ESRF | The European Synchrotron

Magnetic and orbital excitations studied by X-rays

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ESRF – The European Synchrotron

Grenoble, France

Outline

Resonant inelastic X-ray scattering (RIXS)

- ✓ RIXS from a single Cu^{2+} ion
- ✓ RIXS and the magnetic structure factor
- ✓ when magnetic RIXS

RIXS of iridates

- Introduction to the physics of iridates
- RIXS (including XRMS) in iridates
    - ✓ RIXS of a single Ir^{4+} ion
    - ✓ the case of $CaIrO_3$
- Orbital excitations in Ir fluorides
- Magnetic excitations in pure and doped Sr_2IrO_4

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Resonant inelastic X-ray scattering

Resonant – ω_i at an absorption edge (~ 1-10 keV)

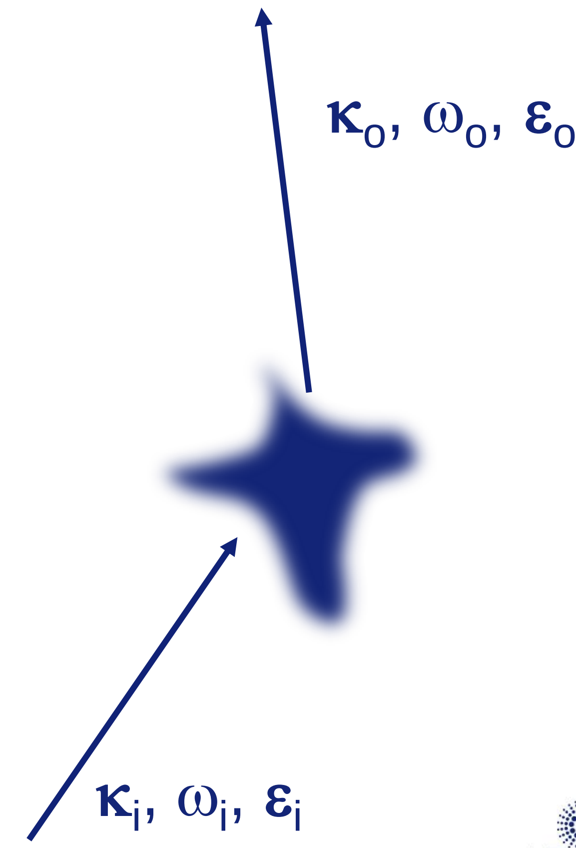
- element sensitivity
- site and orbital selectivity

(In)elastic – energy loss spectroscopy

- probe of excited states (~ 0.01-10 eV)
- …and ground state (REXS/XRMS)

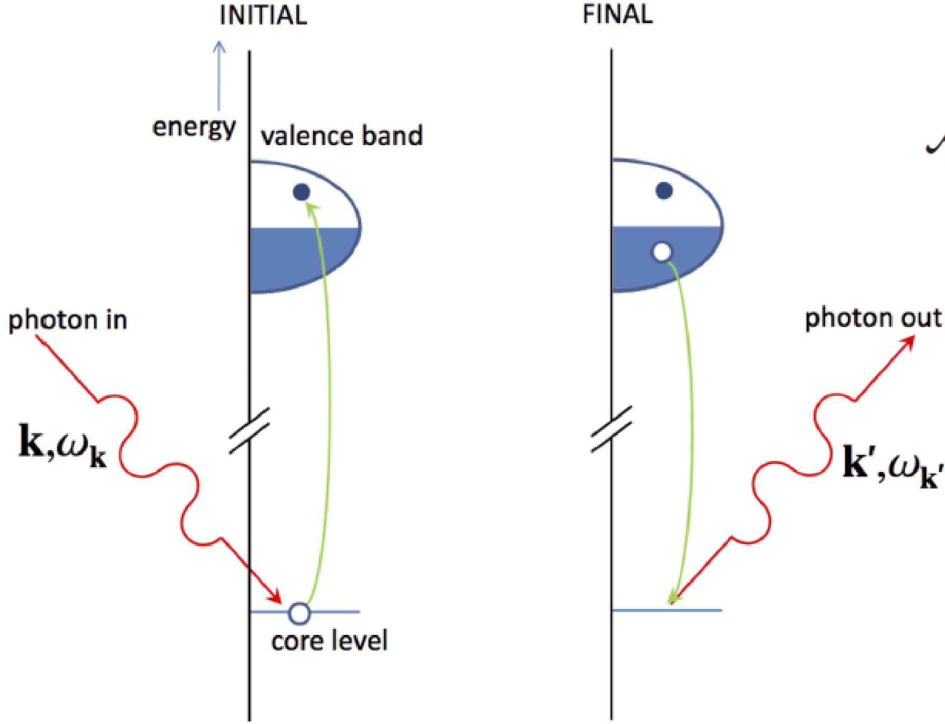
X-ray scattering – photon in/photon out

- neutral excitations
- momentum resolution
- bulk sensitivity



Resonant inelastic X-ray scattering

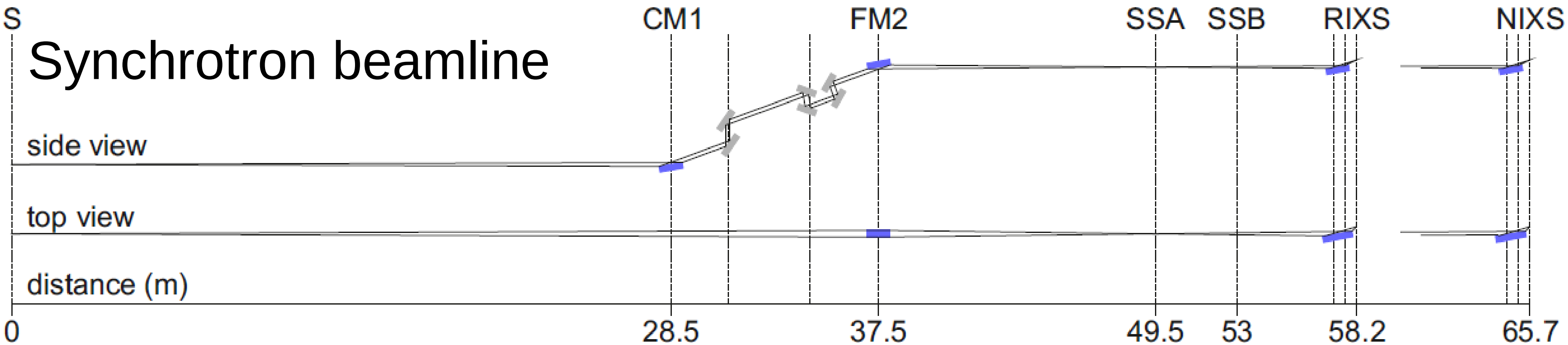
Kramers-Heisenberg equation for the scattering amplitude: a “2-step process”

