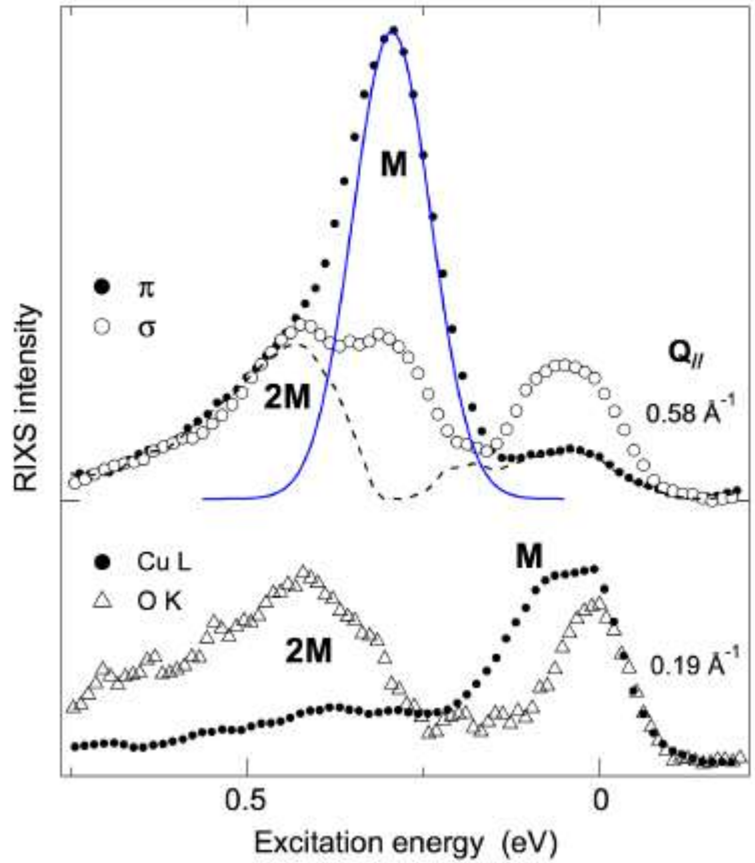
Magnetic excitations in AF cuprates

2008

$Sr_2CuO_2Cl_2$

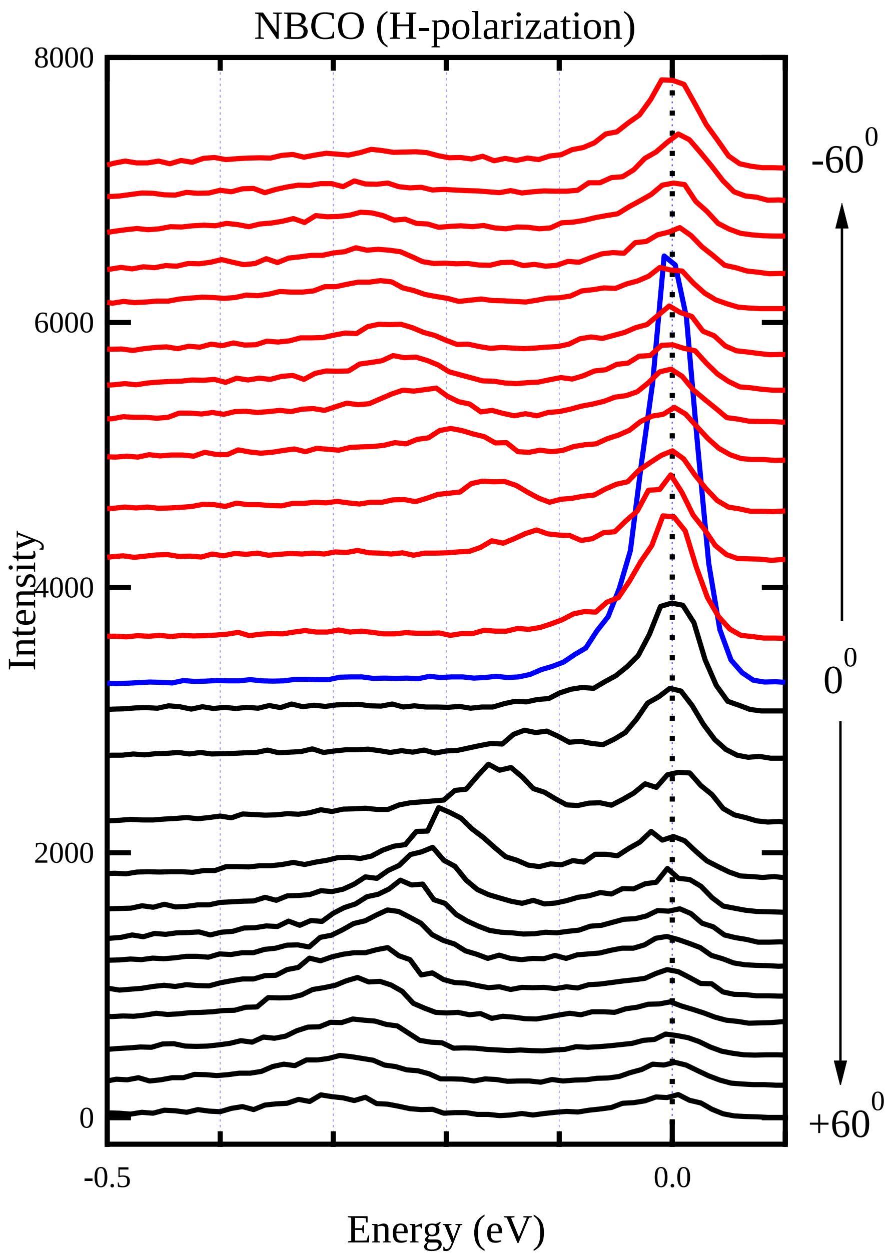
ΔE 0.12 eV





M. Guarise, B. Dalla Piazza, M. Moretti Sala, G. Ghiringhelli, L. Braicovich, H. Berger, J.N. Hancock, D. van der Marel, T. Schmitt, V.N. Strocov, L.J.P. Ament, J. van den Brink, P.-H. Lin, P. Xu, H. M. Rønnow, and M. Grioni. Phys. Rev. Lett. **105**, 157006 (2010)

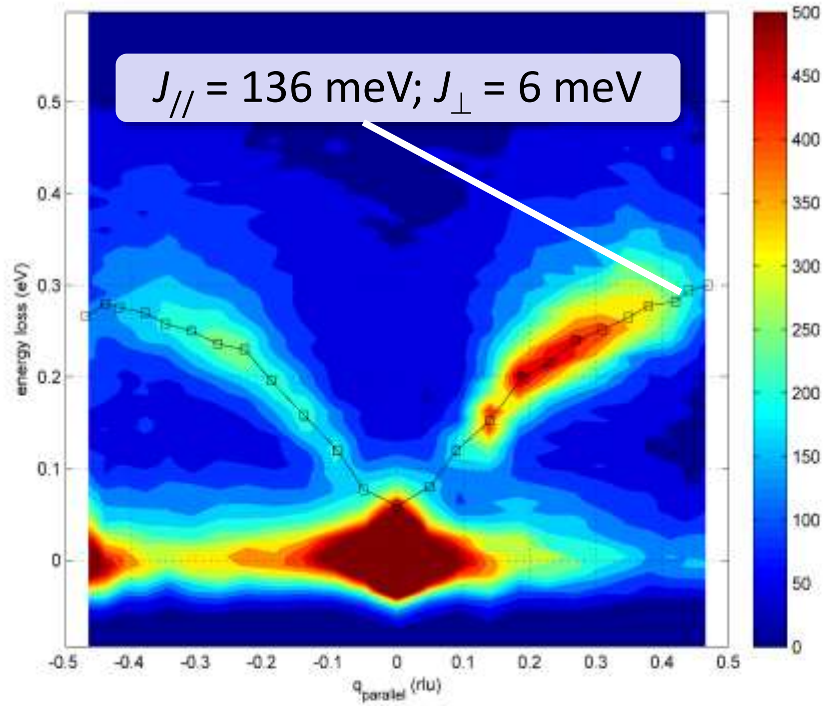
AF $NdBa_2Cu_3O_{6+\delta}$: magnon optical branch



July 2015

BW = 50 meV

π pol. – spin-flip

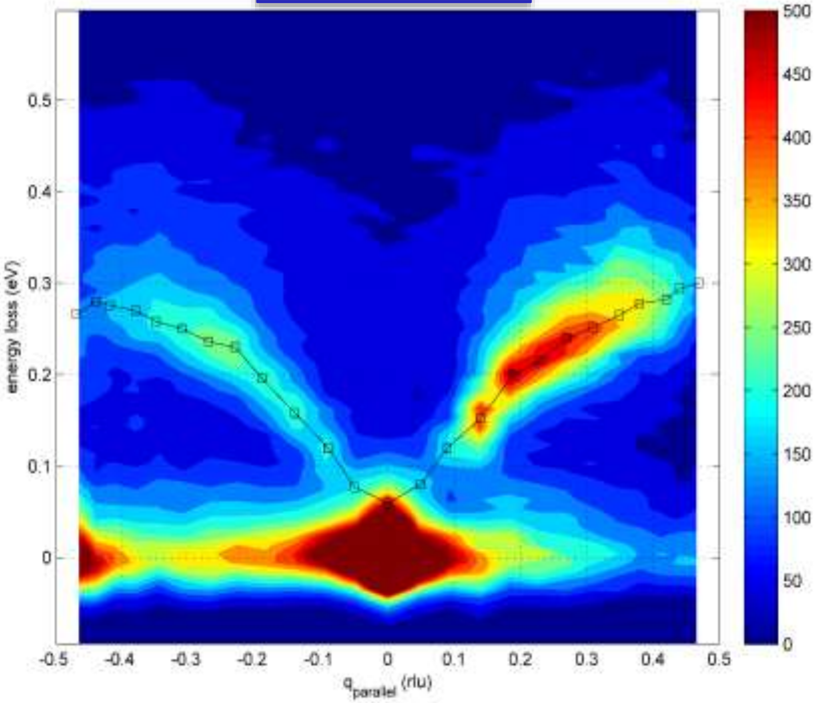


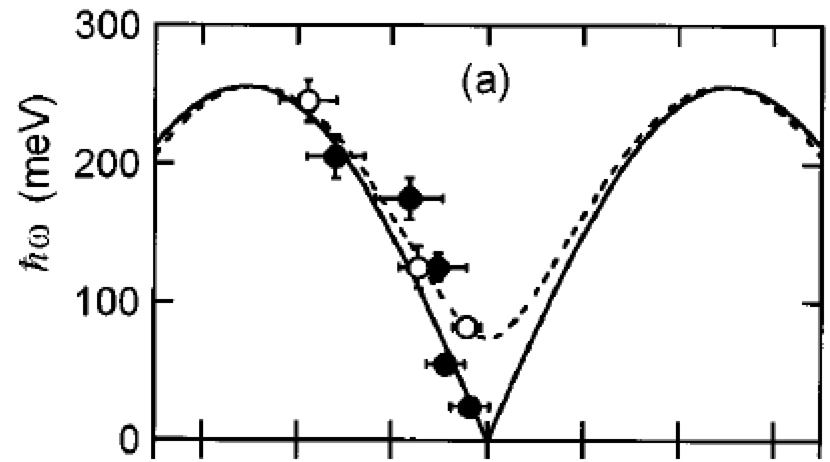
Y.Y. Peng, G. Dellea, M. Minola, G.M. De Luca, M. Salluzzo, M. Le Tacon, B. Keimer, L. Braicovich, N.B. Brookes, and G. Ghiringhelli, unpublished

Comparing RIXS with INS

YBCO AF
INS

NBCO AF
RIXS





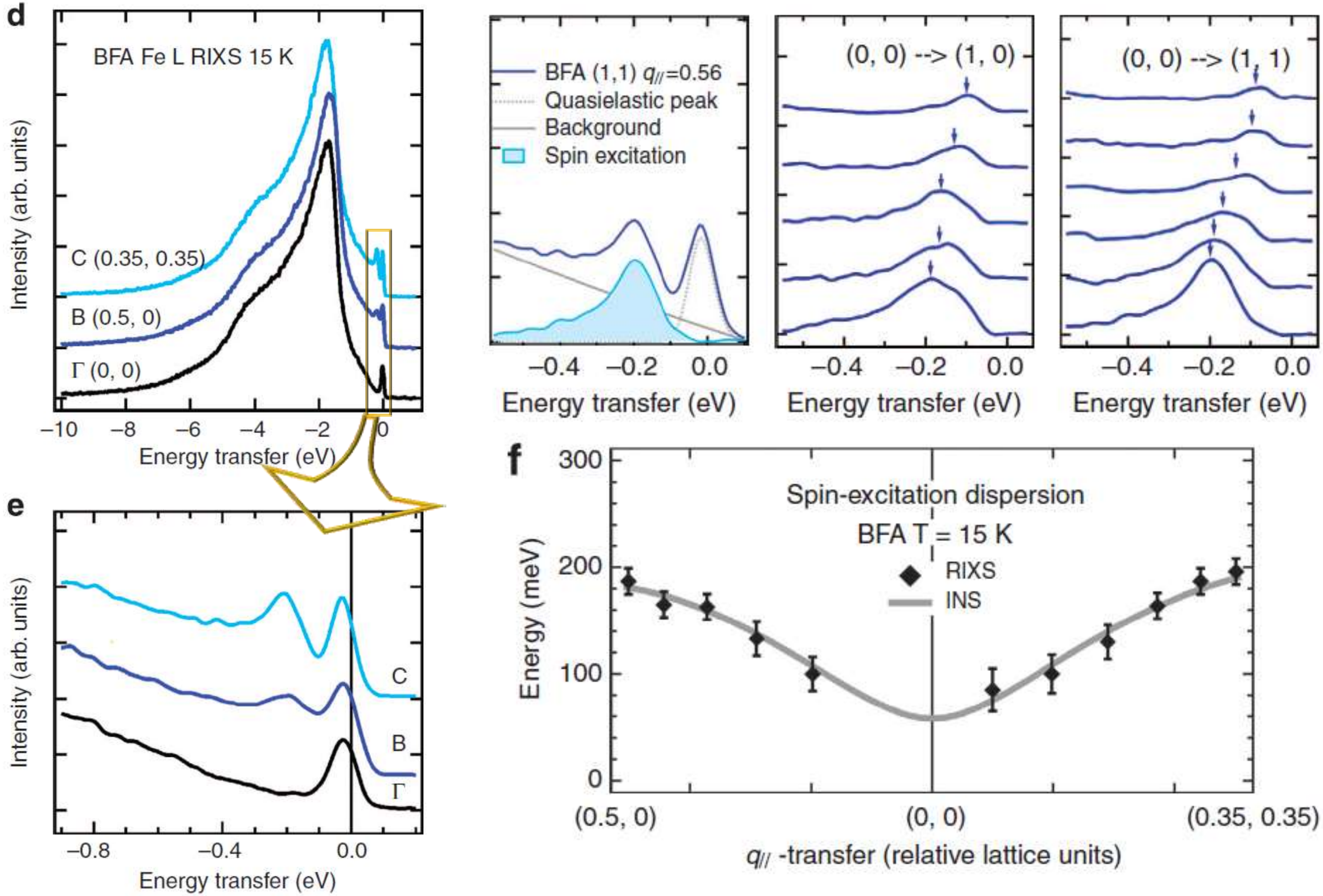
"$YBa_2Cu_3O_{6.15}$ with mass 96 g. […]the resolution in energy was 2 meV and in Q was 0.05 Å^{-1}."

100 nm thick film $NdBa_2CuO_{6.2}$.
BW 55meV, ΔQ=0.02 Ang^{-1}.

S. Hayden et al PRB 54 R6905 (1996)

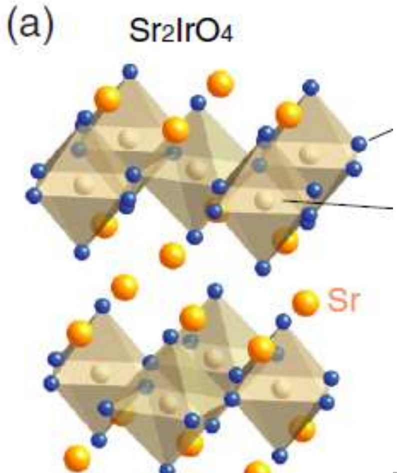
YY Peng, GG et al, unpublished

Magnons at Fe L_3 edge in $BaFe_2As_2$



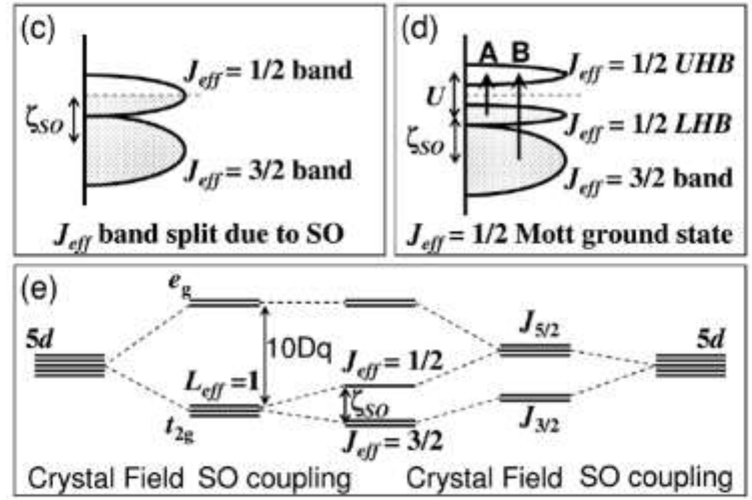
Ke-Jin Zhou, Yao-Bo Huang, Claude Monney, Xi Dai, Vladimir N. Strocov, Nan-Lin Wang, Zhi-Guo Chen, Chenglin Zhang, Pengcheng Dai, Luc Patthey, Jeroen van den Brink, Hong Ding & Thorsten Schmitt, Nature Comm. **4**, 1470 (2013)

Magnetic and orbital excitations in Sr_2IrO_4



Ir^{4+}

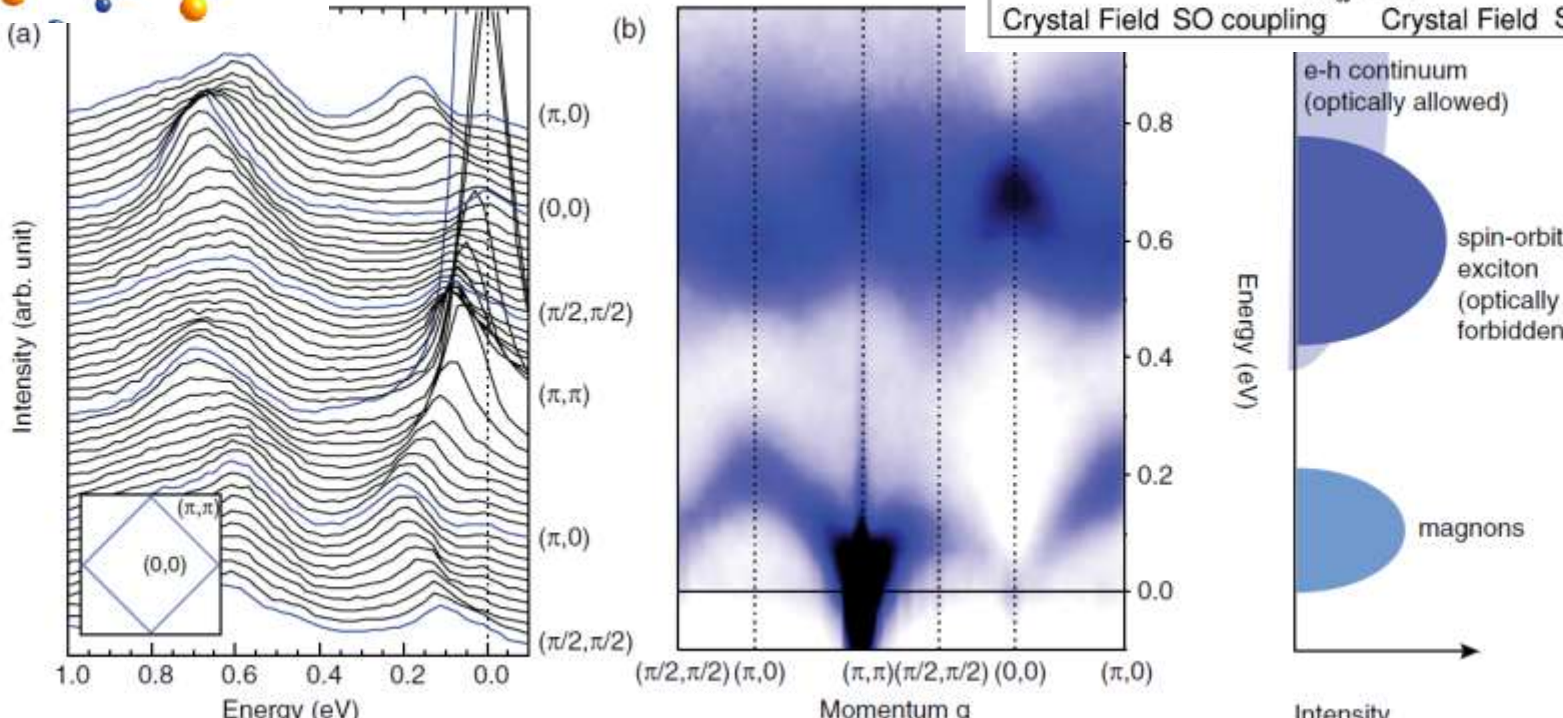
$5d^5$ ⇨



Strong spin-orbit in the 5*d*

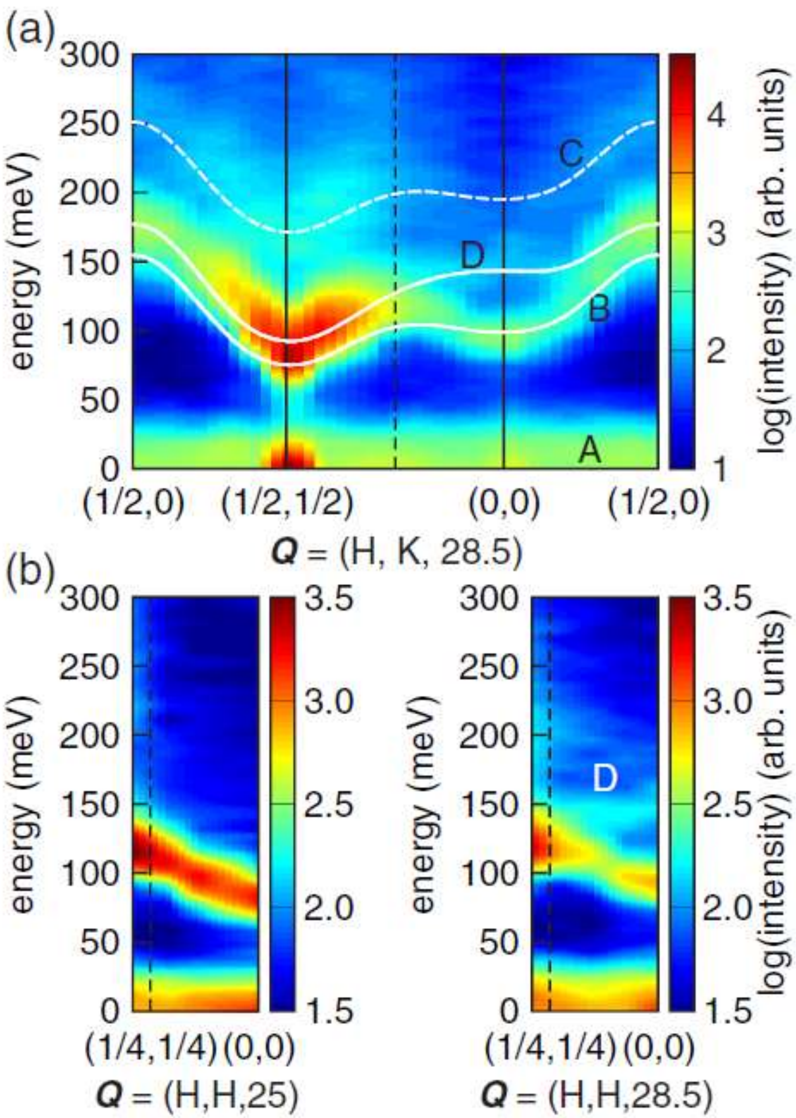
$(\zeta_{SO} \sim 0.4$ eV

L_3 at 11.2 keV

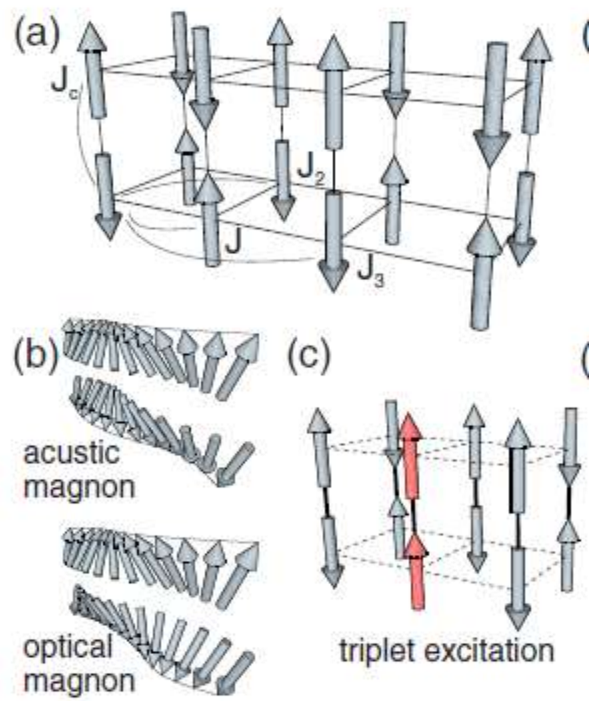


Jungho Kim, D. Casa, M. H. Upton, T. Gog, Young-June Kim, J. F. Mitchell, M. van Veenendaal, M. Daghofer, J. van den Brink, G. Khaliullin, and B. J. Kim, Phys. Rev. Lett. **108**, 177003 (2012)

Magnetic excitations in bilayer iridates



$Sr_3Ir_2O_7$

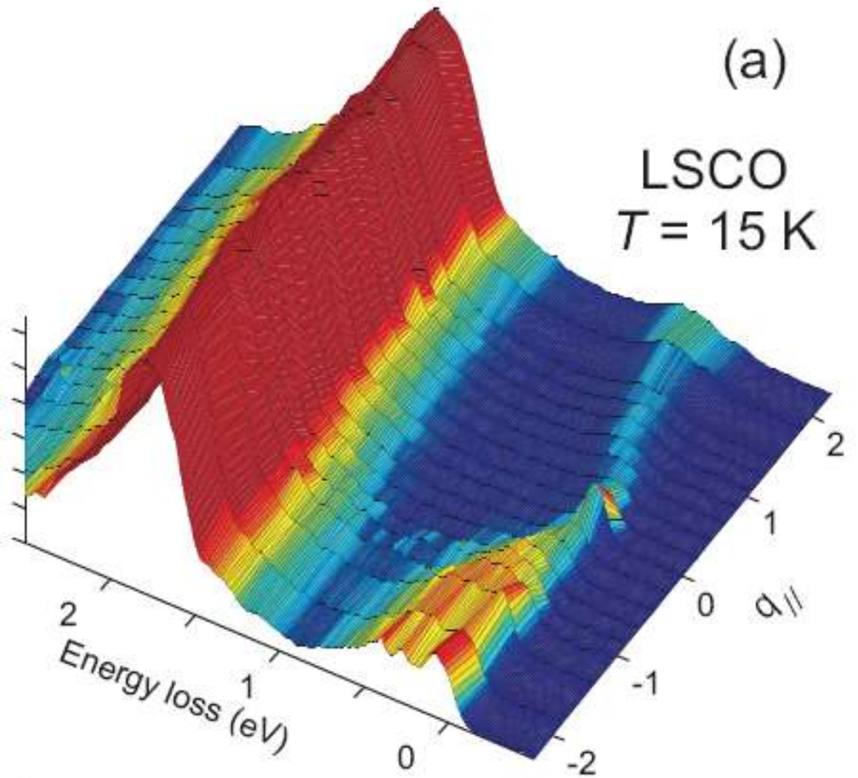


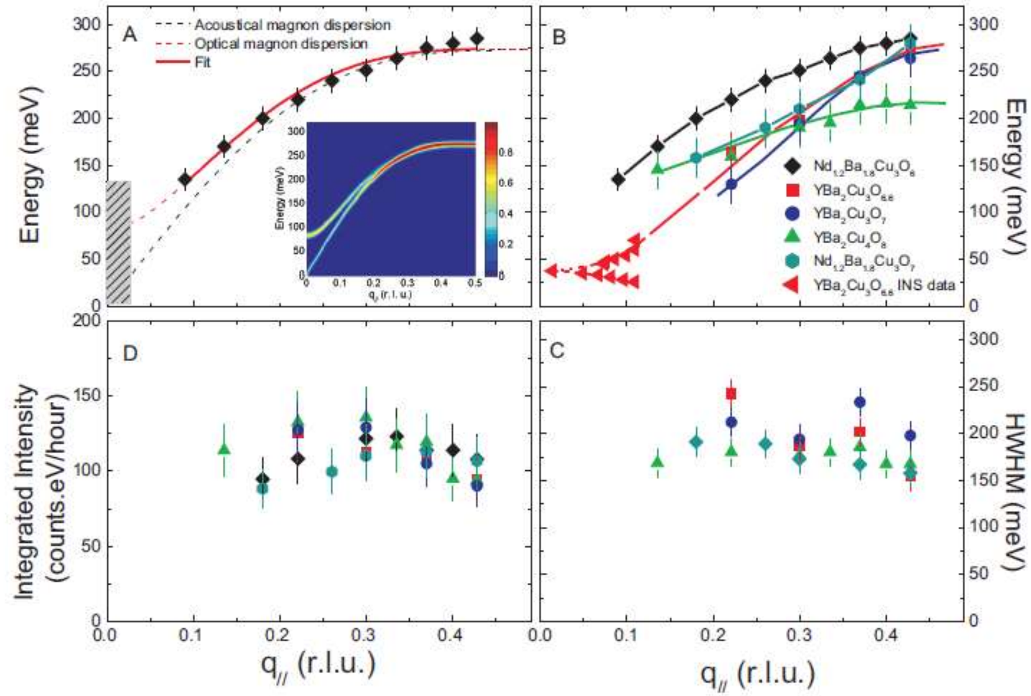
$J_c<J$

$J_c>J$

M. Moretti Sala, et al, PRB **92**, 024405 (2015)

Superconductors: LSCO, YBCO and NdBCO





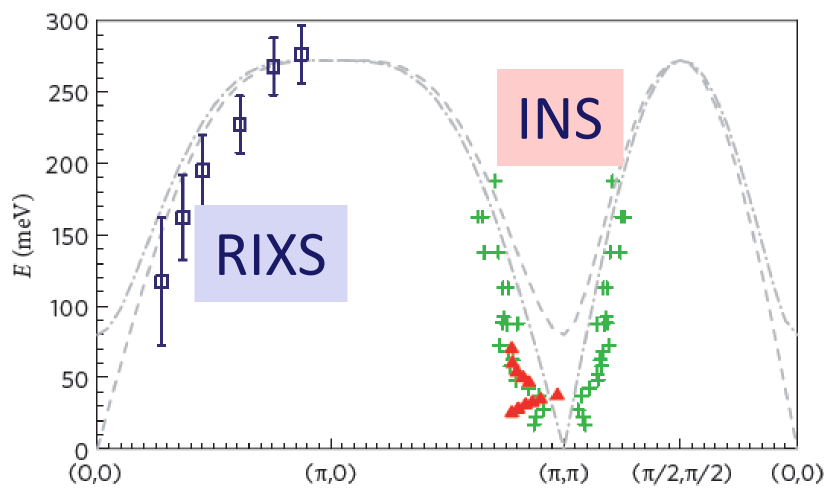
LSCO & NdBCO: 100 nm films on STO. YBCO: detwinned single crystals

Dispersing magnetic excitations are almost as strong in SC as in the AF parent compounds: they can be involved in Cooper pairing

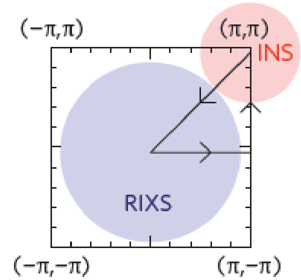
L. Braicovich, J. van den Brink, V. Bisogni, M. Moretti Sala, L. Ament, N.B. Brookes, G.M. de Luca, M. Salluzzo, T. Schmitt, and G. Ghiringhelli PRL **104** 077002 (2010)

M. Le Tacon, G. Ghiringhelli, J. Chaloupka, M. Moretti Sala, V. Hinkov, M.W. Haverkort, M. Minola, M. Bakr, K. J. Zhou, S. Blanco-Canosa, C. Monney, Y. T. Song, G. L. Sun, C. T. Lin, G. M. De Luca, M. Salluzzo, G. Khaliullin, T. Schmitt, L. Braicovich and B. Keimer, Nat. Phys. **7**, 725 (2011)

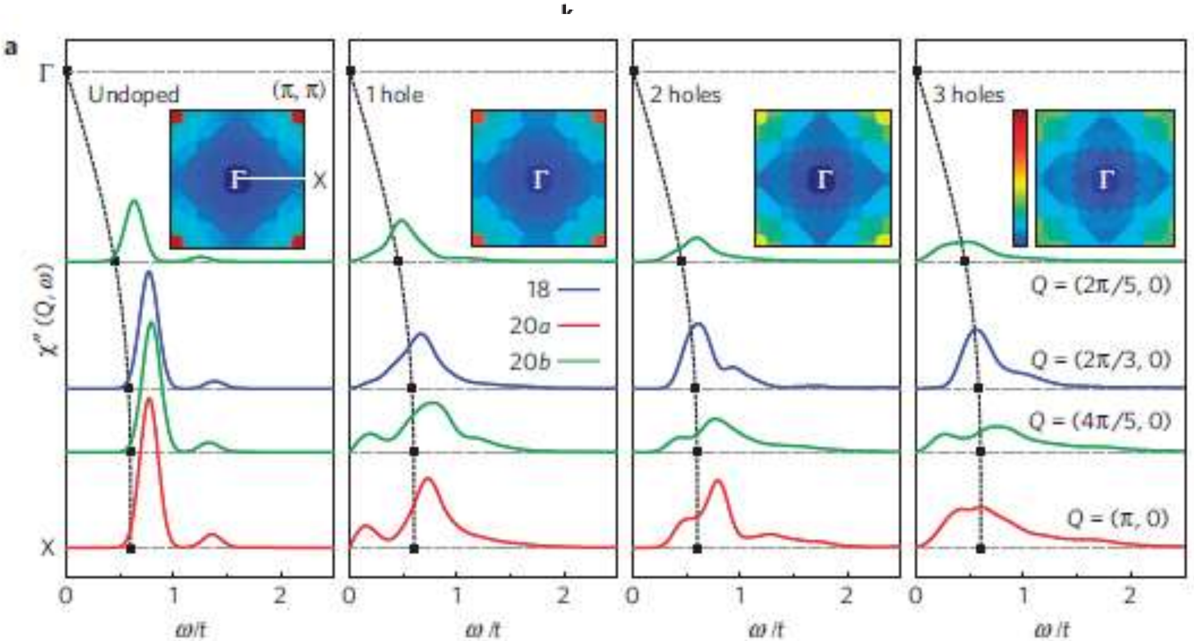
YBCO: doping dependence of χ''

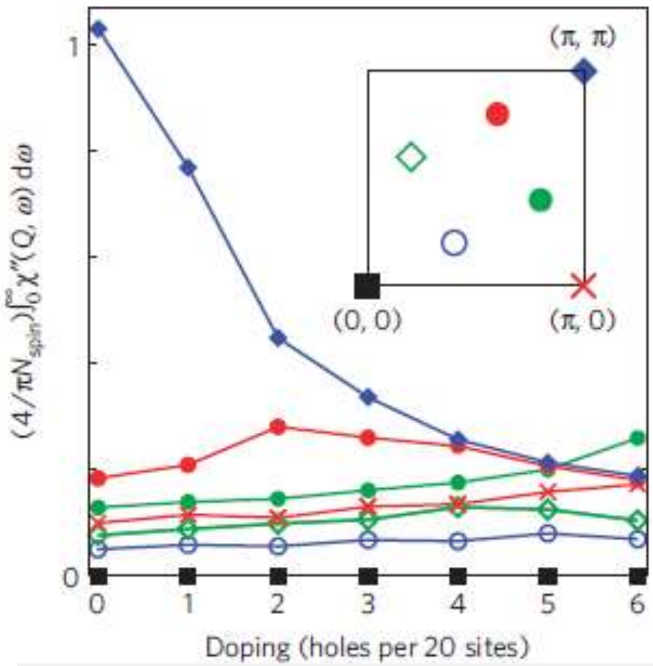


Matthias Vojta, *News and Views*, Nature Physics **7,** 674 (2011)



$YBa_2Cu_3O_{6.6}$

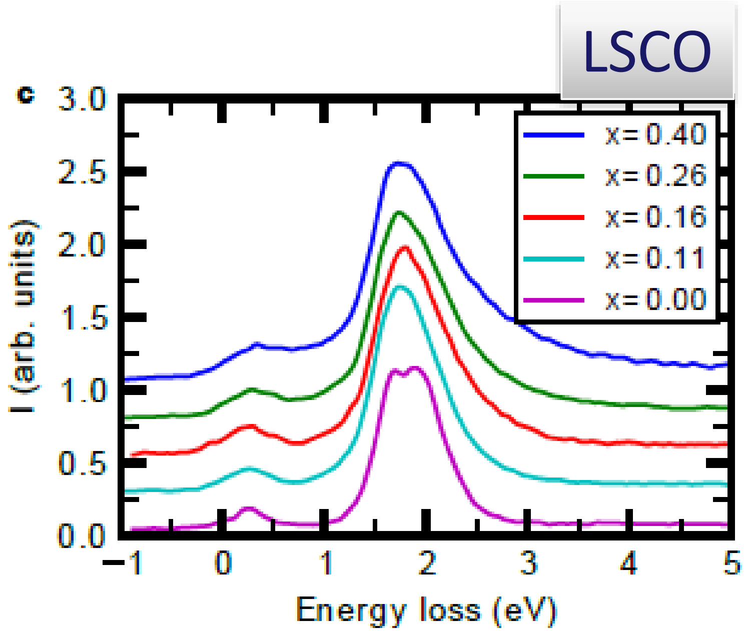




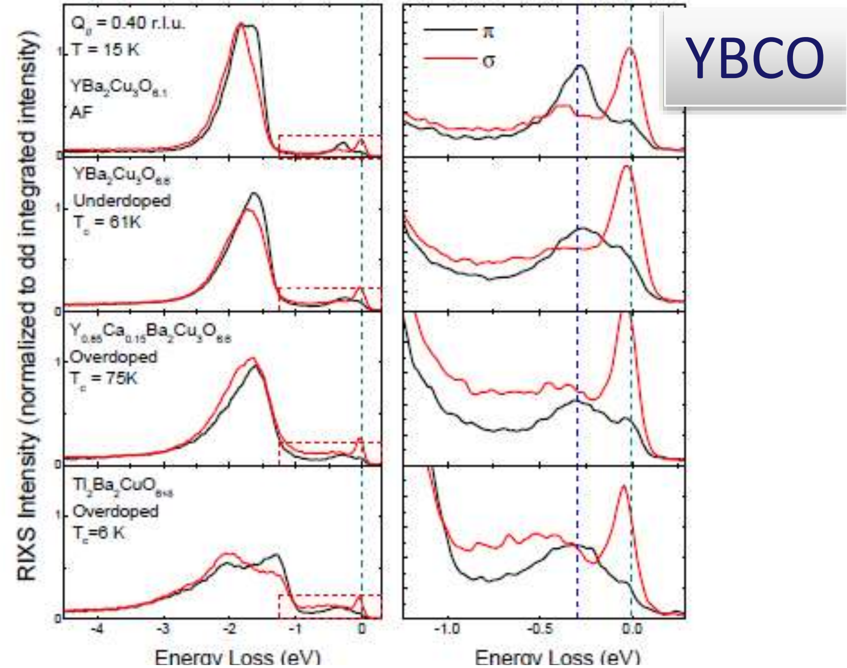
Imaginary part of the spin susceptibility χ''*(Q;w) resulting from exact diagonalization of the t–J model with J/t=0.3 on small cluster. (G. Khaliullin)*