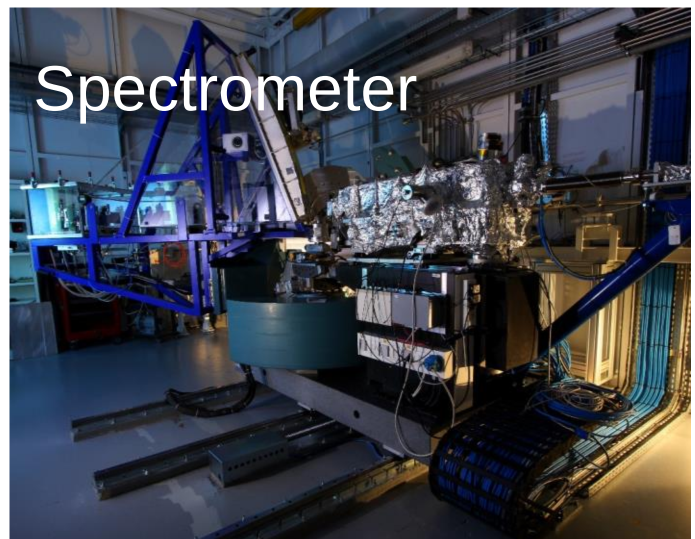


$$\mathcal{A}^{\epsilon\epsilon'}_{|\mathrm{f}\rangle} = \sum_n \frac{\langle \mathrm{f}|\mathcal{D}^\dagger_{\epsilon'}|n\rangle\langle n|\mathcal{D}_\epsilon|\mathrm{g}\rangle}{E_\mathrm{g} - E_n + \hbar\omega + \imath\Gamma_n}$$

L. J. P. Ament et al., Rev. Mod. Phys. 83, 705 (2011)

Resonant inelastic X-ray scattering



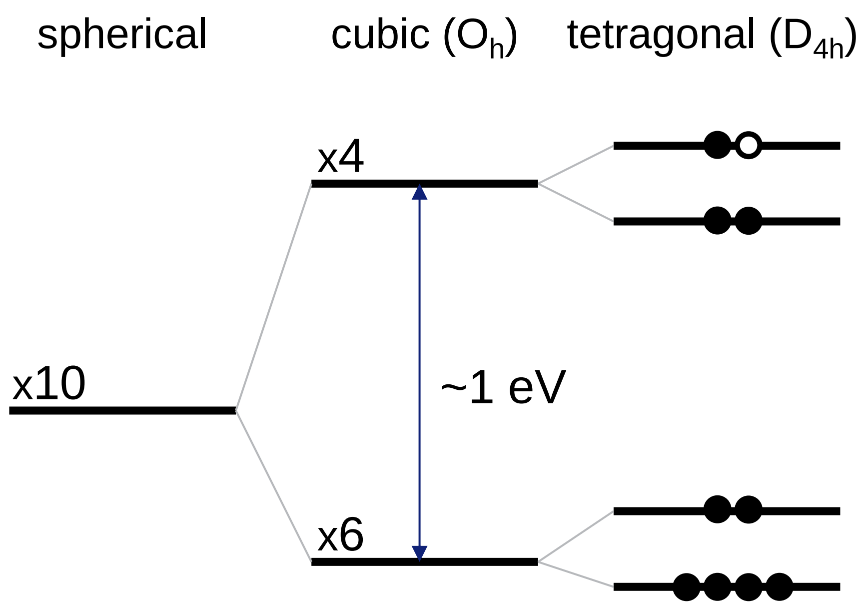


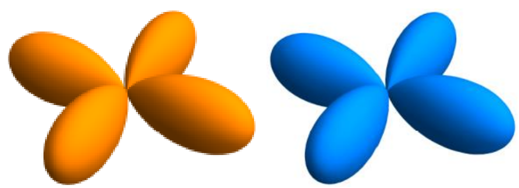
ene. ~ 1-10 keV
flux ~ 10^{13} ph/s
size ~ 10 μm
res. ~ 10 meV

RIXS from a single Cu^{2+} ion

Consider a single Cu^{2+} ion

- $3d^9$ configuration, i.e. one e_g hole system
- trivial multiplet structure (10Dq ~ 1 eV, x^2-y^2 ground state)