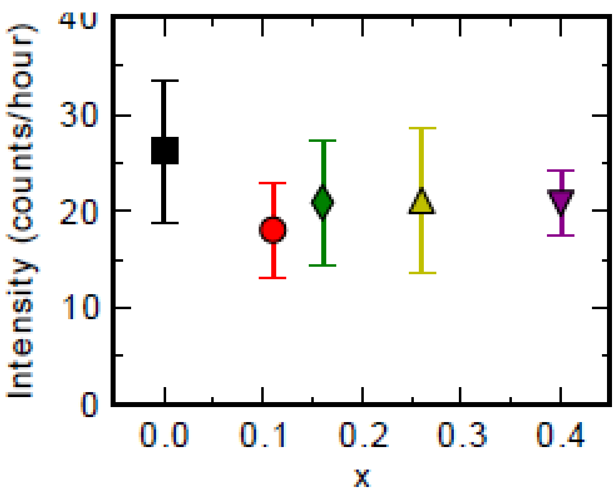
Energy-integrated $\chi''$of the 20-site cluster (normalized) for 7 accessible non-equivalent Q vectors. *(G. Khaliullin)*

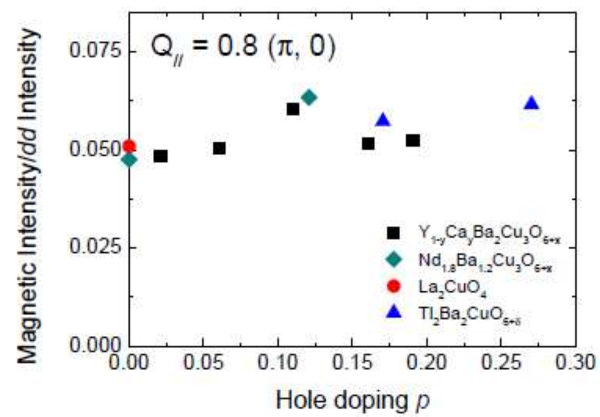
Peristent magnetic excits in overdoped cuprates



π pol



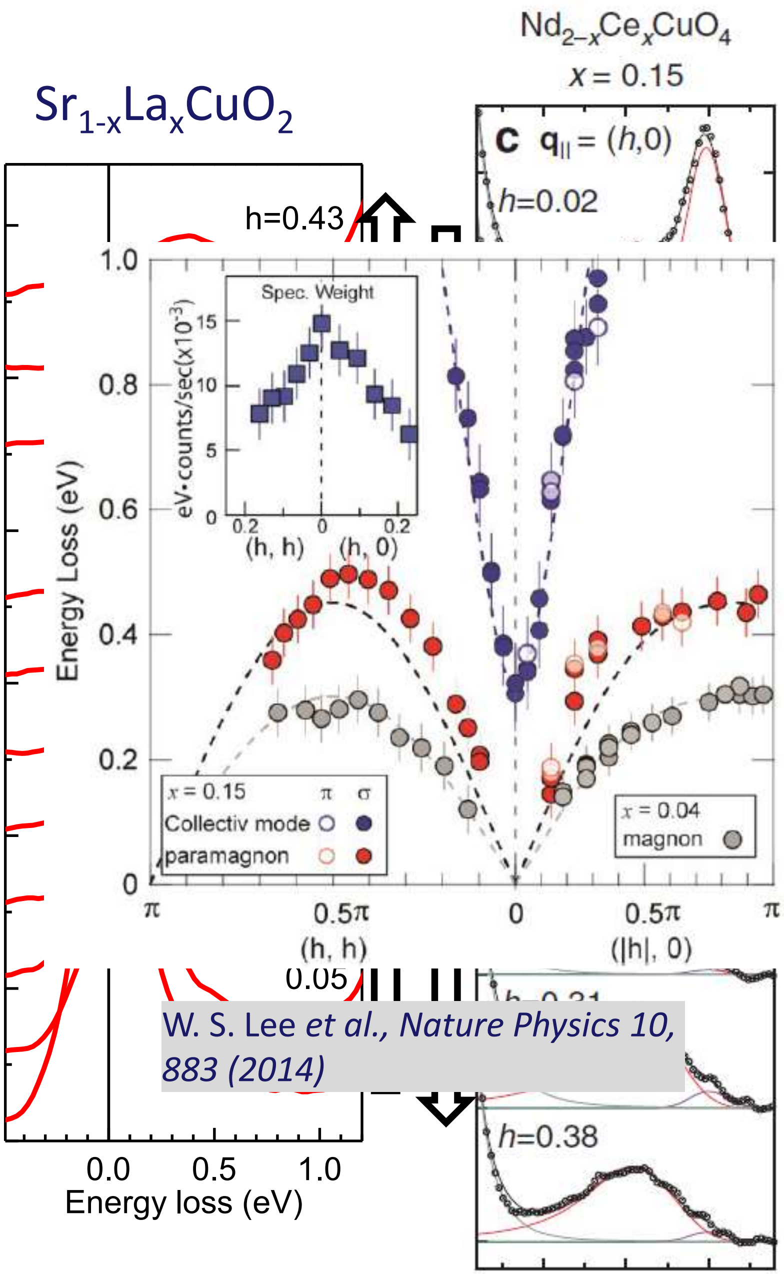




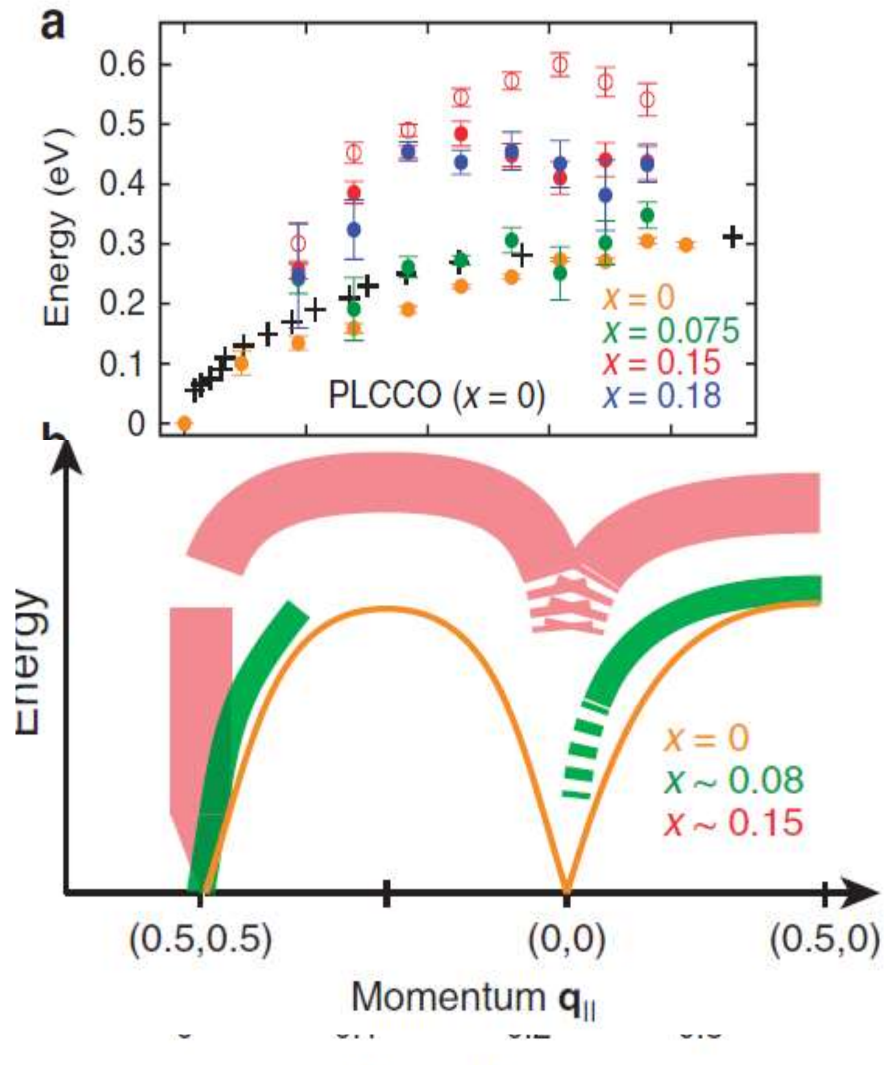
M. P. M. Dean,, . G. Dellea, R. S. Springell, F. Yakhou-Harris, K. Kummer, N. B. Brookes, X. Liu, Y.-J. Sun, J. Strle, T. Schmitt, L. Braicovich, G. Ghiringhelli, I. Bozovic, and J. P. Hill, Nat. Mater. **12**, 1019 (2013)

M. Le Tacon, M. Minola, D. C. Peets, M. Moretti Sala, S. Blanco-Canosa, V. Hinkov, R. Liang, D. A. Bonn, W. N. Hardy, C. T. Lin, T. Schmitt, L. Braicovich, G. Ghiringhelli, and B. Keimer, Phys. Rev. B 88, 020501 (2013)

Spin excitations in e-doped SC

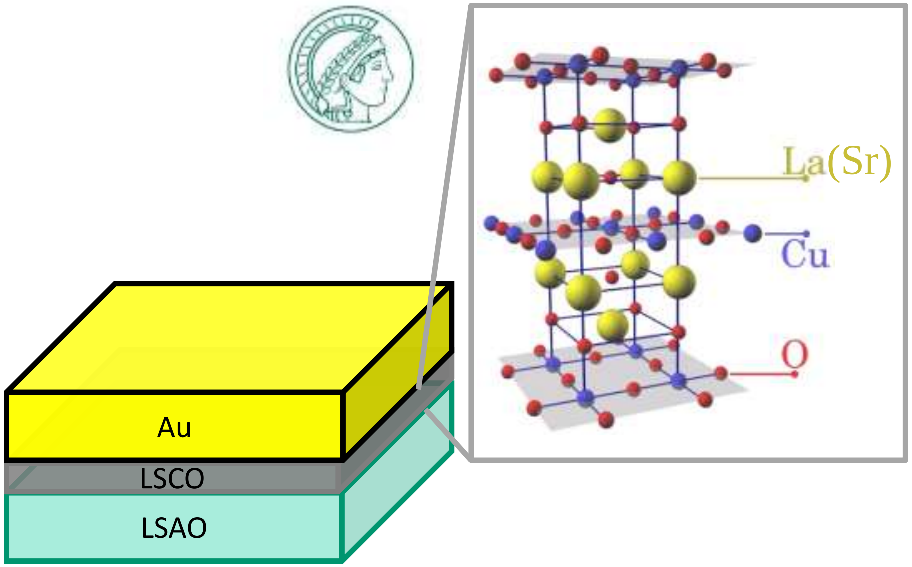


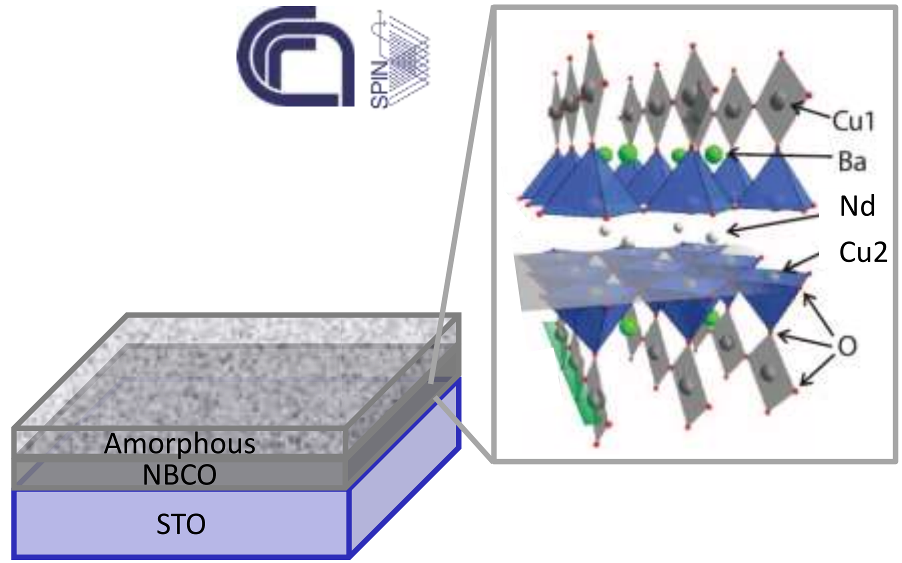
W. S. Lee *et al., Nature Physics 10, 883 (2014)*



K. Ishii, M. Fujita, T. Sasaki, M. Minola, G. Dellea, C. Mazzoli, K. Kummer, G. Ghiringhelli, L. Braicovich, T. Tohyama, K. Tsutsumi, K. Sato, R. Kajimoto, K. Ikeuchi, K. Yamada, M. Yoshida, M. Kurooka & J. Mizuki, Nat. Comm. **5**, 3714 (2014)

Individual cuprate layers





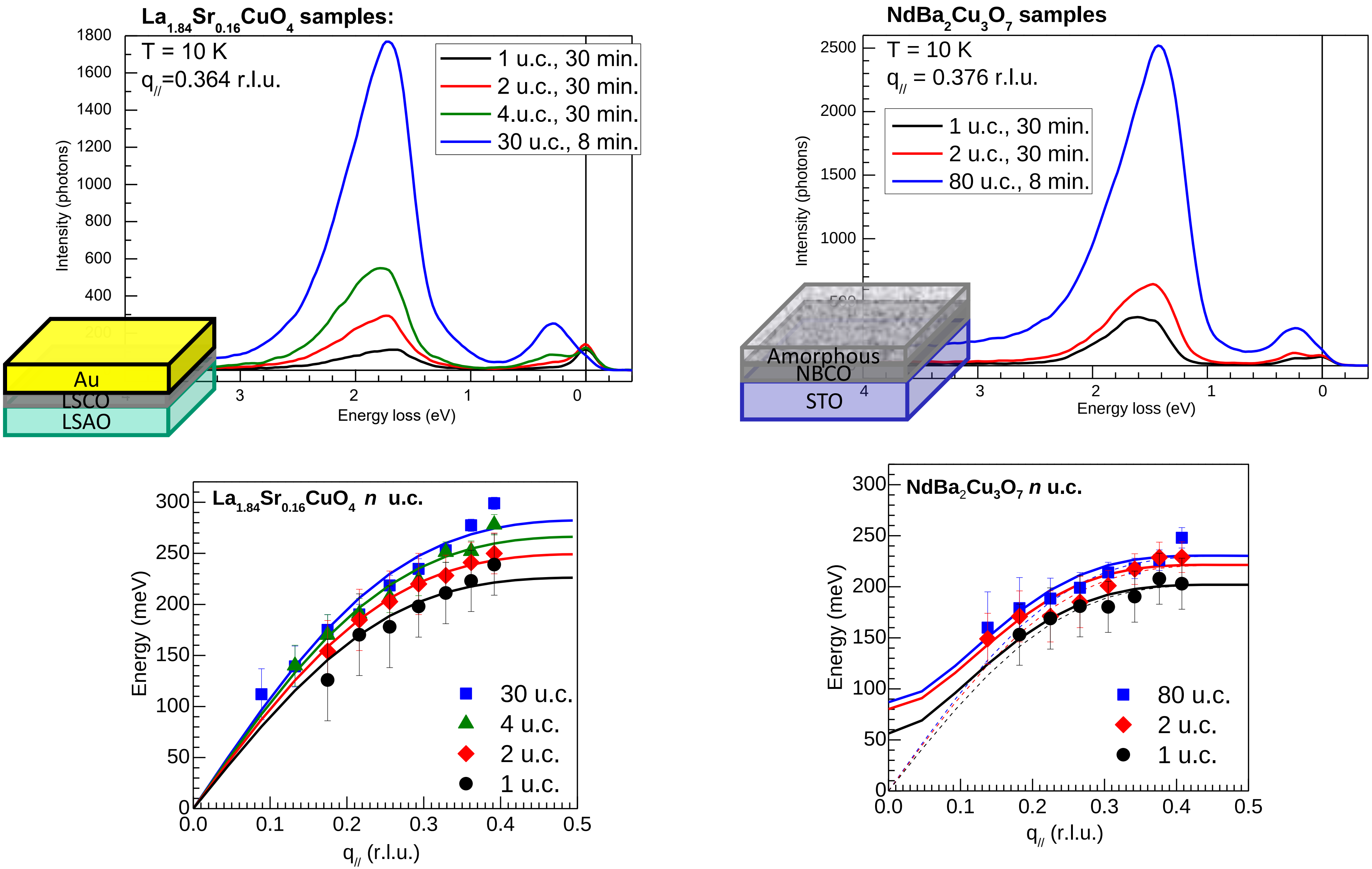
- **$La_{2-x}Sr_xCuO_4$ (LSCO) opt. doped *x*=0.16**
- **Au capping** (10÷15 nm thick)
  - 1 u.c. non-SC
  - 2 u.c. $T_c \approx 20$ K
  - 4 u.c. $T_c \approx 25$ K
  - 30 u.c. $T_c \approx 38$ K

Grown by G. Logvenov, G. Cristiani, F. Baiutti

- **$NdBa_2Cu_3O_7$ (NBCO) optimally doped**
- **Amorphous NBCO capping** (equivalent thickness of 2 u.c.)
  - 1 u.c. non-SC
  - 2 u.c. $T_c \approx 93$ K
  - 80 u.c. $T_c \approx 93$ K

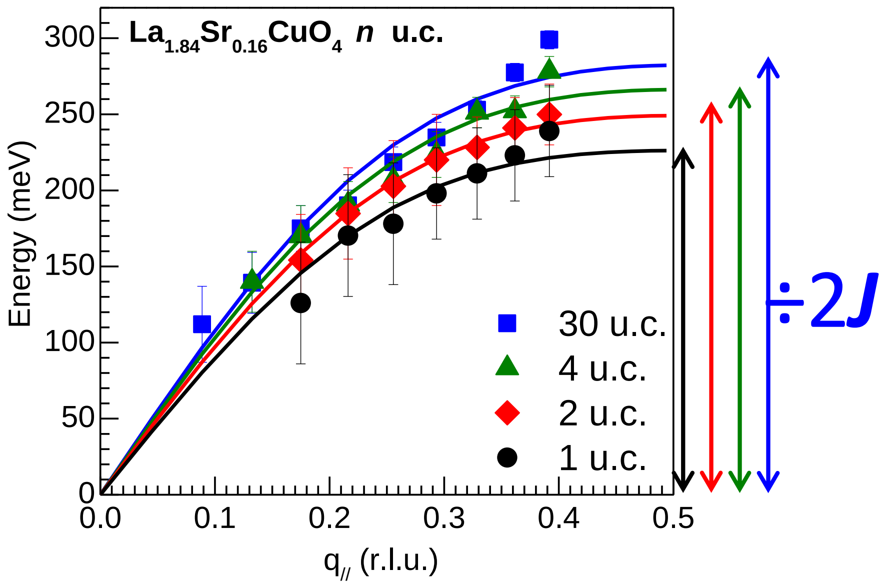
Grown by M. Salluzzo

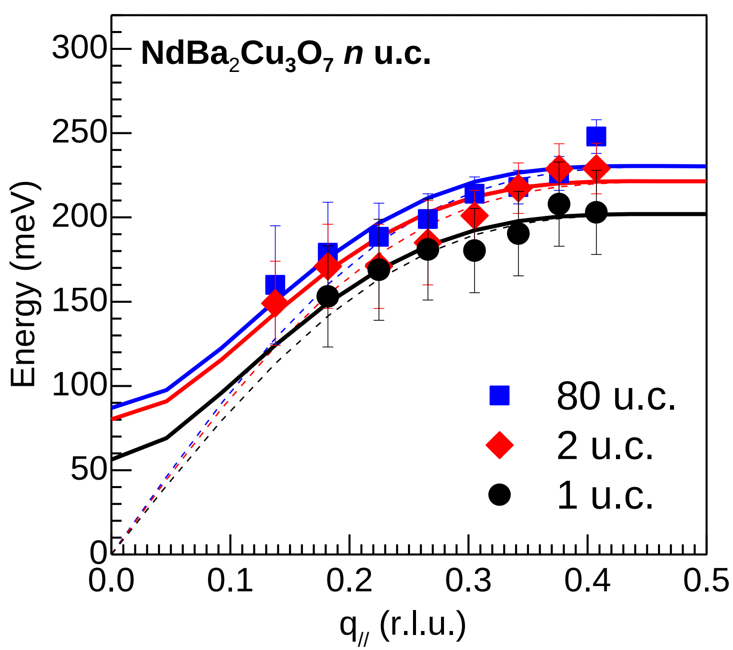
Exceptional sensitivity of RIXS

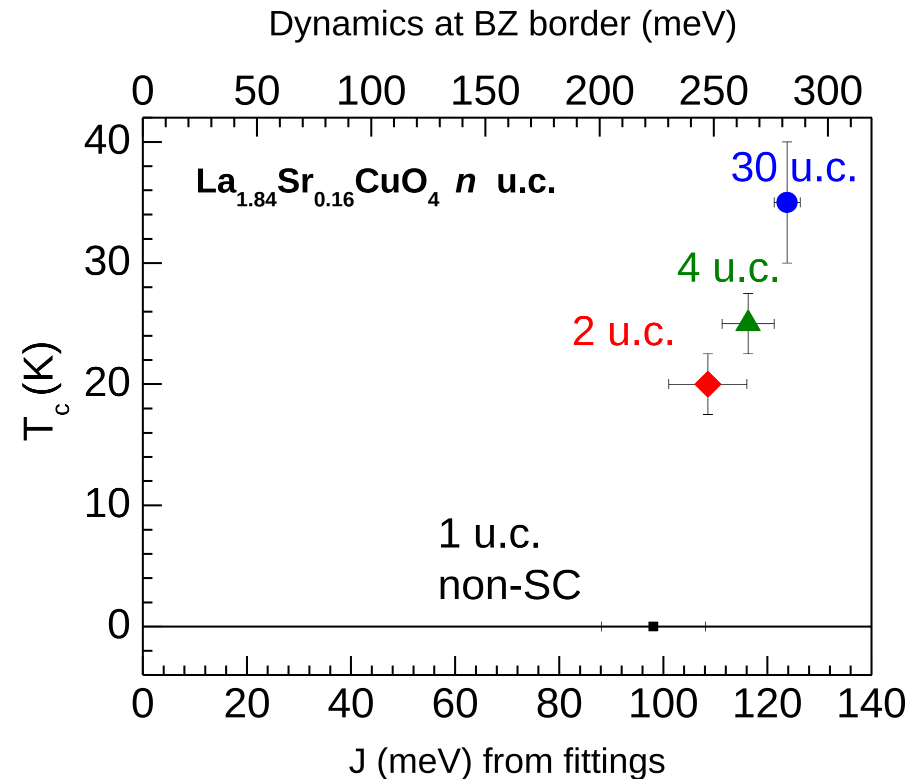


Experiment at ADRESS beam line of SLS: **M. Minola**, G. Dellea, M. Salluzzo, V. Bisogni, T. Schmitt, M. Le Tacon, G. Logvenov, G. Cristiani, F. Baiutti, B. Keimer, L. Braicovich, G. Ghiringhelli, unpublished

Small effect of thickness on *J*



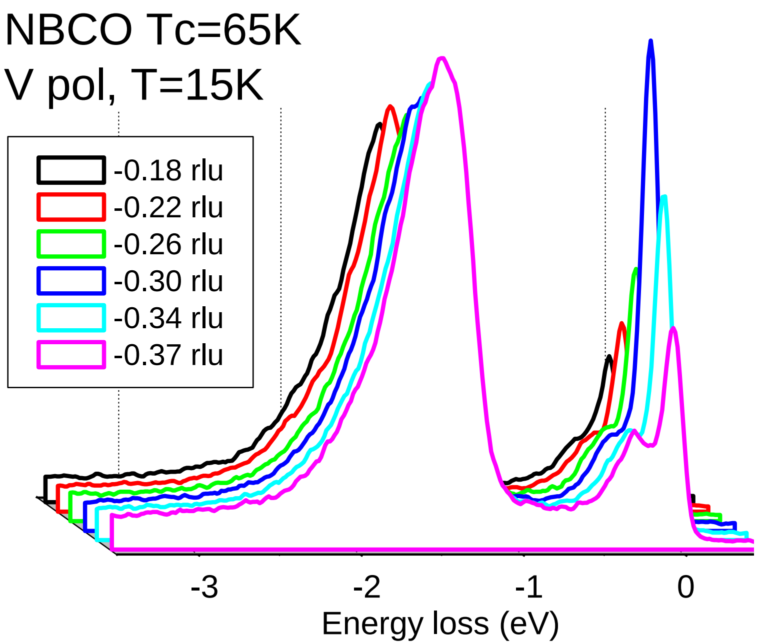


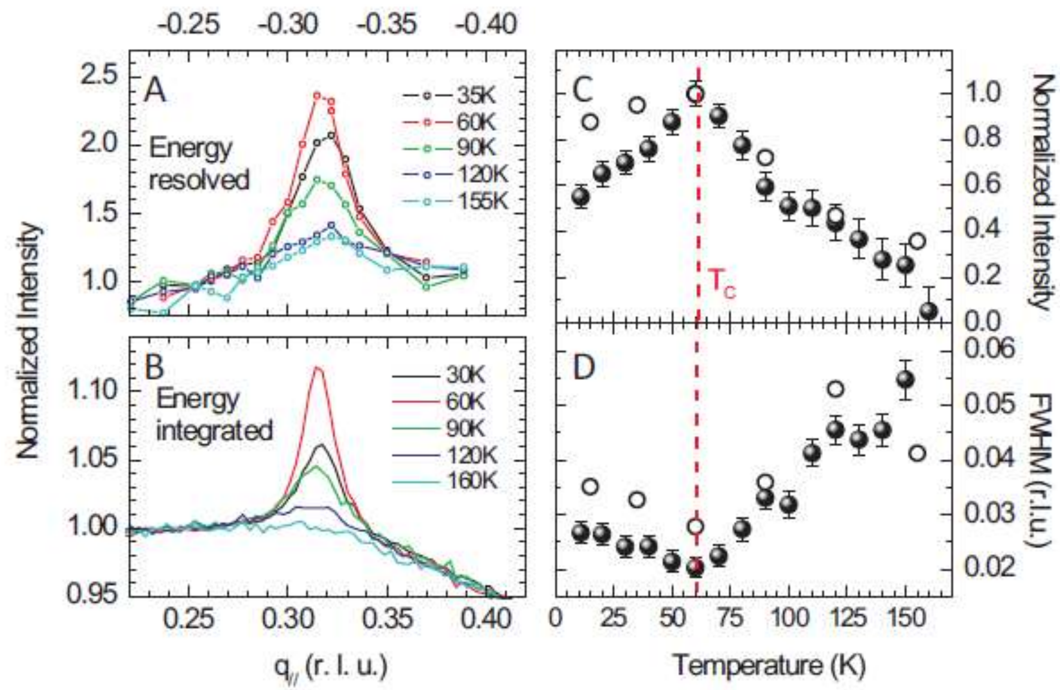


Linear spin-wave theory fittings

J decreases at lower film thickness, but only by 20% from bulk to 1 uc

RIXS revealed Charge Order in HTcS





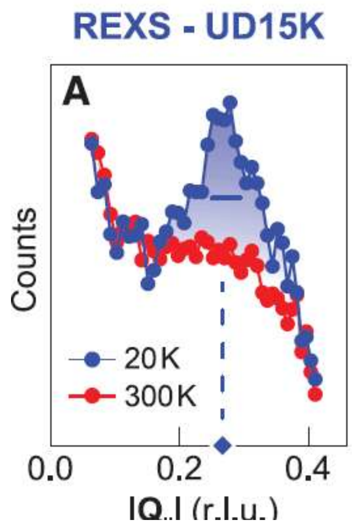
Max intensity at Tc: CO compete with SC

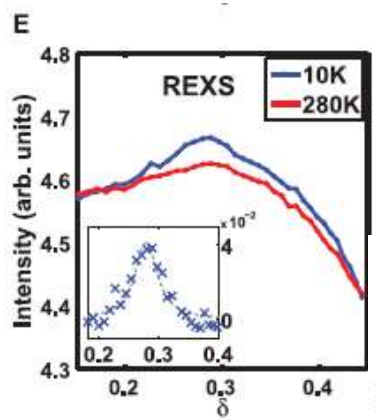
G. Ghiringhelli, M. Le Tacon, M. Minola, S. Blanco-Canosa, C. Mazzoli, N.B. Brookes, G.M. De Luca, A. Frano, D. G. Hawthorn, F. He, T. Loew, M. Moretti Sala, D.C. Peets, M. Salluzzo, E. Schierle, R. Sutarto, G. A. Sawatzky, E. Weschke, B. Keimer, L. Braicovich, Science **337**, 821 (2012)

RXS (at Cu L_3 and O K) in combination with STM, XRD and NMR has demonstrated that CO is ubiquitous in cuprates

UD Bi2201, Bi2212, Hg1201 and OPD Bi2212

Bi2201 and Bi2212 underdoped

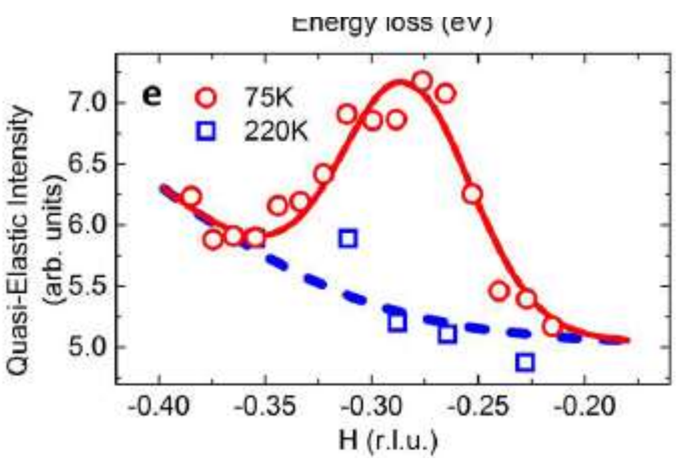




R. Comin et al, Science 343, 390 (2014;

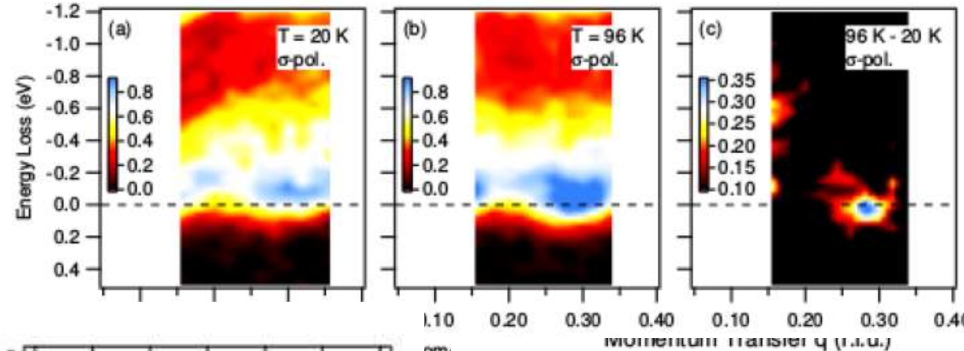
Eduardo H. da Silva Neto et al, Science 343, 393 (2014

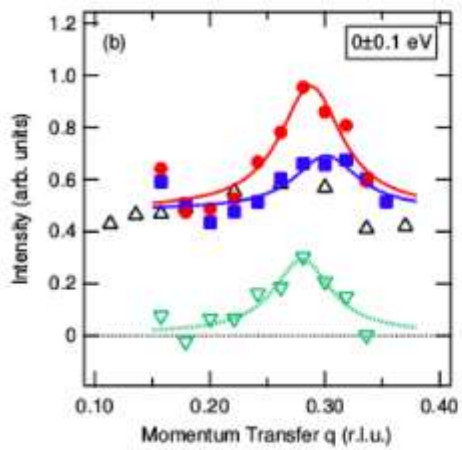
Hg1201 underdoped

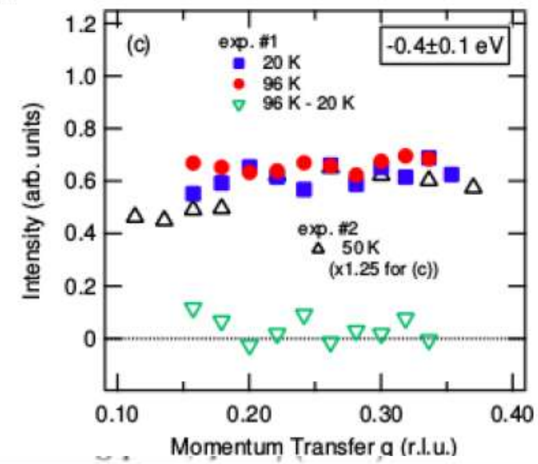


W. Tabis et al, Nat. Comm. 6875 (2014)

Bi2212 optimally doped







Sample	p	T_c	$q_{\parallel}$ (r.l.u.)	ξ (Å)	refs.
Bi2201	0.115	15	0.265	26	[16]
Bi2201	0.130	22	0.257	23	[16]
Bi2201	0.145	30	0.243	21	[16]
Bi2212	0.09	45	0.30	24	[15]
Bi2212	0.160	98	0.28	<24 (at T_c)	this work
YBCO	0.115	61	0.32	∼60 (at T_c)	[8, 10]
LBCO	0.125	2.5	0.236	∼200	[4, 6–8]
LBCO	0.155	30	0.244	∼240 (15 – 25 K)	[4, 6–8]

M. Hashimoto, G. Ghiringhelli et al, PRB 89 220511 (2014)

ENERGY RESOLUTION: progress in the last 20 years

La_2CuO_4
Cu 2*p* → 3*d*
Photon energy ~ 931 eV

Δ*E* ~1.6 eV

K. Ichikawa *et al.*, J. Electron Spectrosc. Relat. Phenom.**78**, 183 (1996).

Δ*E* ~1.2 eV

L. C. Duda *et al.*, J. Electron Spectrosc. Relat. Phenom. **110–111**, 275 (2000).

Δ*E* ~0.8 eV

AXES @ ID08, 2003. G. Ghiringhelli *et al.*, Phys Rev Lett. **92**, 117406 (2004).

Δ*E* ~0.45 eV

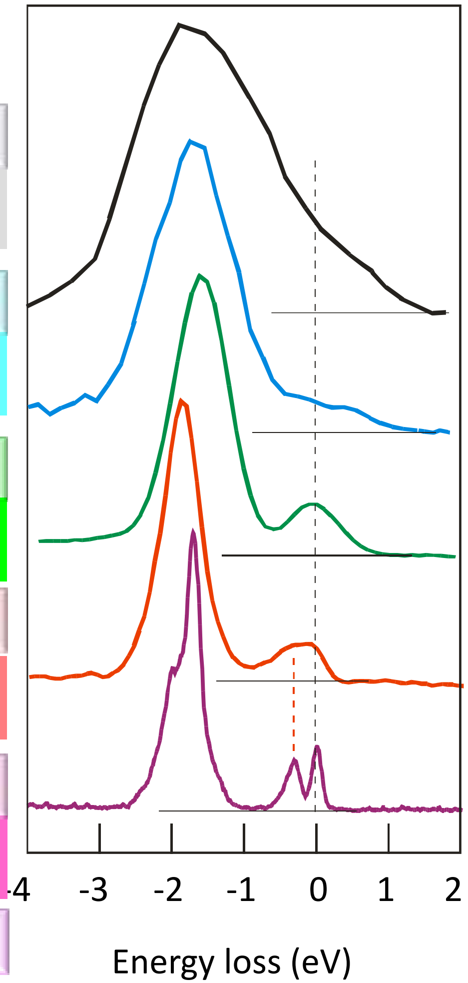
AXES @ ID08, 2007. L. Braicovich *et al.*, arXiv:0807:1140v1, (2008).

Δ*E* ~0.13 eV

SAXES @ SLS, 2008. G. Ghiringhelli, L. Braicovich, T. Schmit *et al.*, unpublished

Δ*E* ~0.050 eV

Δ*E* ~0.030 eV



2000 Uppsala

2003 **2007** ESRF + AXES

2008 SLS + SAXES

2015 **2016** ESRF + ERIXS

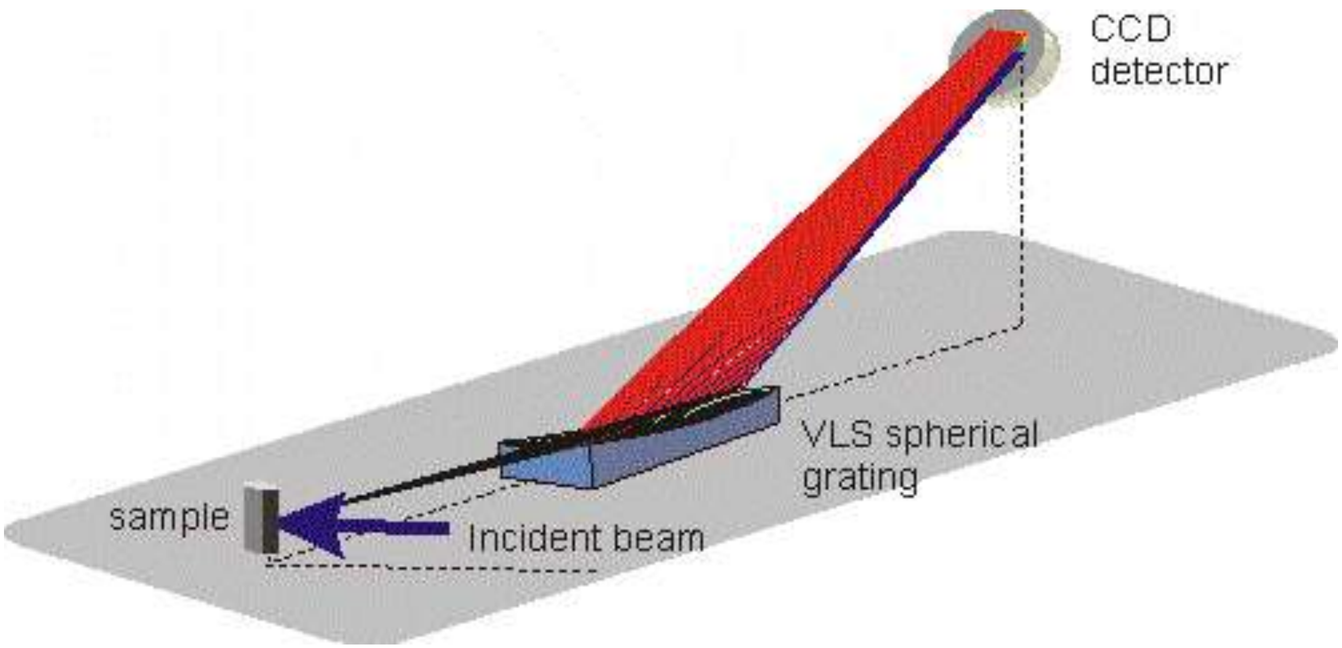
Combined resolving power has increased by a factor 30

Soft x-ray RIXS instrumentation

High resolution mono, small x-ray spot on the sample

Grating spectrometer: optimized efficiency, high resolution

The main limiting factor is INTENSITY!!!!