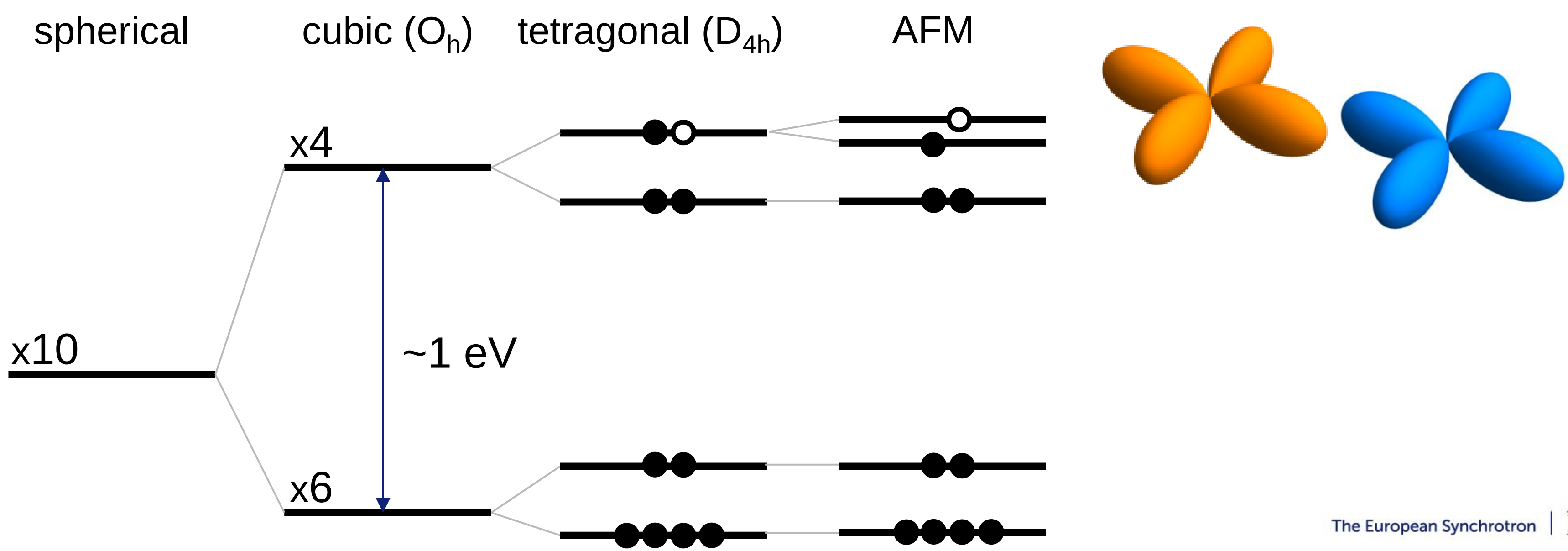




RIXS from a single Cu^{2+} ion

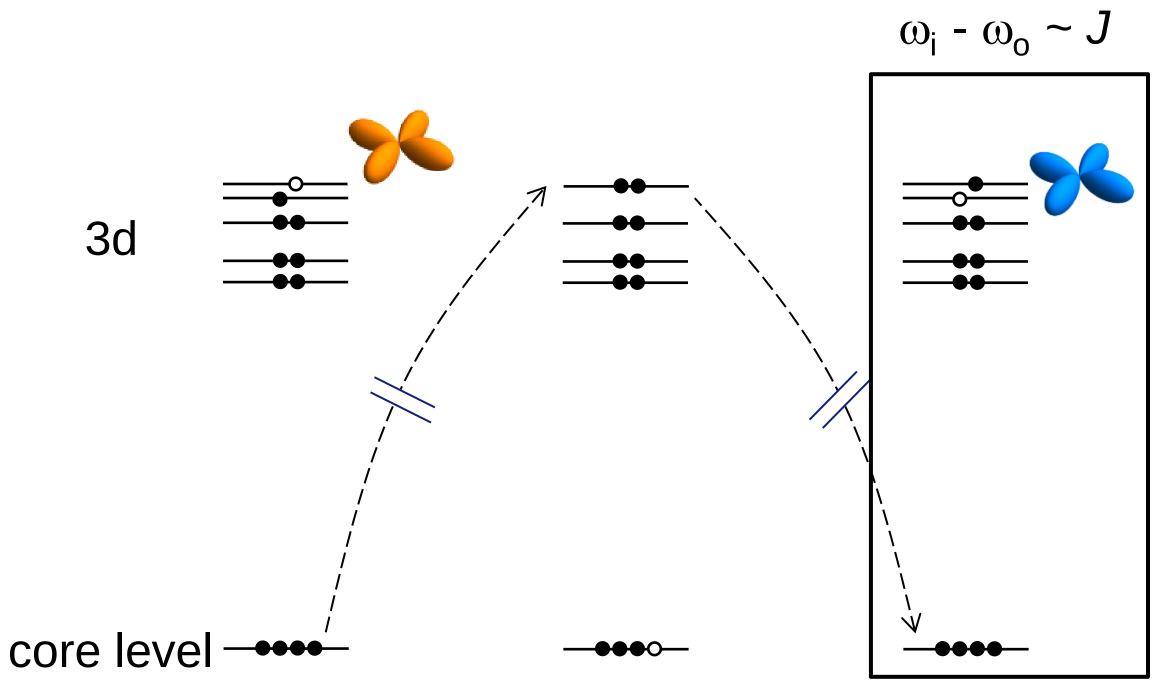
Consider a single Cu^{2+} ion within CuO_2 planes with AFM order

- $3d^9$ configuration, i.e. one e_g hole system
- trivial multiplet structure (10Dq ~ 1 eV, x^2-y^2 ground state)
- the degeneracy of the ground state is lifted (J ~ 100 meV)



RIXS from a single Cu^{2+} ion

Spin-flip is a possible final state of the RIXS process. But…



RIXS from a single Cu^{2+} ion

…how can optical (spin-conserving) transitions give rise to spin-flip?

$$\mathcal{A}^{\epsilon\epsilon'}_{|\ \rangle} = \sum_n \frac{\langle\ |\mathcal{D}^\dagger_{\epsilon'}|n\rangle\langle n|\mathcal{D}_\epsilon|\ \rangle}{E_\mathrm{g} - E_n + \hbar\omega + \imath\Gamma_n}$$

$$\propto \sum_n \langle\ |\mathcal{D}^\dagger_{\epsilon'}|n\rangle\langle n|\mathcal{D}_\epsilon|\ \rangle$$

$$\mathcal{D}_\epsilon = \qquad \mathcal{D}_\mathbf{x}, \qquad \mathcal{D}_\mathbf{y}, \qquad \mathcal{D}_\mathbf{z}$$

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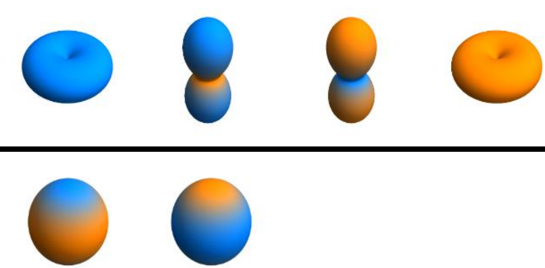
$$\propto \sum_n \langle\ |\mathcal{D}^\dagger_{\epsilon'}|n\rangle\langle n|\mathcal{D}_\epsilon|\ \rangle$$

L$_3$ edge $2p_{3/2}$

L$_2$ edge $2p_{1/2}$



RIXS from a single Cu^{2+} ion

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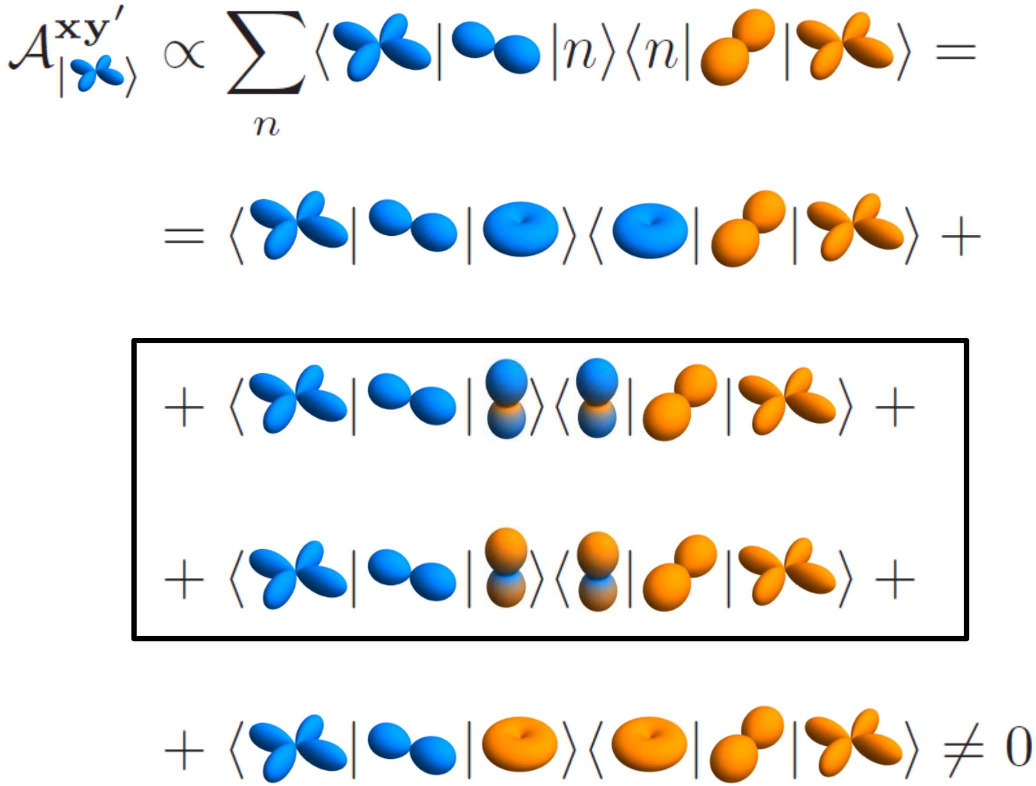


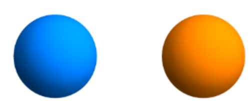
RIXS from a single Cu^{2+} ion

Spin-flip is allowed by spin-orbit coupling in the intermediate state (e.g. $2p_{3/2}$ in case of L_3 edge RIXS)

$$\mathcal{A}^{L_3}_{\text{spin}-\text{flip}} = \begin{array}{c} \begin{array}{ccc} x & y & z \end{array} \\ \begin{pmatrix} 0 & \imath\sin\theta_s & 0 \\ -\imath\sin\theta_s & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \end{array} \begin{array}{c} x' \\ y' \\ z' \end{array}$$