From AXES (ESRF, ID08) to SAXES (SLS, ADRESS)

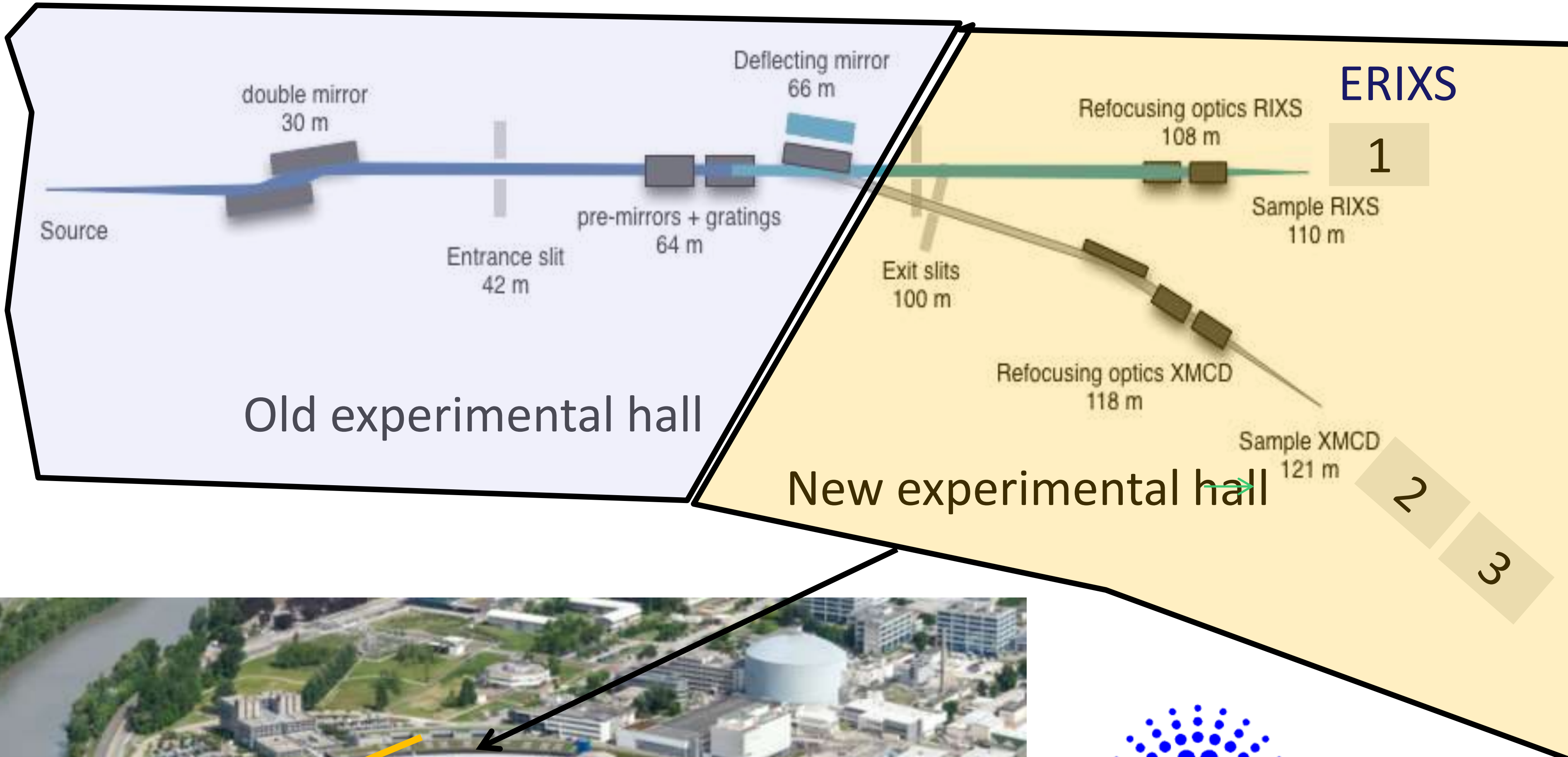
Since 1994: AXES at beam line ID08 of the ESRF
L = 2.2 m
Design: ***E*/Δ*E* = 2,000 at Cu L_3 (930 eV)**
2010: ***E*/Δ*E* = 5,000 at Cu L_3**

Since 2007: SAXES at beam line ADRESS of the SLS
L = 5.0 m
Design: ***E*/Δ*E* = 12,000 at Cu L_3**
2011: ***E*/Δ*E* = 11,000 at Cu L_3**

C. Dallera *et al*. J. Synchrotron Radiat. **3**, 231 (1996)
G. Ghiringhelli *et al.*, Rev. Sci. Instrum. **69**, 1610 (1998)
M. Dinardo *et al.*, Nucl, Instrum. Meth A **570**, 176 (2007)

G. Ghiringhelli, et al Rev. Sci. Instrum. **77**, 113108 (2006)
V. Strocov, T. Schmitt, L. Patthey et al, J. Synch. Rad., 17, 631 (2010).

New ID32 at the ESRF



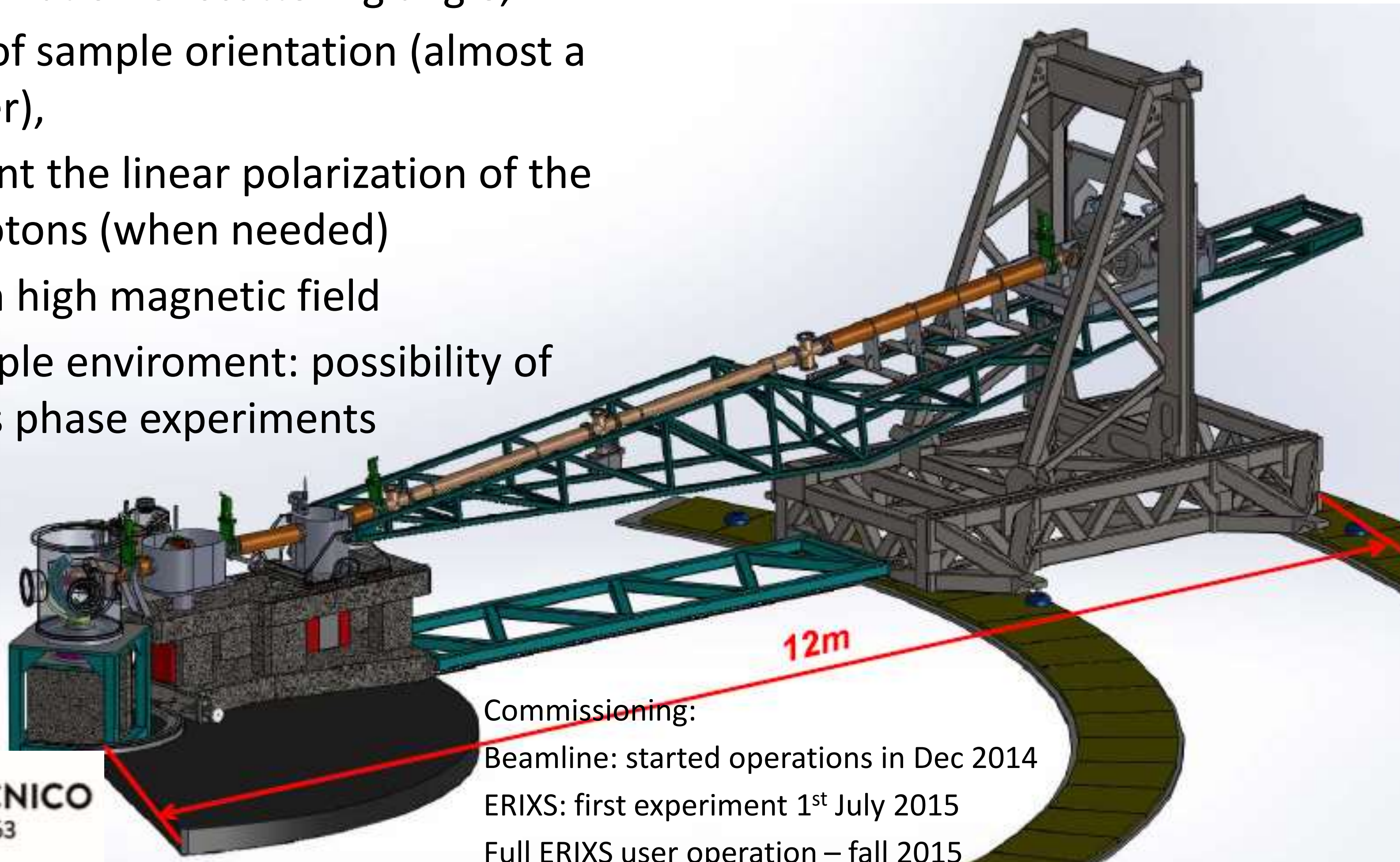
ESRF

ЄRIXS spectrometer at the new ID32

ESRF Upgrade program,
N.B. Brookes, F. Yakhou,
GG et al

FEATURES:

- $E/\Delta E > 20{,}000$ below 1000 eV from day one (50 meV at Cu L_3) and $E/\Delta E > 30{,}000$ ultimate
- continous variation of scattering angle,
- full control of sample orientation (almost a diffractometer),
- measurement the linear polarization of the scattered photons (when needed)
- optionally in high magnetic field
- flexible sample enviroment: possibility of liquid and gas phase experiments



Commissioning:
Beamline: started operations in Dec 2014
ERIXS: first experiment 1st July 2015
Full ERIXS user operation – fall 2015

ESRF

POLITECNICO MILANO 1863

ERIXS@ID32, ESRF, 27/04/2014

ERIXS, 27/04/2014

ERIXS Spectrometer

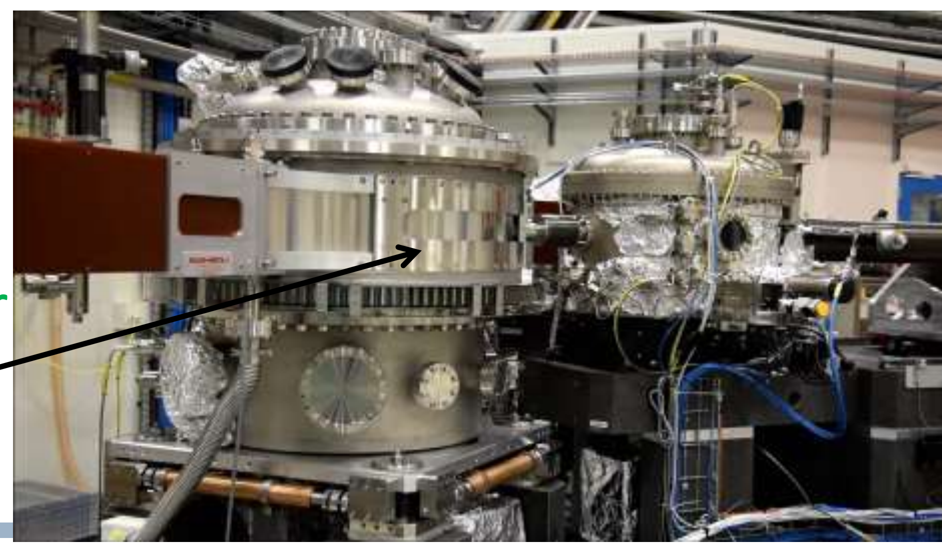
Continuously rotating scattering arm

Speed x8.6

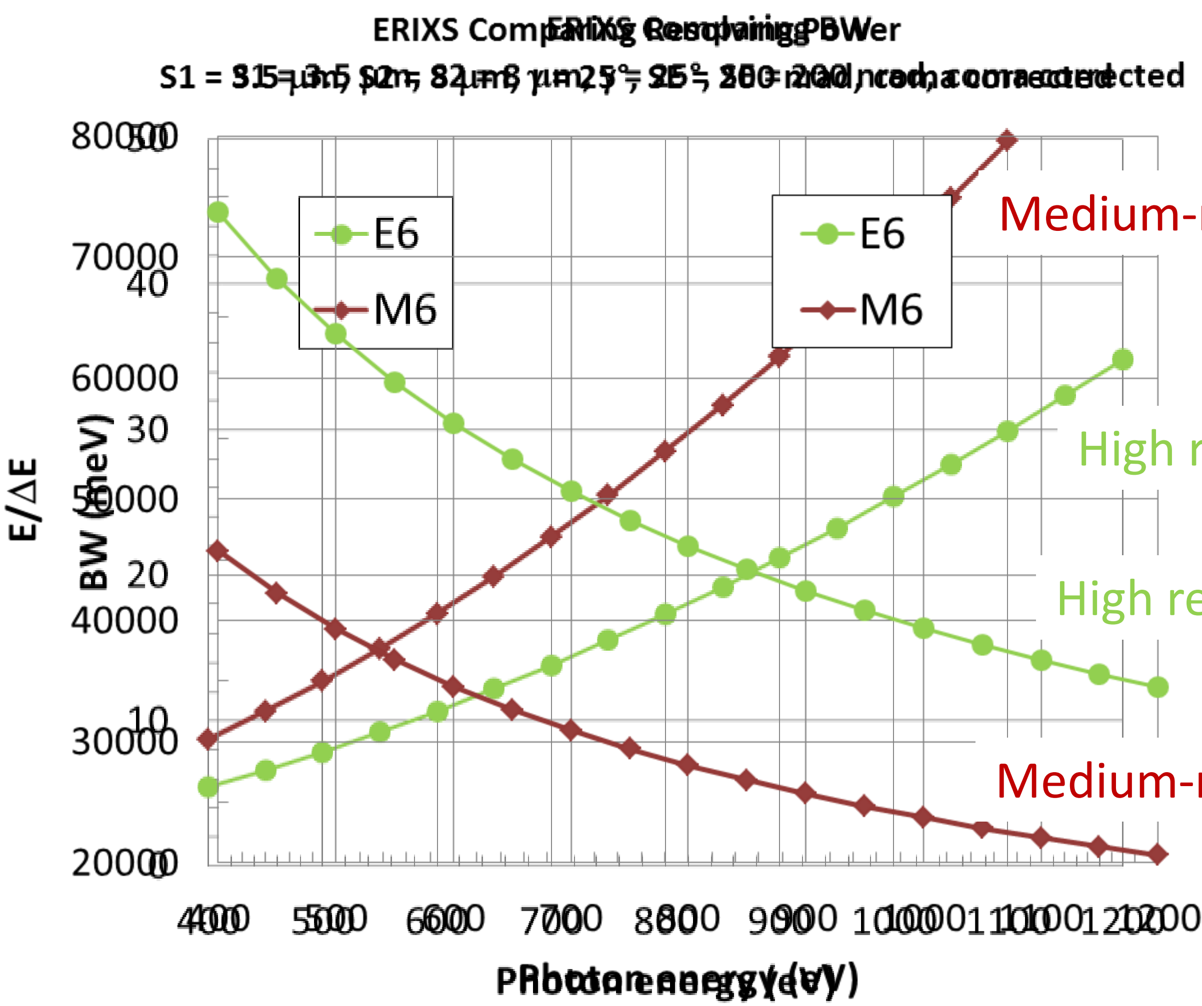
In-vacuum Huber diffractometer

Sample chamber CINEL – Italy

ESRF



ERIXS: Expected resolving power



Medium-resolution grating: 1400 mm^{-1}

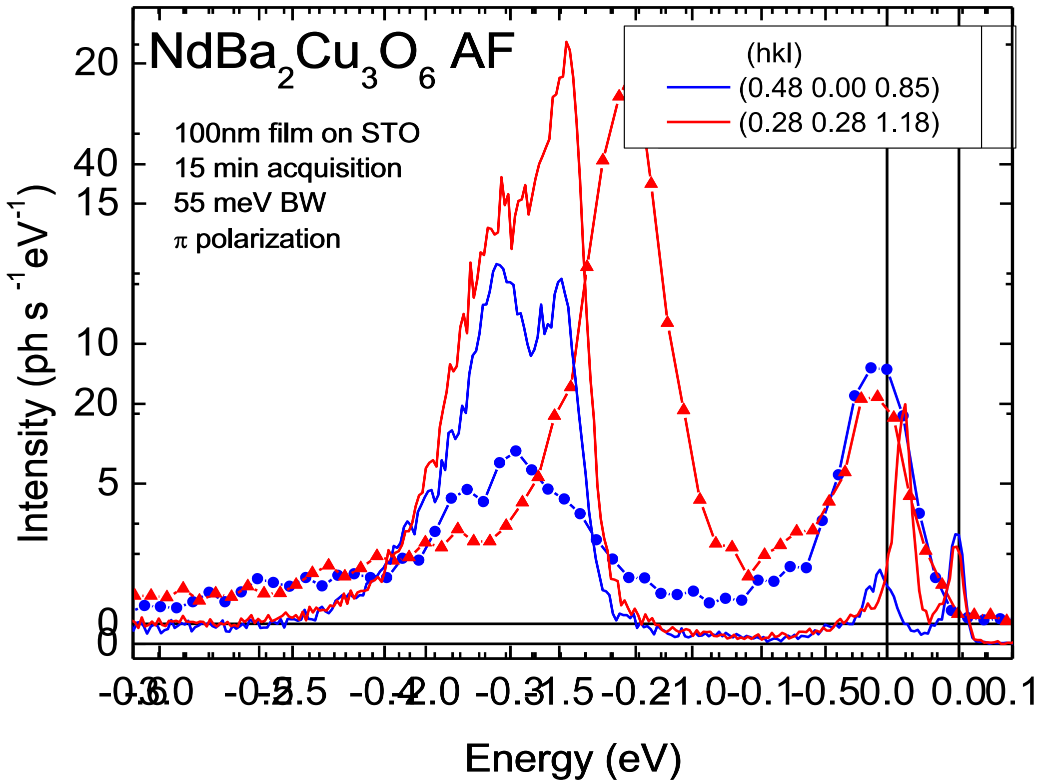
High resolution grating: 2400 mm^{-1}

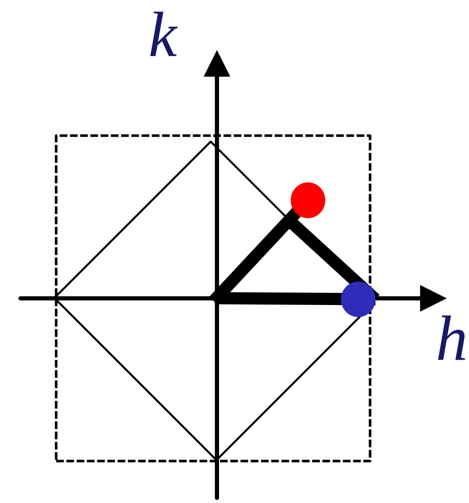
High resolution grating: 2400 mm^{-1}

Medium-resolution grating: 1400 mm^{-1}

NBCO magnetic spectra

Resolution: 55 meV



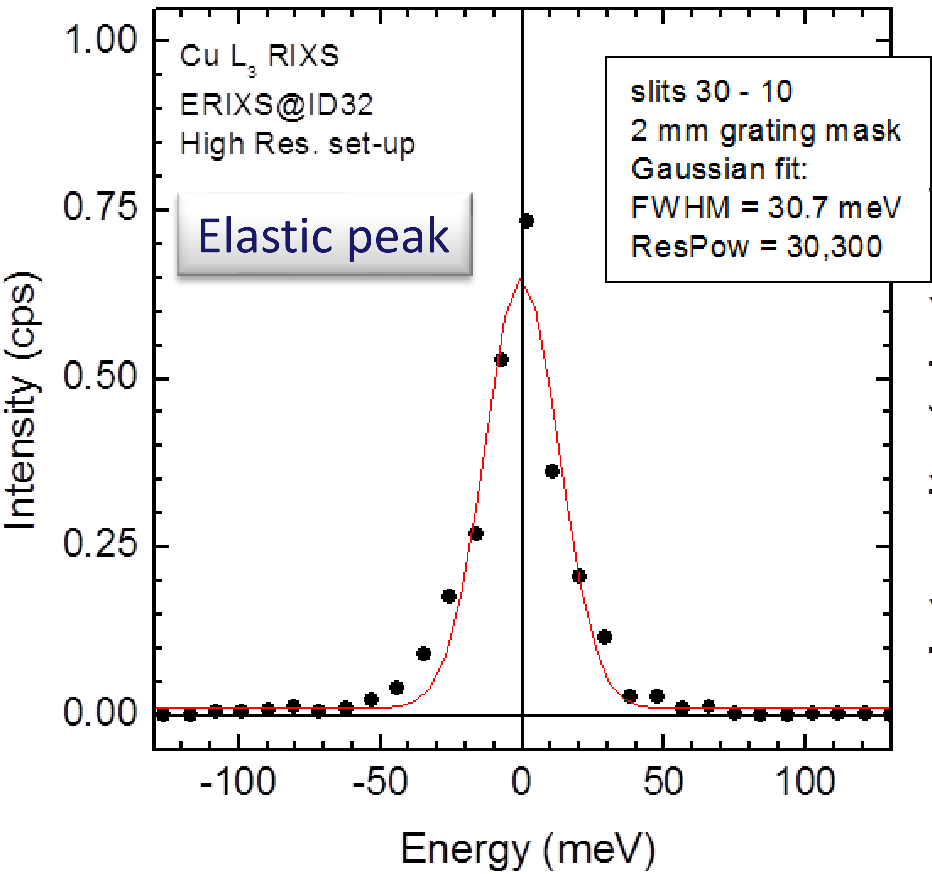


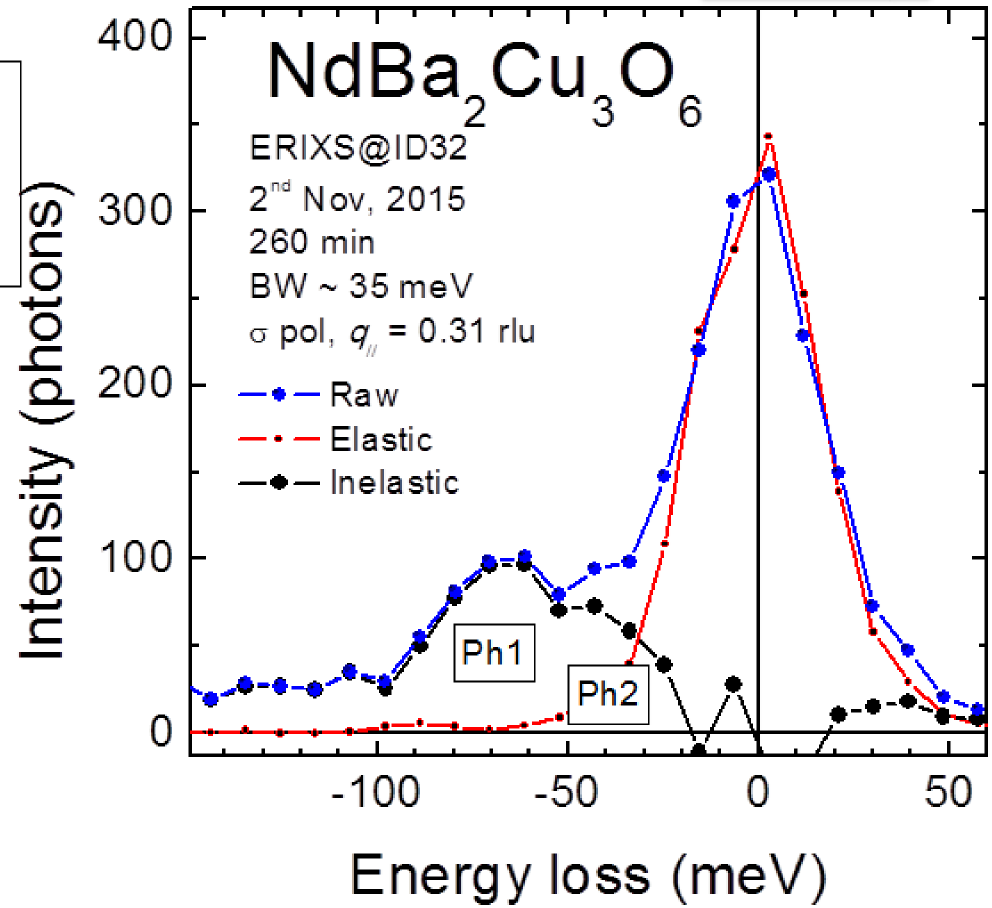
ERIXS: ultra-high resolving power (Nov 2015)

Resolution: 31 meV

High resolution grating: 2400 mm^{-1}

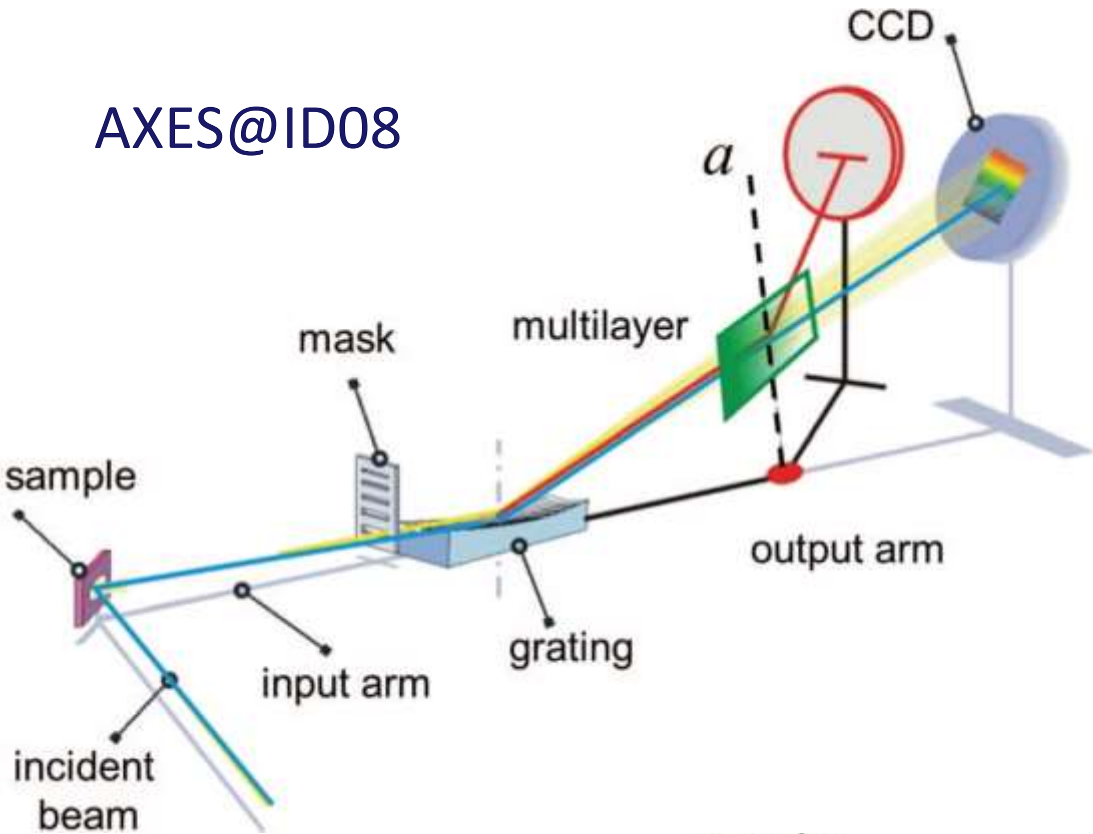
Phonons



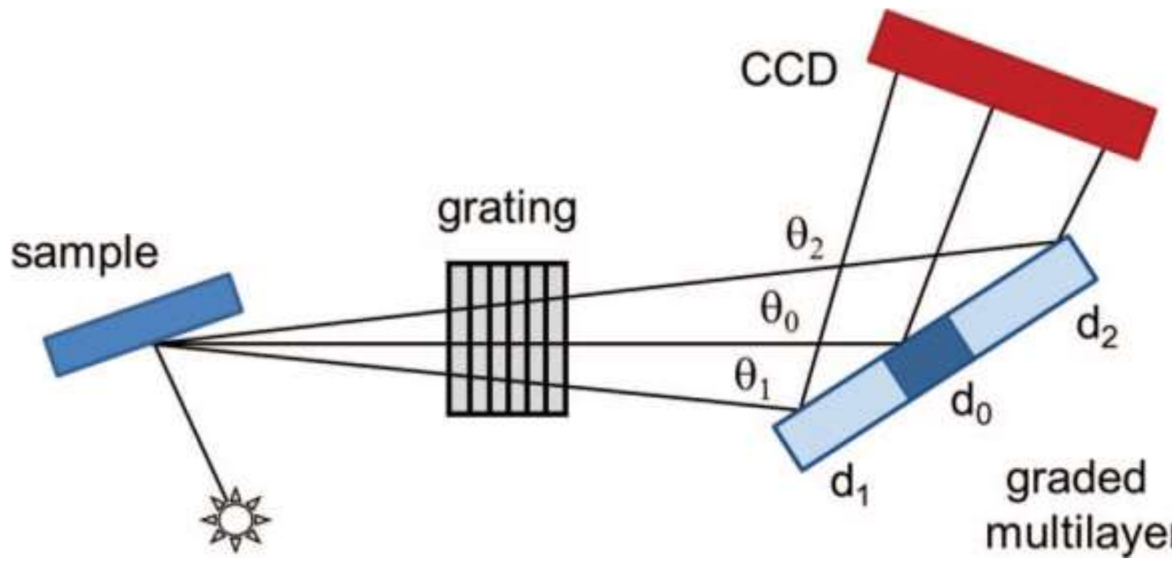


A polarimeter for RIXS spectrometer

AXES@ID08

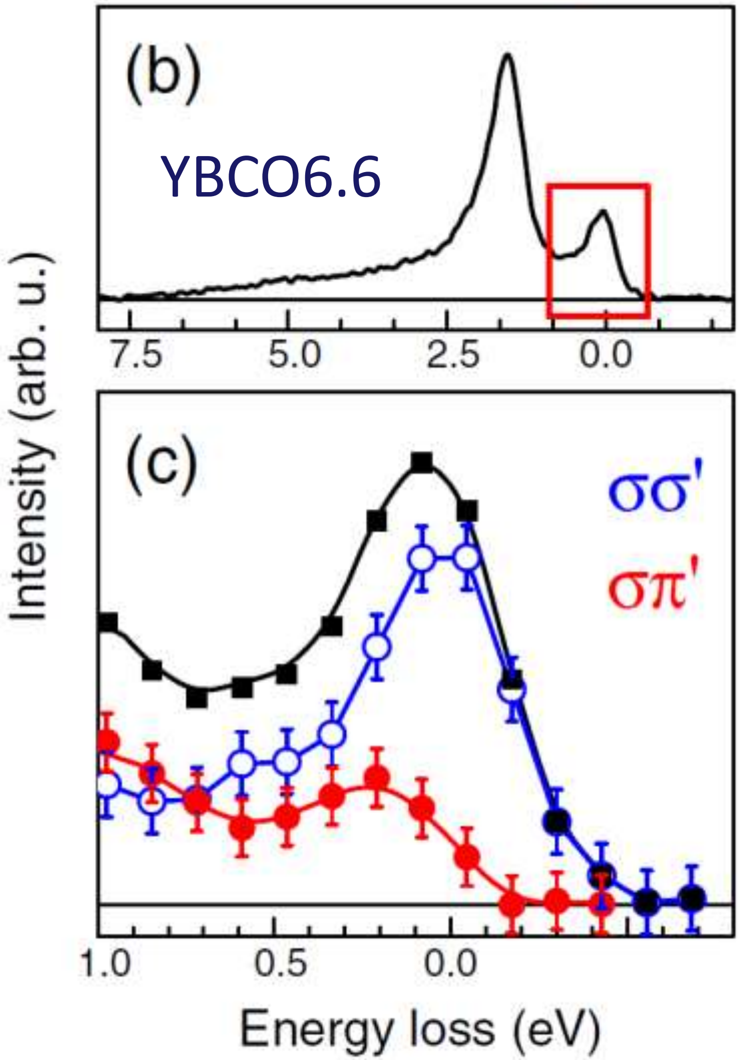


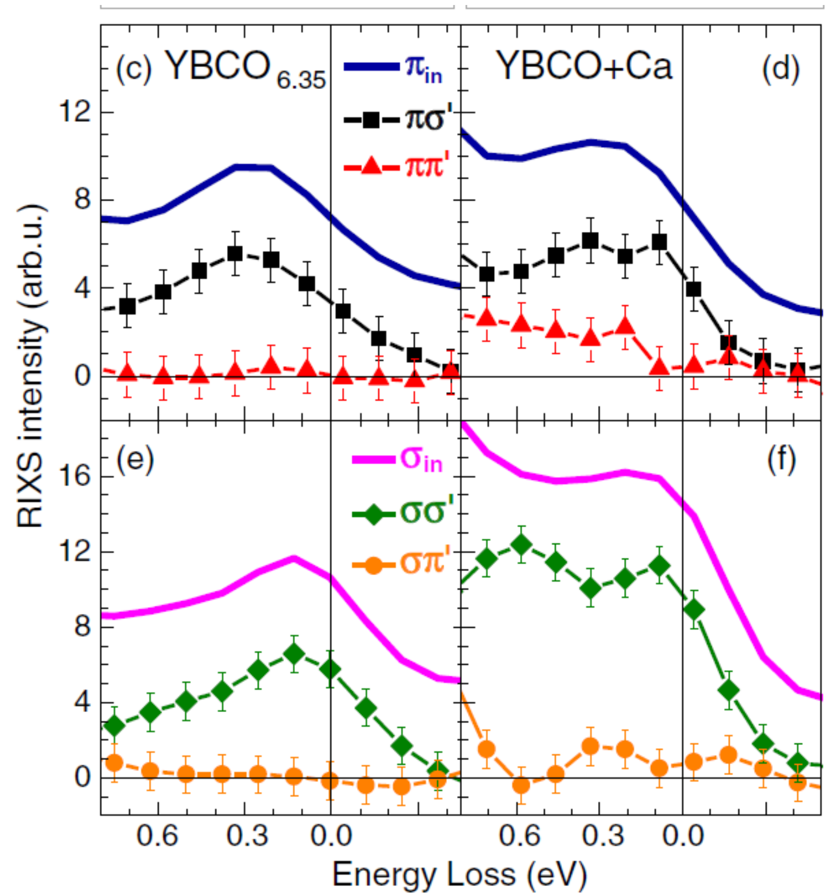
The CCD was rotated when the ML was inserted in the beam. One ML could cover limited energy range (Ni and Cu $L_{2,3}$).



L. Braicovich, M. Minola, G. Dellea, M. Le Tacon, M. Moretti Sala, C. Morawe, J.-Ch. Peffen, R. Supruangnet, F. Yakhou, G. Ghiringhelli, and N. B. Brookes, Rev. Sci. Instrum. **85**,115104 (2014)

AXES Polarimeter

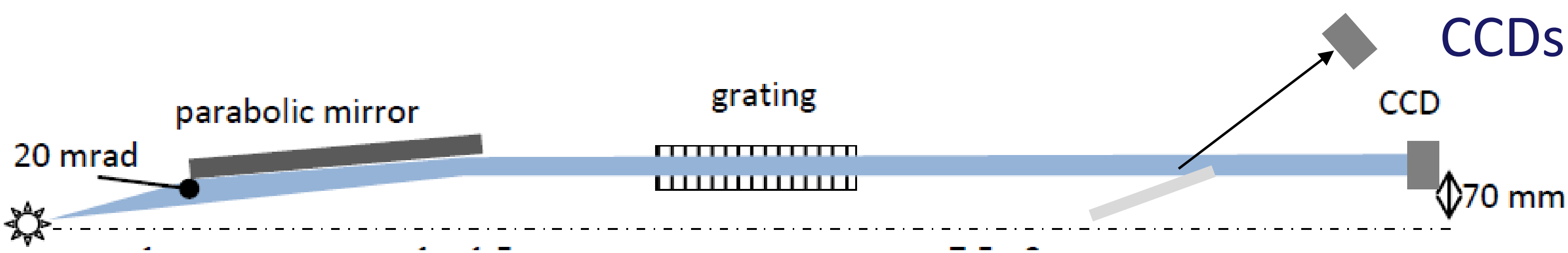


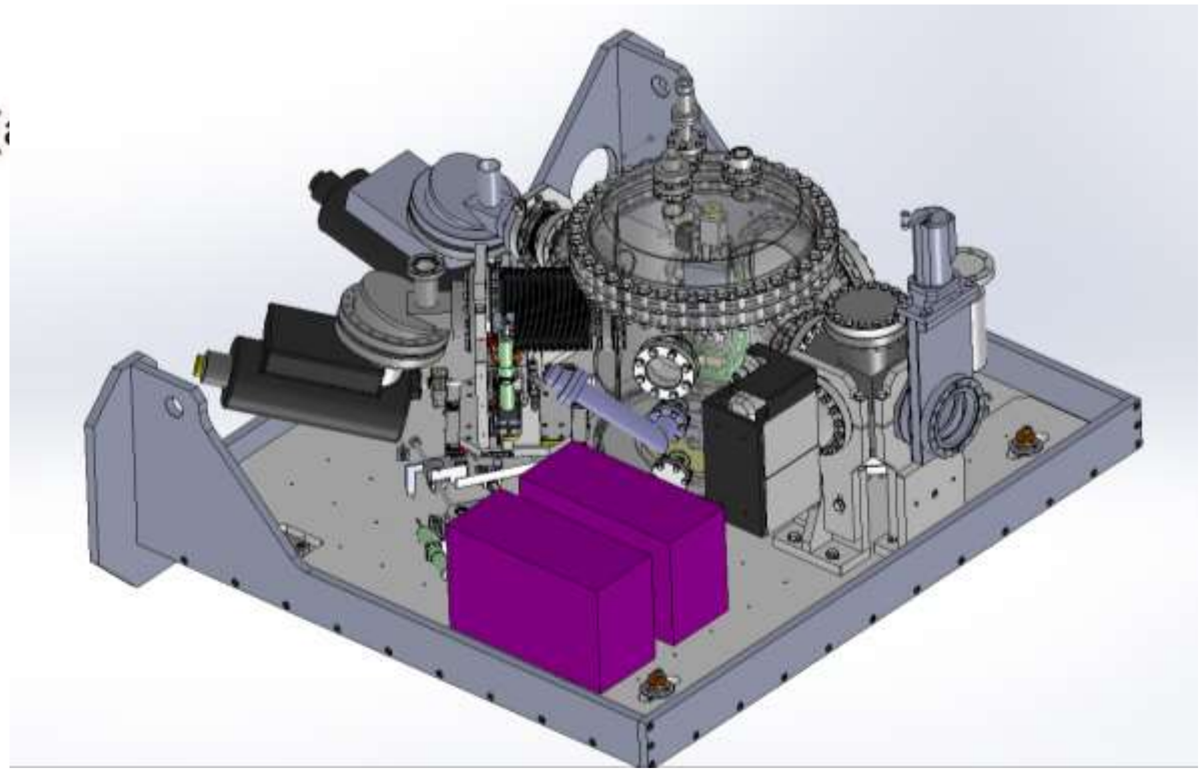


M. Minola, G. Dellea, H. Gretarsson, Y. Y. Peng, Y. Lu, J. Porras, T. Loew, F. Yakhou, N. B. Brookes, Y. B. Huang, J. Pelliciari, T. Schmitt, G. Ghiringhelli, B. Keimer, L. Braicovich, and M. Le Tacon, Phys. Rev. Lett. 114, 217003 (2015)

ERIXS Polarimeter

2 identical CCDs



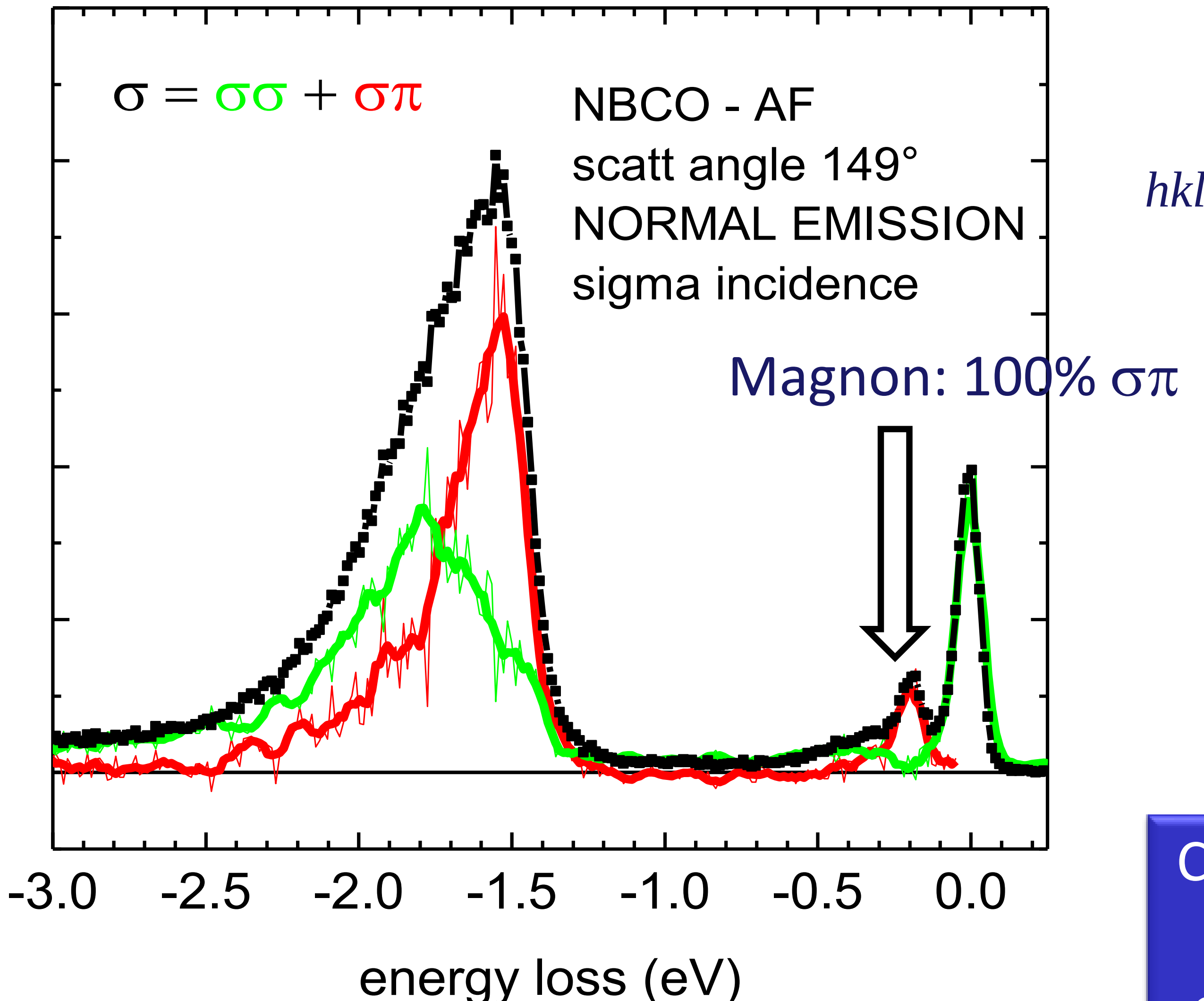


Covering with 2 ML mirrors most of the 520-1000 eV range

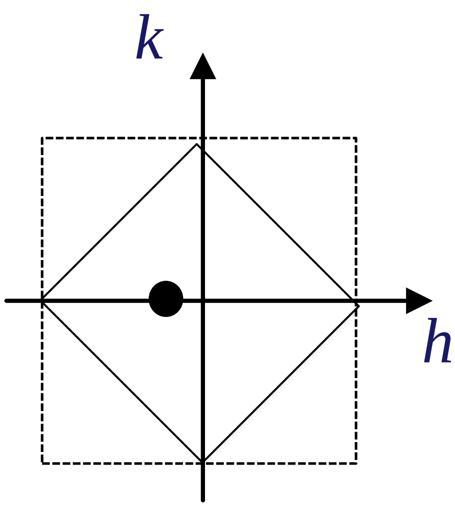
An evolution of the prototype made for AXES@ESRF that was used for real measurements

L. Braicovich, M. Minola, G. Dellea, M. Le Tacon, M. Moretti Sala, C. Morawe, J.-Ch. Peffen, R. Supruangnet, F. Yakhou, G. Ghiringhelli, and N. B. Brookes, Rev. Sci Instrum. 85, 115104 (2014)

ERIXS Polarimeter: first spectrum



hkl = (-0.10, 0.00, 1.63)



Commissioning has just started, not open to users yet.