No spin-flip in the absence of spin-orbit coupling

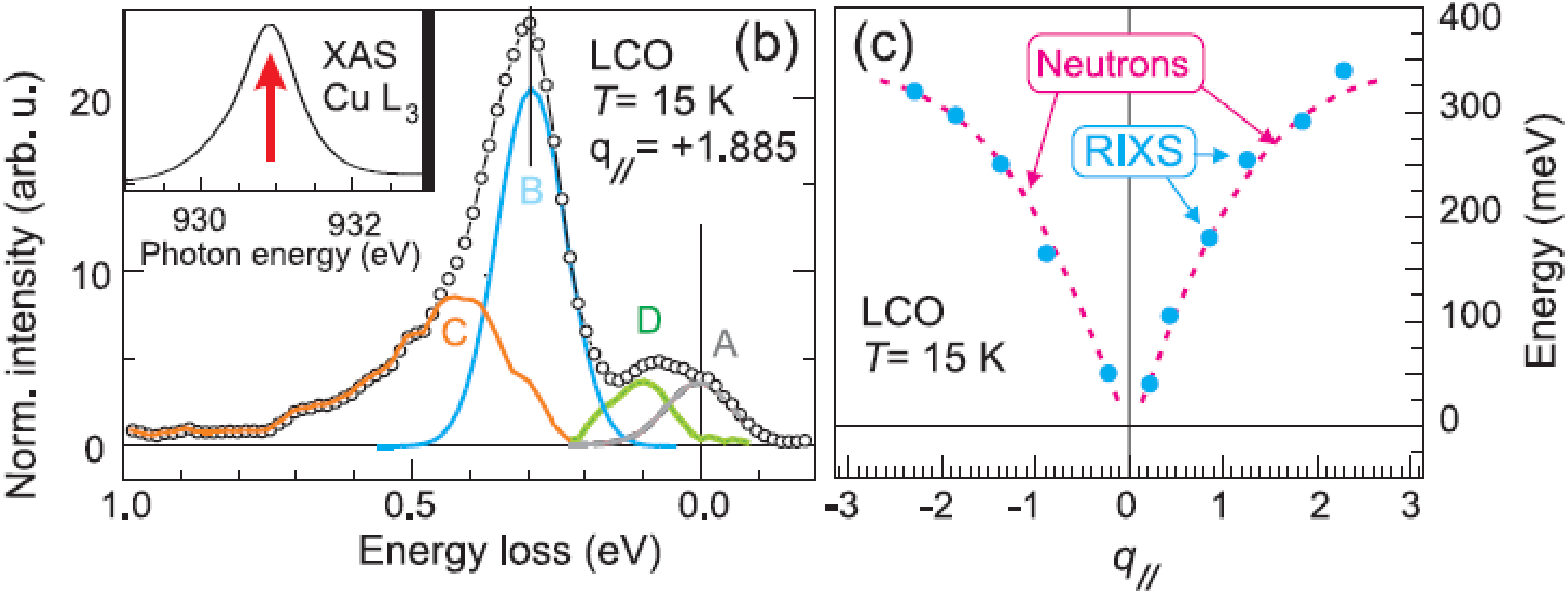
K edge $1s$



L. J. P. Ament et al., Phys. Rev. Lett. 103, 117003 (2009)
M. Moretti Sala, New J. Phys. 13, 043026 (2011)

RIXS from a single Cu^{2+} ion

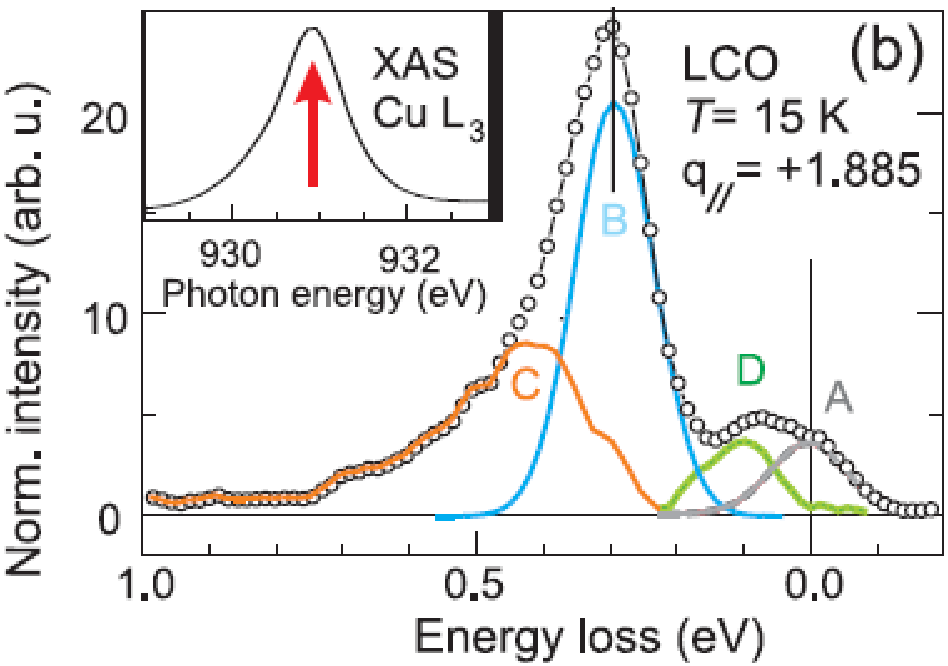
First RIXS evidence of magnons in 2010

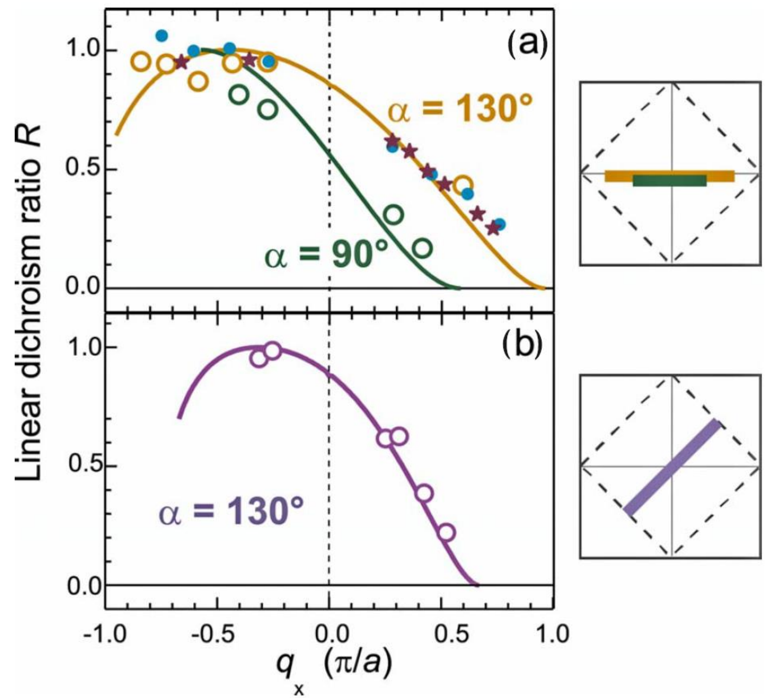


L. Braicovich et al., Phys. Rev. Lett. 104, 077002 (2010)
R. Coldea et al., Phys. Rev. Lett. 96, 174533 (2010) – INS

RIXS from a single Cu^{2+} ion

Semi-quantitative agreement with single Cu^{2+} ion model

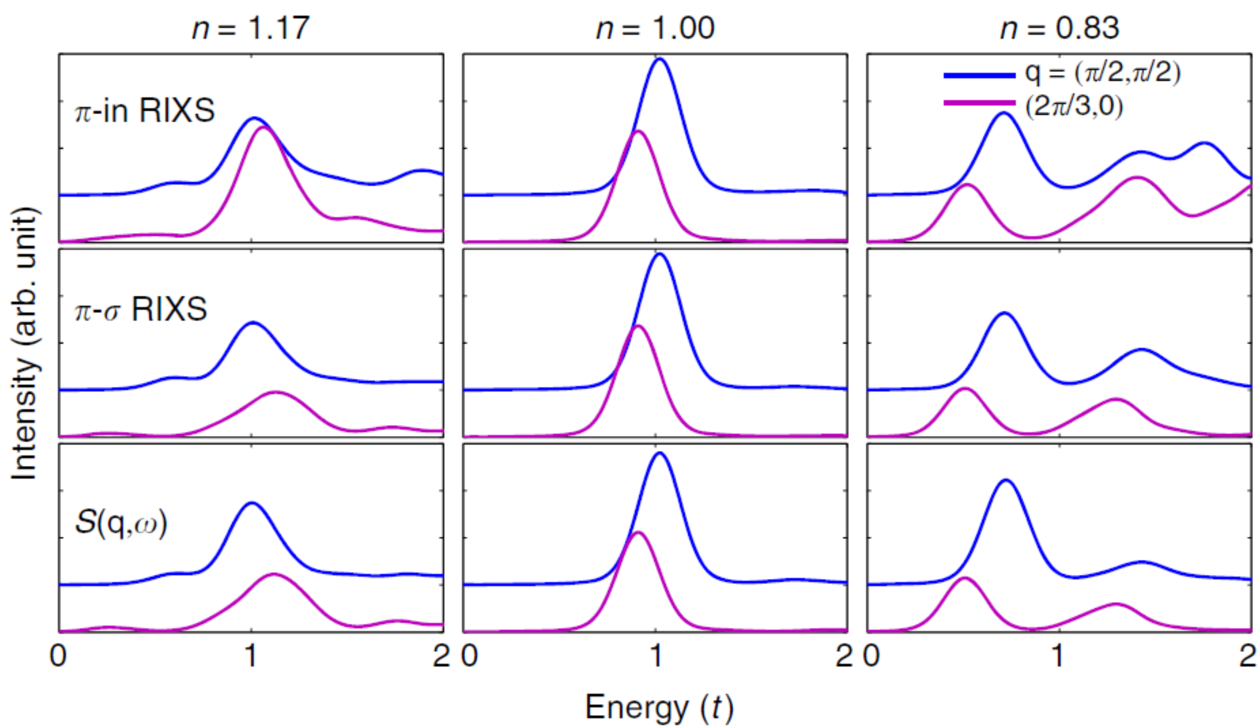




L. Braicovich et al., Phys. Rev. Lett. 104, 077002 (2010)
L. Braicovich et al., Phys. Rev. B 81, 174533 (2010)