ERIXS and the other HR soft-RIXS projects

SR FACILITY	E/ΔE (combined)	Length	YEAR	NOTES
ESRF, ERIXS@ID32	30,000	11 m	2015	With Polarimeter
DIAMOND, IXS	40,000	14 m	2017	
MAX IV, Veritas	40,000	?	2017	Rowland Geometry
NSLS II, Centurion@SIX	70,000	15 m	2017	Hettrick-Underwood, 50 nrad slope error, 1 um spot on sample
European XFEL	20,000	5 m	2018	For non linear RIXS and pump-probe time-resolved RIXS

Heisenberg RIXS: SCS beam line of European XFEL

*h*RIXS consortium

The Heisenberg RIXS project at the European XFEL

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Marco Grioni, Ecole Polytechnique Fédérale de Lausanne

Rafael Abela, Paul-Scherrer-Institute

Torsten Schmitt, Paul-Scherrer-Institute

Bernhard Keimer, Max-Planck-Institut für Festkörperforschung Stuttgart

Simone Techert, Max-Planck-Institut für biophysikalische Chemie Göttingen

Jan-Erik Rubensson, Uppsala University

Joseph Nordgren, Uppsala University

Hao Tjeng, Max-Planck-Institut für Chemische Physik fester Stoffe Dresden

Matias Bargheer, Uni Potsdam

Jeroen van den Brink, IFW Dresden

Simo Huotari, Oulu, Finland

Maurits W. Haverkort, Max-Planck-Institut für Festkörperforschung Stuttgart

Frank de Groot, Utrecht University

Stefan Eisebitt, Technische Universität Berlin

Andrea Cavalleri, CFEL Hamburg, Oxford U.

Marc Simon, Laboratoire Chimie-Physique-Matière et Rayonnement, SOLEIL, Paris

Alexei Erko, Helmholtz-Zentrum Berlin

Alexander Föhlisch, Helmholtz-Zentrum Berlin and Uni Potsdam

*h*RIXS working group

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European XFEL: **Andreas Scherz**

Politecnico Milano: **Ying Ying Peng, Giacomo Ghiringhelli**

Uni Potsdam: **Alexander Föhlisch**

Unique performance of XFEL in brillance and time structure

+ Average brillance/brightness:

ESRF ID32: 5×10^{15} ph/s

NSLS II: $6\text{-}8 \times 10^{15}$ ph/s

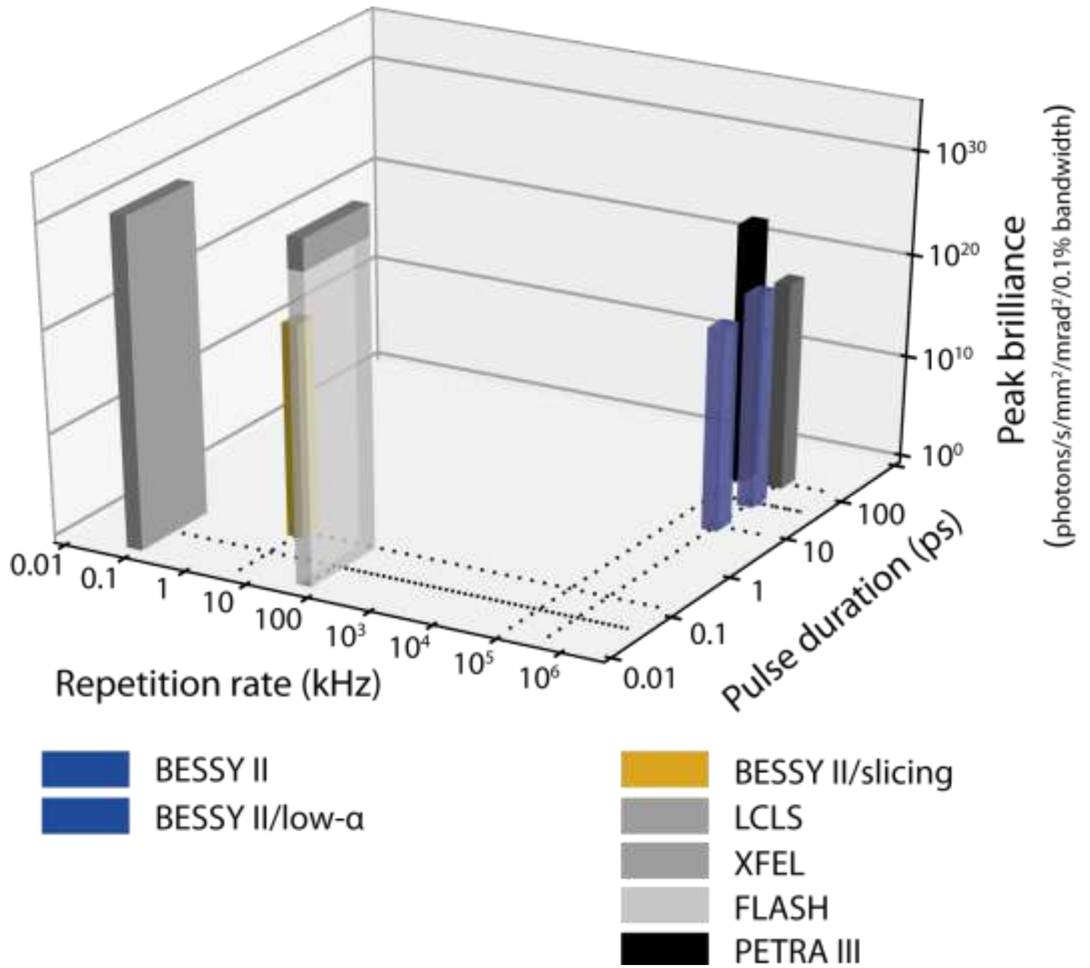
XFEL SASE3: 5×10^{17} ph/s

→ energy resolution

→ polarization analysis

→ rapid data acquisition

→ 3D-momentum-transfer



+ peak brillance

+ femtosecond time structure:

→ Driven and transient phases

→ Chemical pathways

→ Non-linear X-ray science

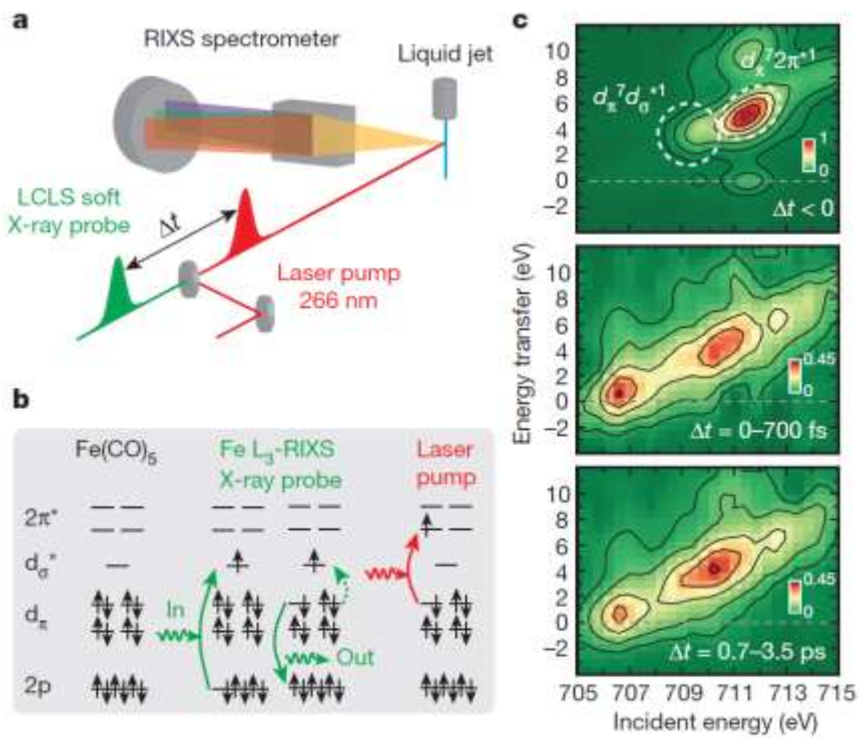
Pump-Probe RIXS

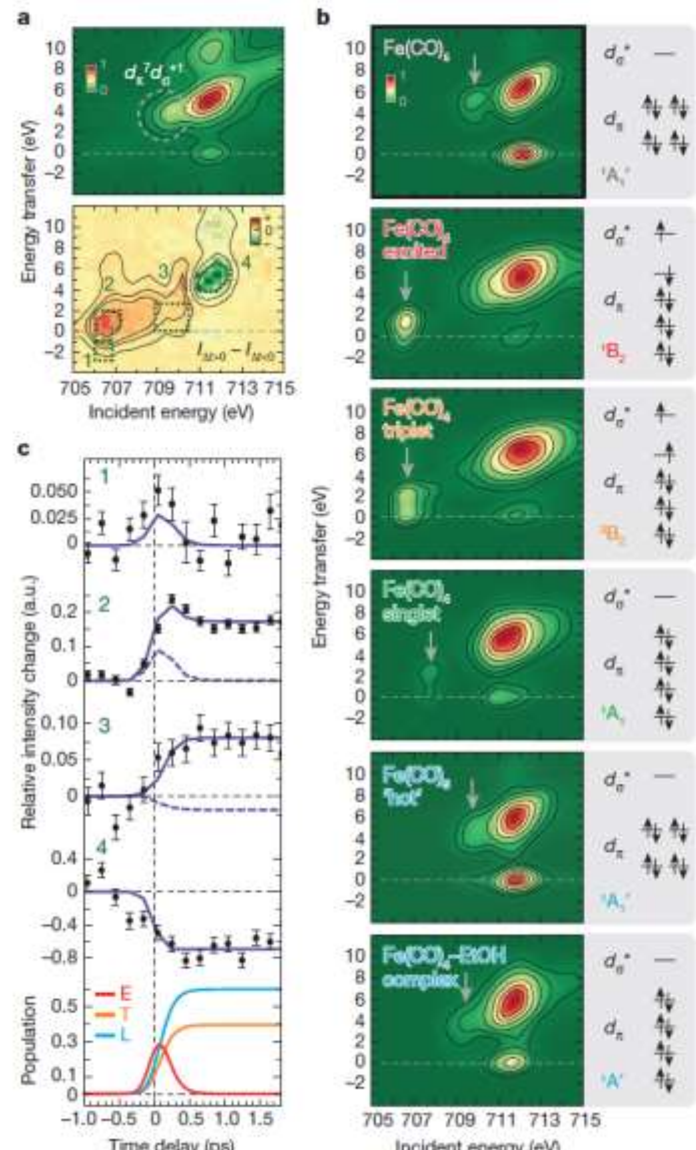
LETTER

doi:10.1038/nature14296

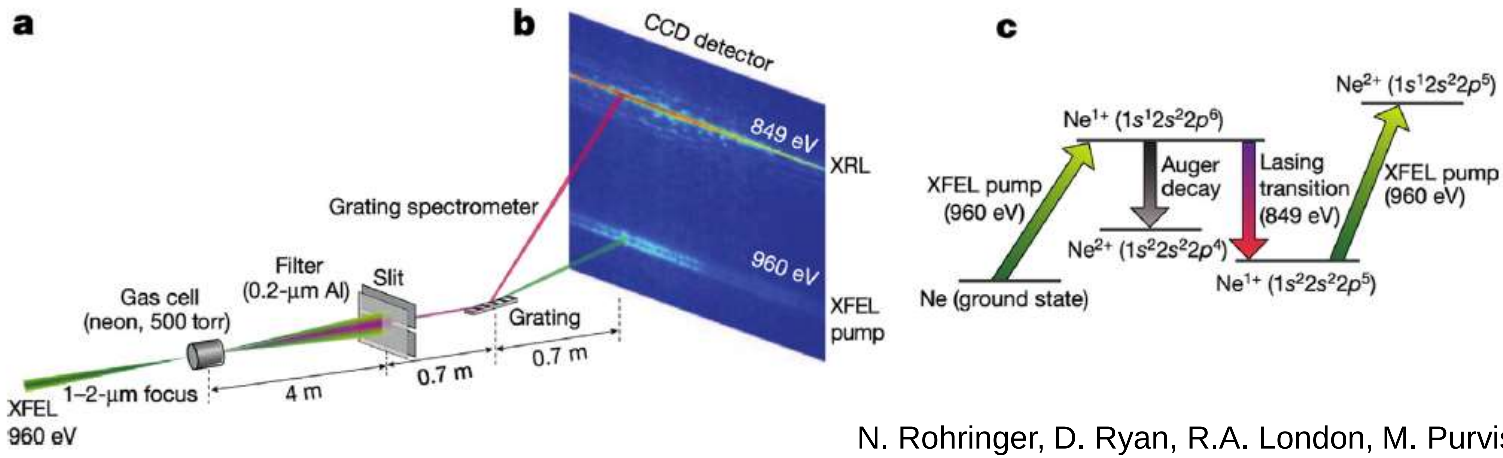
Orbital-specific mapping of the ligand exchange dynamics of $Fe(CO)_5$ in solution

Ph. Wernet[1], K. Kunnus[1,2], I. Josefsson[3], I. Rajkovic[4]†, W. Quevedo[4]†, M. Beye[1], S. Schreck[1,2], S. Grübel[4]†, M. Scholz[4], D. Nordlund[5], W. Zhang[6]†, R. W. Hartsock[6], W. F. Schlotter[7], J. J. Turner[7], B. Kennedy[8]†, F. Hennies[8], F. M. F. de Groot[9], K. J. Gaffney[6], S. Techert[4,10,11], M. Odelius[3] & A. Föhlisch[1,2]

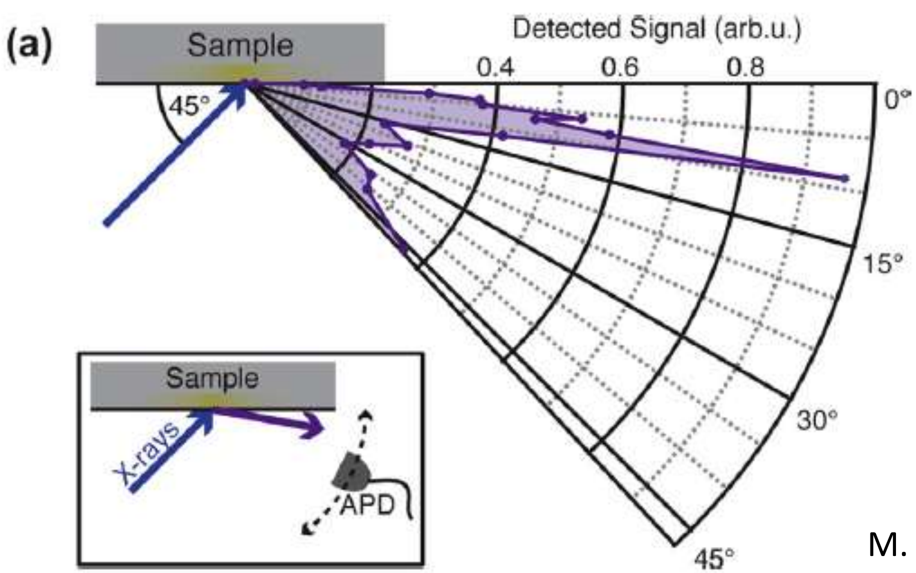




Stimulated emission



N. Rohringer, D. Ryan, R.A. London, M. Purvis, F. Albert, J. Dunn, et al., Atomicinner-shell X-ray laser at 1.46 nanometres pumped by an X-ray free-electronlaser, Nature 481 (2012) 488–491



M. Beye, S. Schreck, F. Sorgenfrei, C. Trabant, N. Pontius, C. Schüßler-Langeheine, et al., Stimulated X-ray emission for materials science, Nature501 (2013) 191–194,

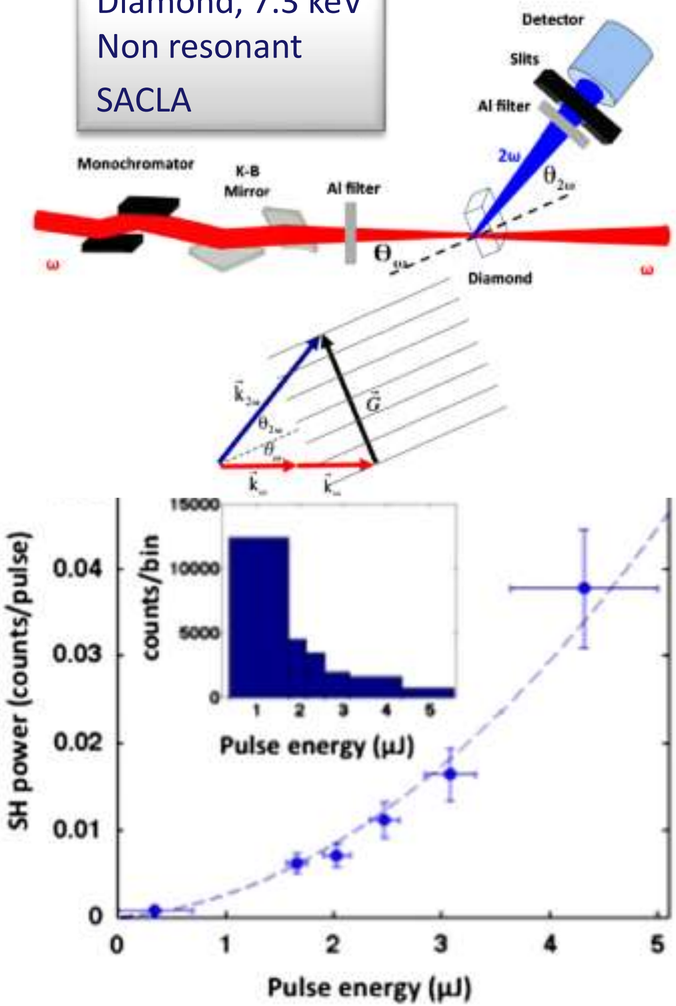
RIXS at XFEL: beyond 1 photon at a time

PRL 112, 163901 (2014) PHYSICAL REVIEW LETTERS

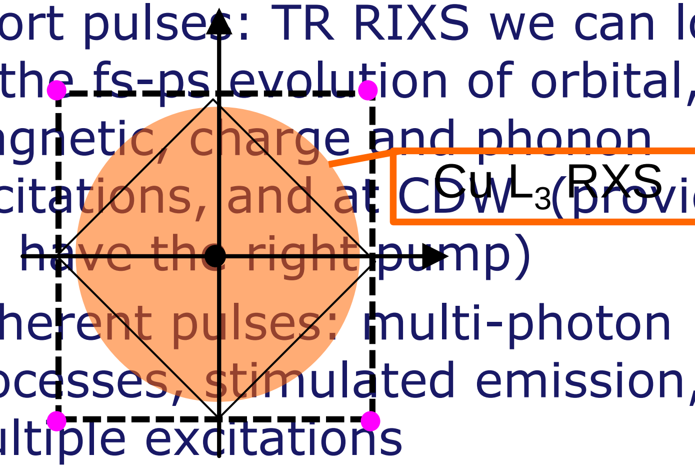
X-Ray Second Harmonic Generation

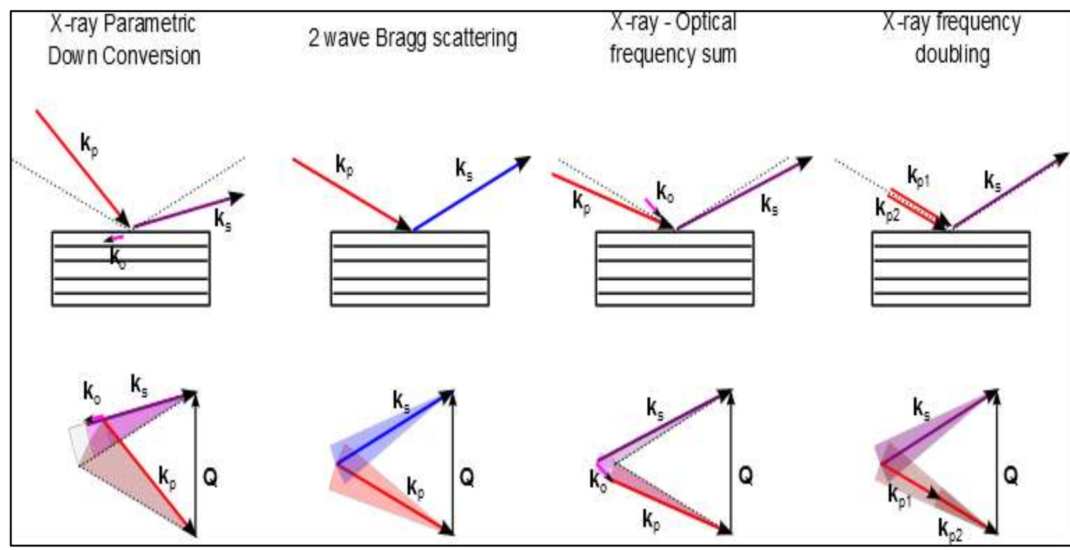
S. Shwartz,[1,2,*] M. Fuchs,[3,4] J. B. Hastings,[5] Y. Inubushi,[6] T. Ishikawa,[6] T.