RIXS and the magnetic structure factor

RIXS in the cross-polarisation channel is reminescent of S(***q***,ω)



C. J. Jia et al., Nat. Comm. (2014) – exact diagonalisation calculations

when magnetic RIXS

The natural probe of magnetism is neutrons...

- direct coupling to magnetic moments
- easy interpretation of results – $S(\boldsymbol{q}, \omega)$
- unbeatable energy resolution (INS vs. RIXS)

...but unfavourable when

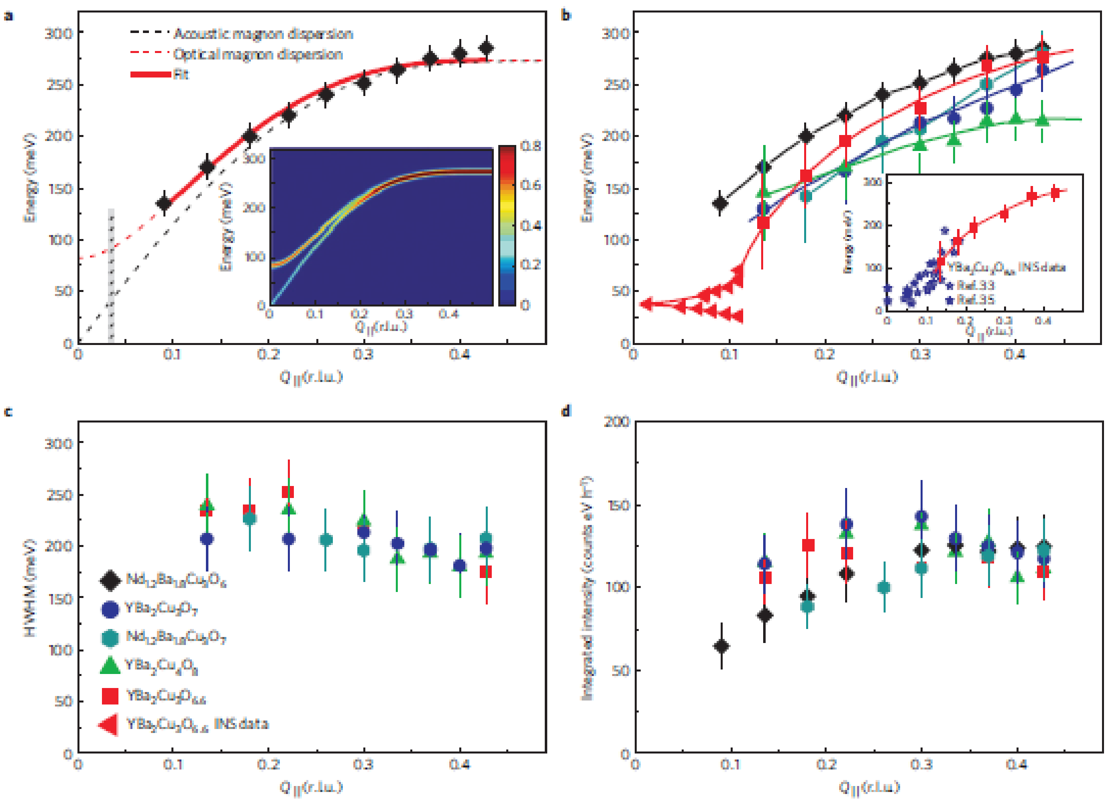
- small sample volumes
- strong neutron absorbers

RIXS if energy resolution is good enough

~ 10 meV nowadays (continuously improving)

when magnetic RIXS

Intense paramagnon excitations in the YBa_2CuO_{6+x} family of high-temperature superconductors

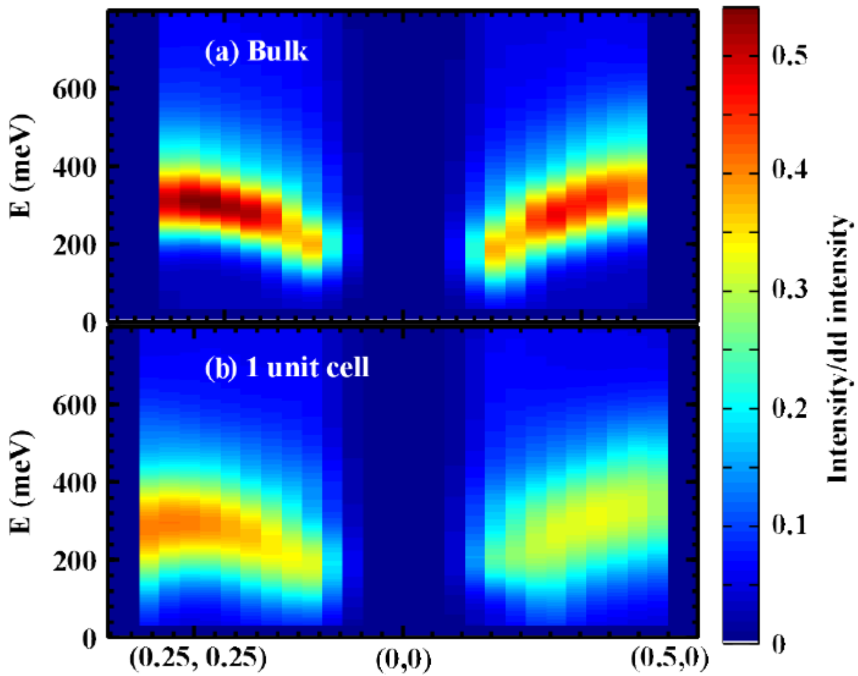


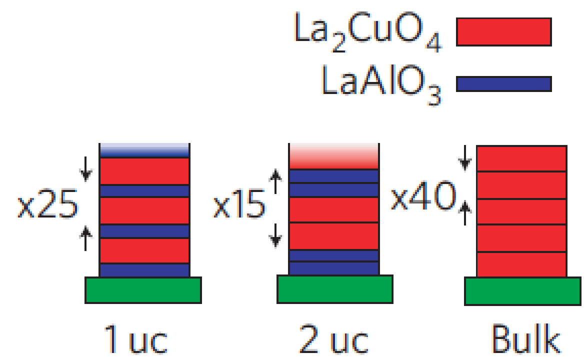
M. Le Tacon et al., Nat. Phys. 7, 725 (2011)

M. Guarise et al., Nat. Commun. 5, 5760 (2014); M. P. M. Dean, et al., Phys. Rev. B 90, 220506 (2014)

when magnetic RIXS

Spin excitations in isolated single La_2CuO_4 layers



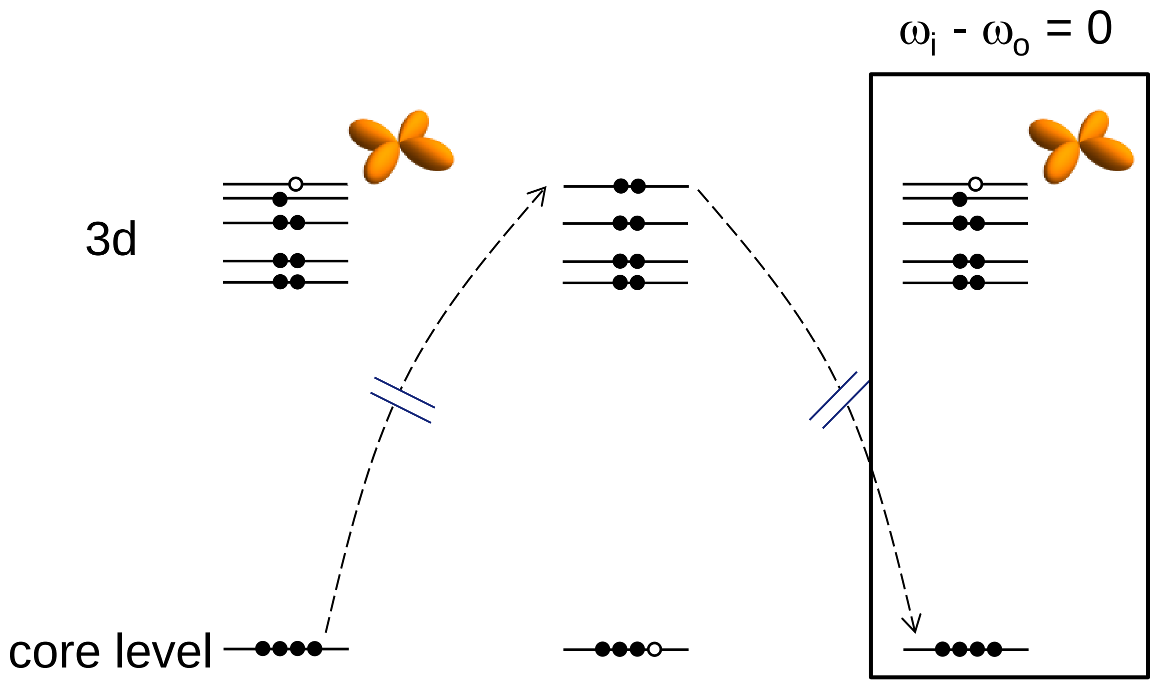


13.2 Å x 25 = 330 Å

M. P. M. Dean et al., Nat. Mat. 11, 850–854 (2012)

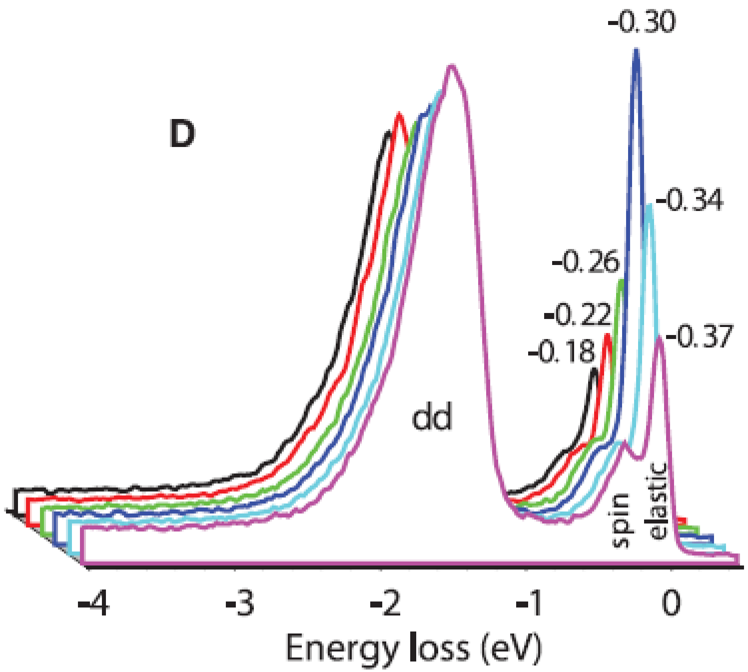
RIXS from a single Cu^{2+} ion

REXS is RIXS back to the ground state



RIXS from a single Cu^{2+} ion

Long-range incommensurate charge fluctuations in $(Y,Nd)Ba_2Cu_3O_{6+x}$



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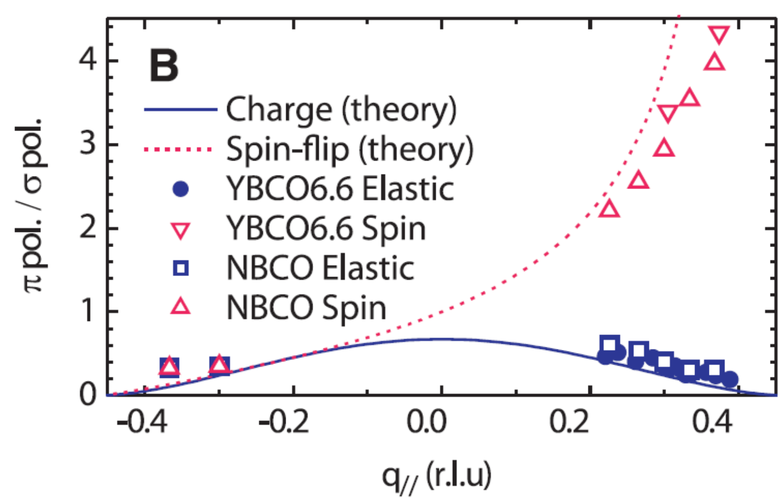