Diamond, 7.3 keV
Non resonant
SACLA

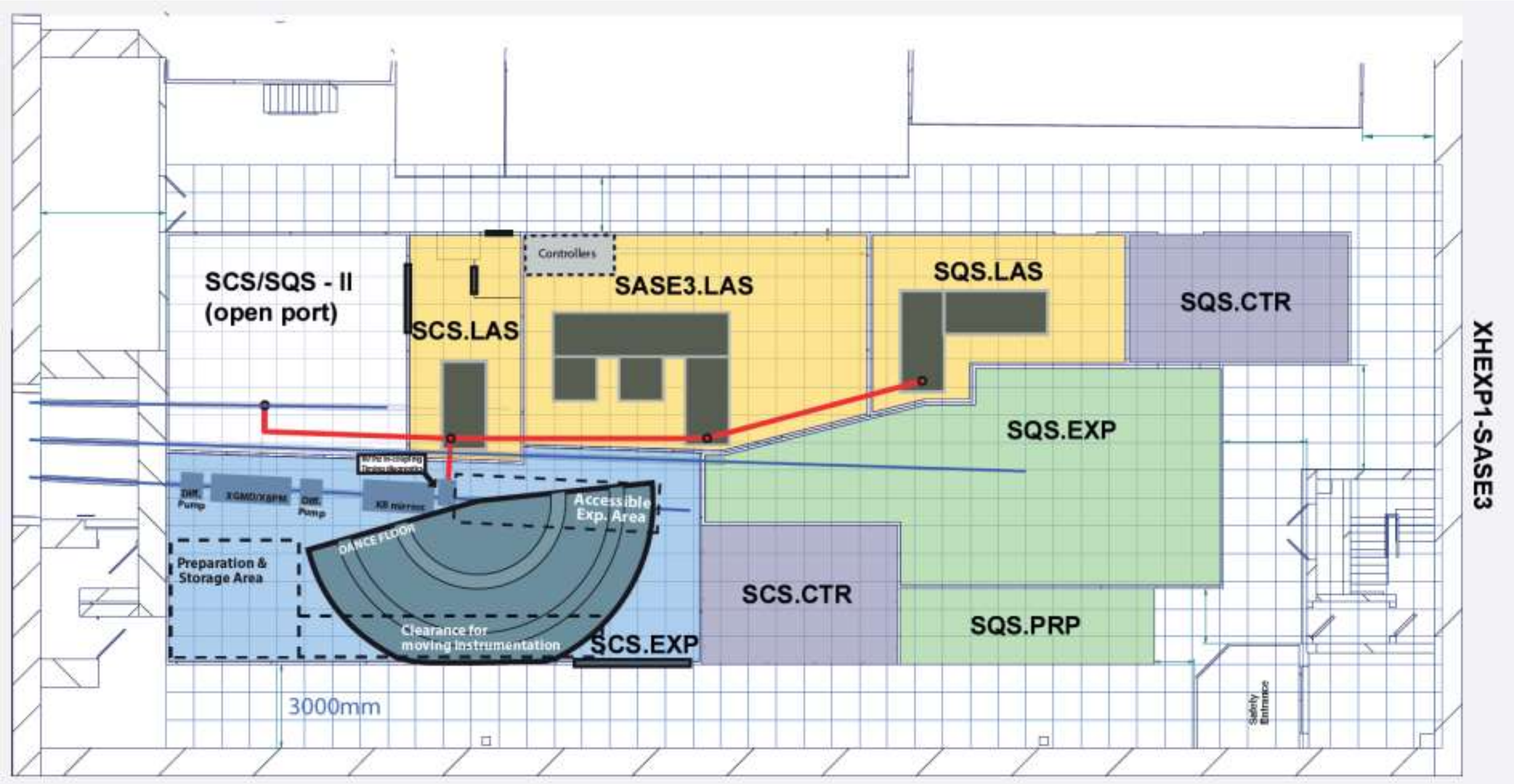


- Short pulses: TR RIXS we can look at the fs-ps evolution of orbital, magnetic, charge and phonon excitations, and at CDW (provided we have the right pump)
- Coherent pulses: multi-photon processes, stimulated emission, multiple excitations





hRIXS: boundary conditions

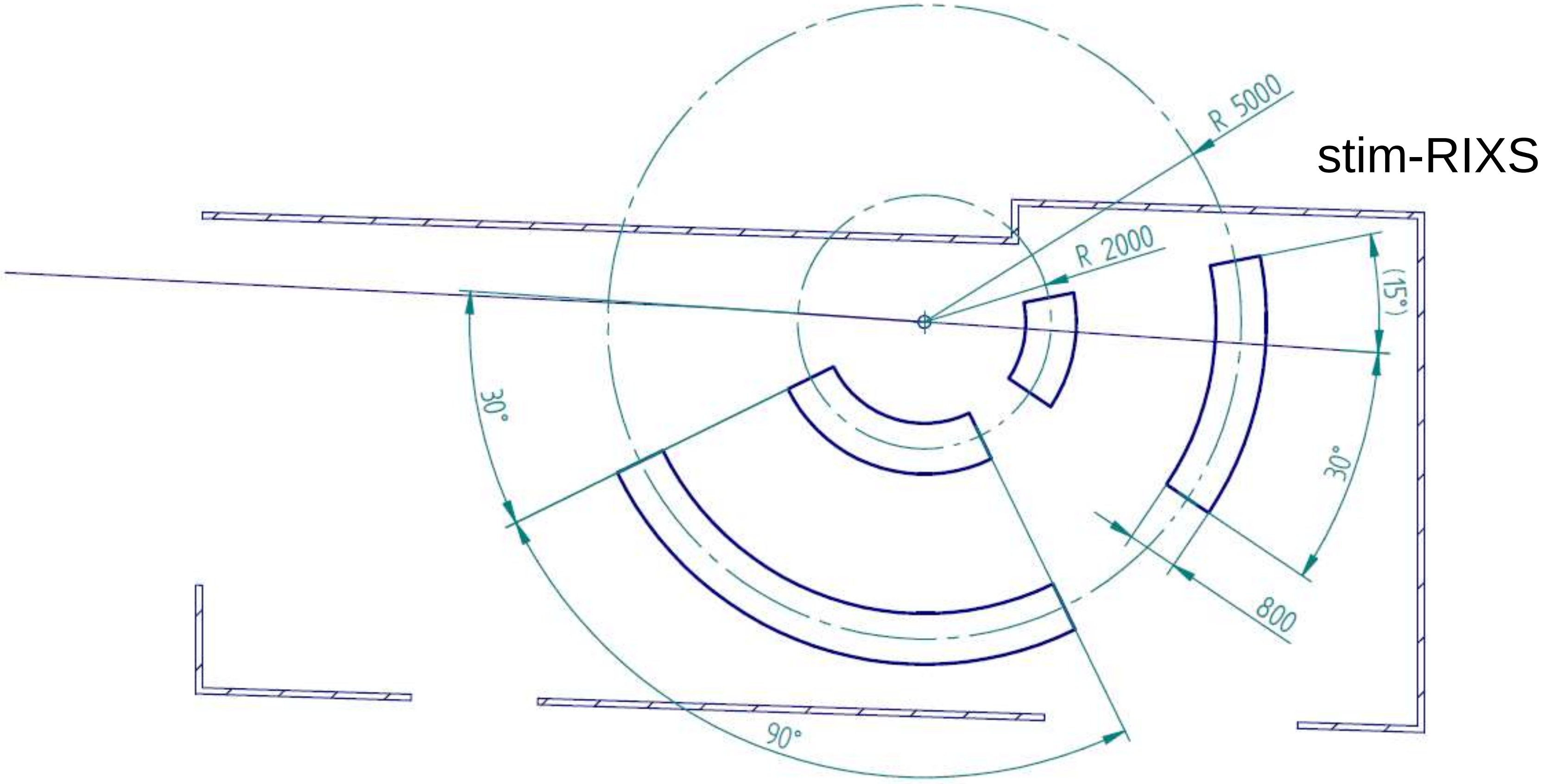


hRIXS: boundary conditions

5-6 m scattering arm

Continuous rotation in backscattering (2θ = 60° - 150°)

Possibility of full forward scattering (2θ = 0° - 20°)

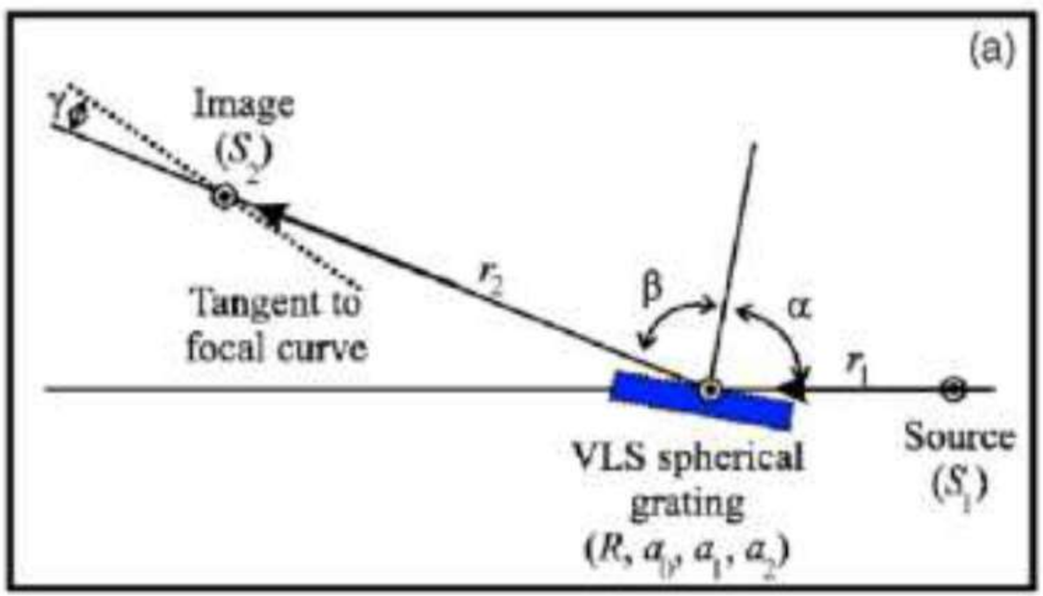


We privilege flexibility

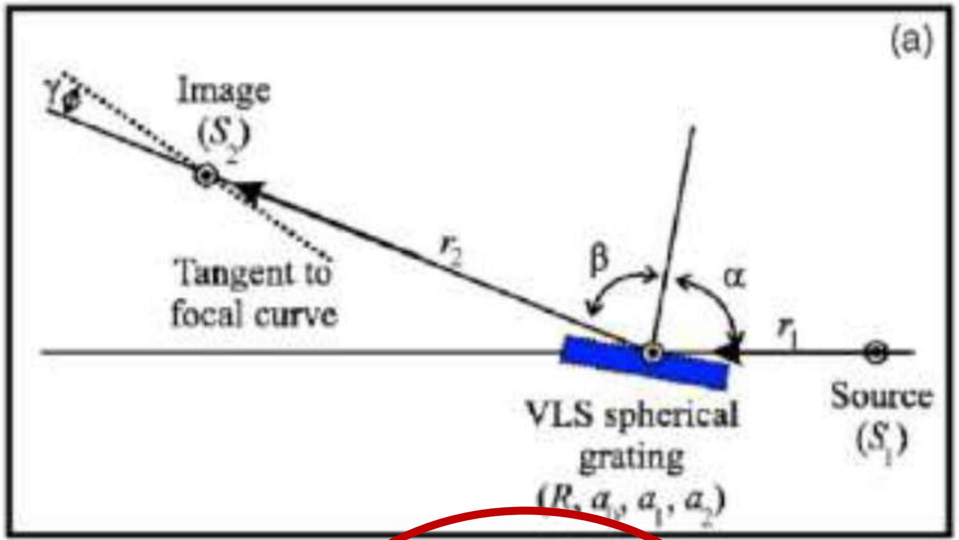
hRIXS will have to work with

- different source size (defocusing on sample to reduce beam damage in some cases)
- different detector resolution (from 10 micron for high resolution CCD to 100-200 micron for pixelated fast detectors)
- 5 m maximum length, that gives at least 5 mrad horizontal acceptance with 1” detectors

Therefore we abandoned the option of the collimating/refocusing mirror, to keep strictly



Optimization of grating parameters



A (1:1)
S_1=10 μm
S_2=10 μm

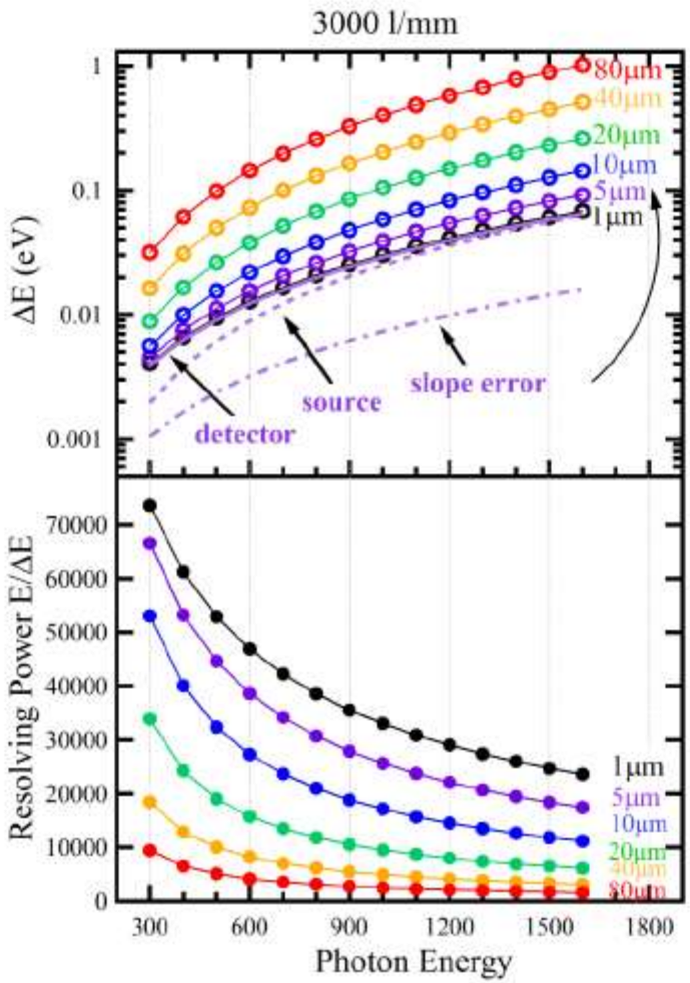
B (1:2)
S_1=5 μm
S_2=10 μm

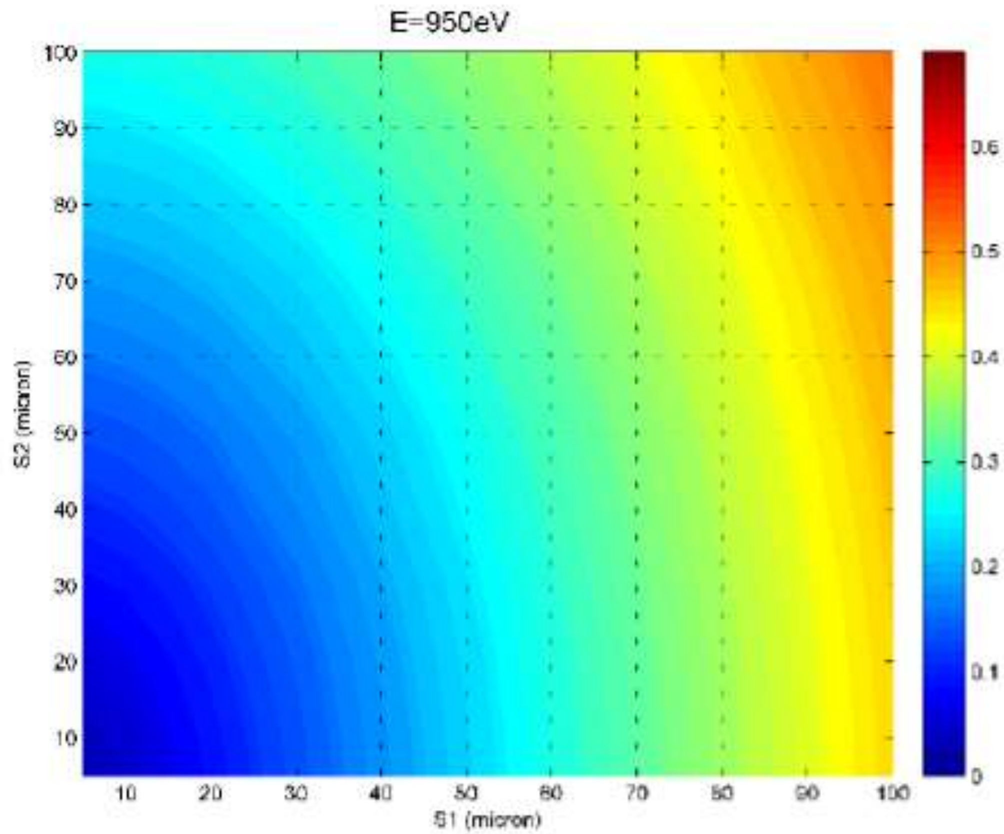
C (1:5)
S_1=20 μm
S_2=100 μm

D (1:20)
S_1=5 μm
S_2=100 μm

E (5:1)
S_1=50 μm
S_2=10 μm

Parameter optimization





Operational end 2017 (?)

YY Peng, GG, *hRIXS optical design report* (2015)

Bibliography

REVIEWS OF MODERN PHYSICS, VOLUME 83, APRIL–JUNE 2011

Resonant inelastic x-ray scattering studies of elementary excitations

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Thanks

Insights into the high temperature superconducting cuprates from resonant inelastic X-ray scattering

M.P.M. Dean

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Volume 376, 15 February 2015, Pages 3–13