G. Ghiringhelli et al., Science 337, 821-825 (2012)

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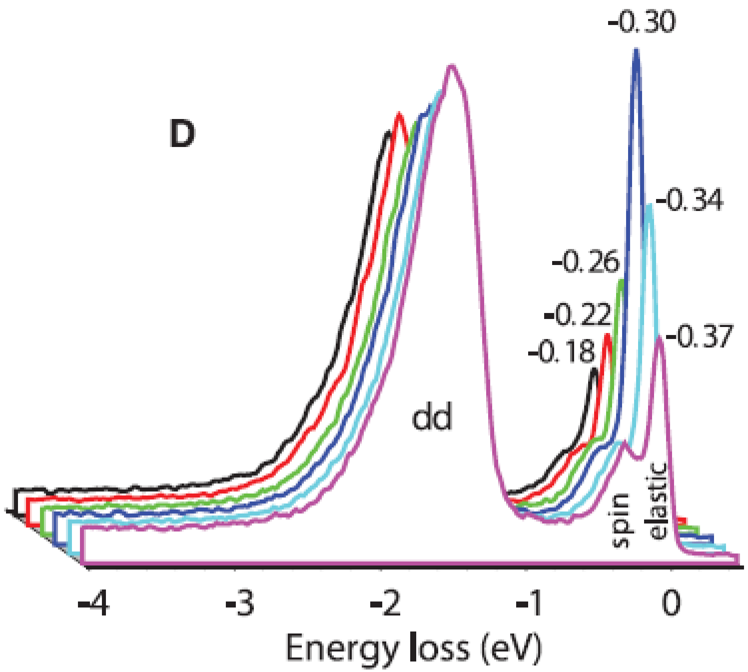
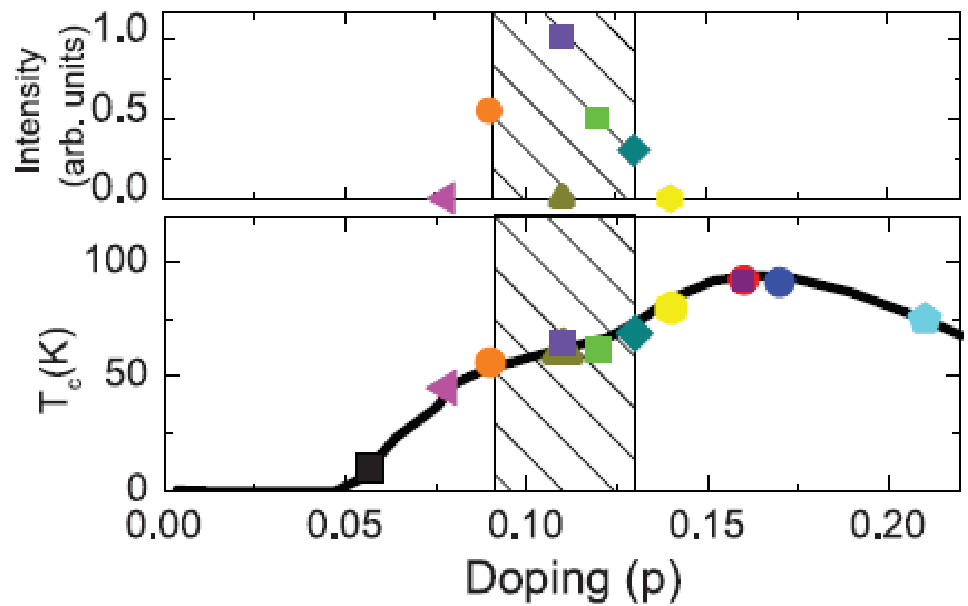
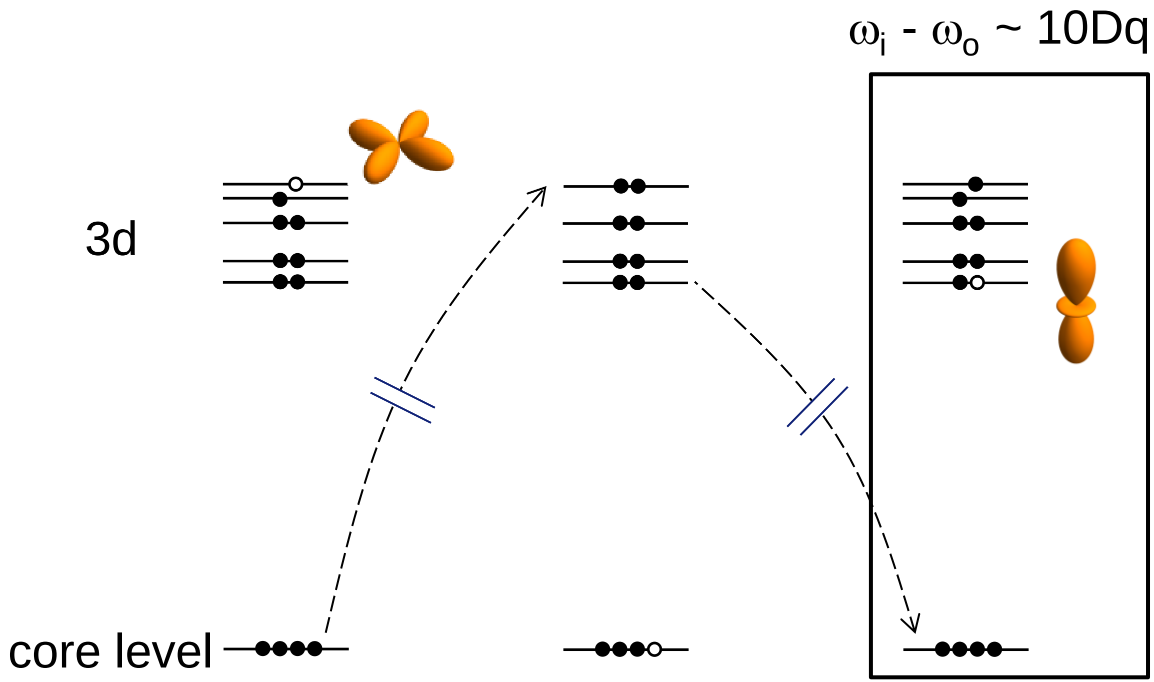




G. Ghiringhelli et al., Science 337, 821-825 (2012)

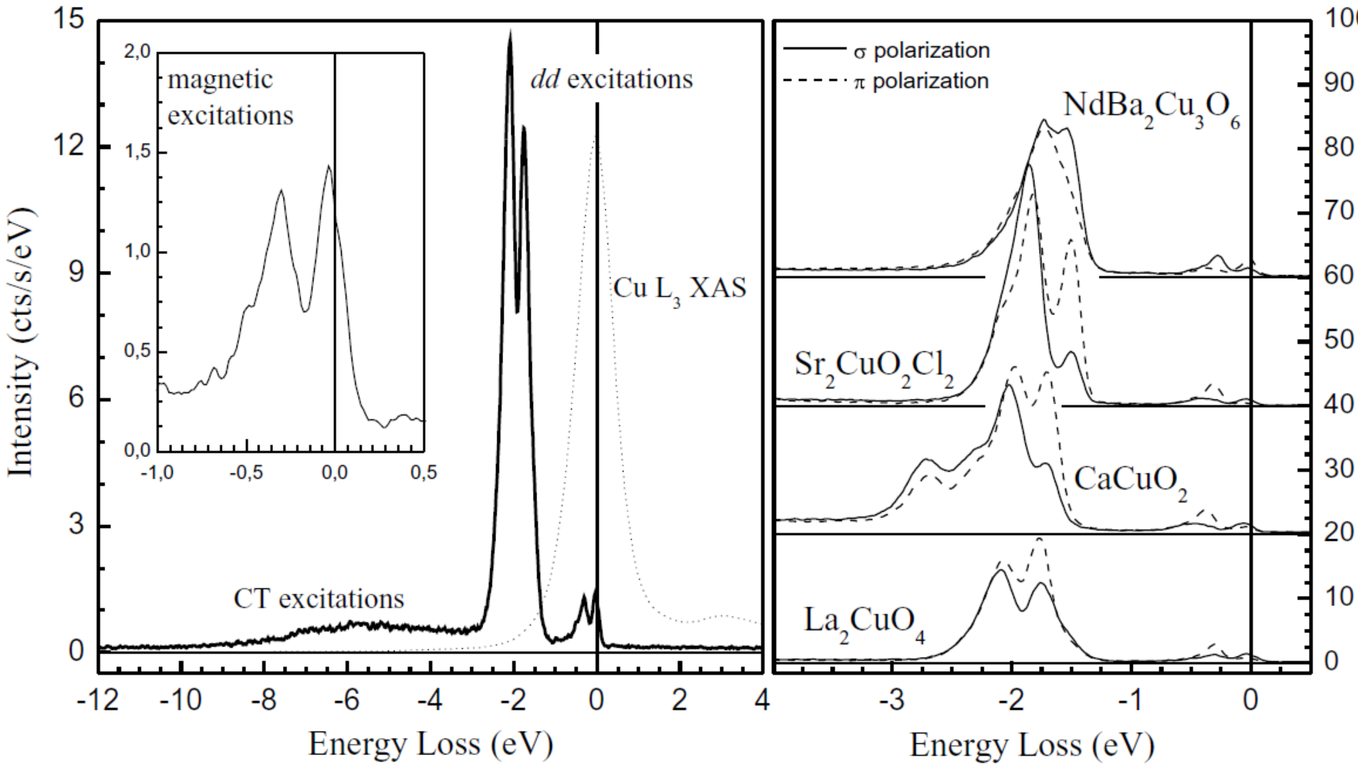
RIXS from a single Cu^{2+} ion

Orbital-flip is also a possible final state of the RIXS process



RIXS from a single Cu^{2+} ion

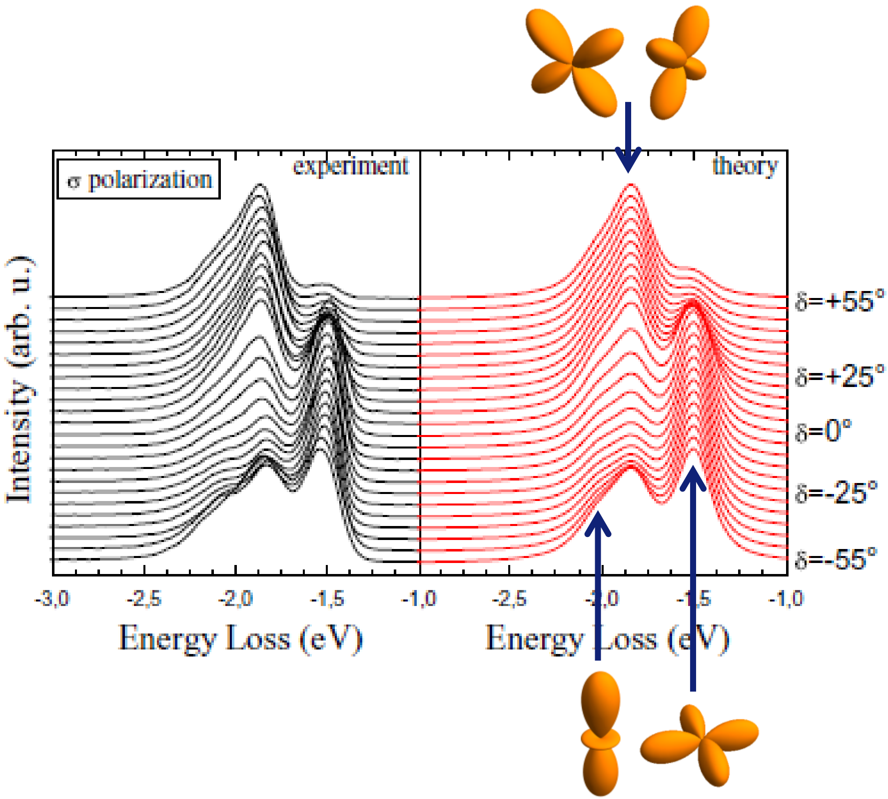
Excitations to crystal field states (d-d excitations)



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

RIXS from a single Cu^{2+} ion

Symmetry of the final states from their angular dependence



$$\mathcal{A}^{\epsilon\epsilon'}_{|\ \rangle} = \sum_n \frac{\langle\ |\mathcal{D}^{\dagger}_{\epsilon'}|n\rangle\langle n|\mathcal{D}_{\epsilon}|\ \rangle}{E_{\rm g} - E_n + \hbar\omega + \imath\Gamma_n}$$

	La_2CuO_4	$Sr_2CuO_2Cl_2$	$CaCuO_2$	$Sr_{0.5}Ca_{0.5}CuO_2$	$NdBa_2Cu_3O_7$
$E_{3z^2-r^2}$ ($\Gamma_{3z^2-r^2}$) (eV)	1.70 (0.14)	1.97 (0.10)	2.65 (0.12)	2.66	1.98 (0.18)
E_{xy} (Γ_{xy}) (eV)	1.80 (0.10)	1.50 (0.08)	1.64 (0.09)	1.56	1.52 (0.10)
$E_{xz/yz}$ ($\Gamma_{xz/yz}$) (eV)	2.12 (0.14)	1.84 (0.10)	1.95 (0.12)	1.93	1.75 (0.12)
ΔE_{e_g}	1.70	1.97	2.65	2.66	1.98
$\Delta E_{t_{2g}}$	0.32	0.33	0.31	0.36	0.23
$10Dq$ (eV)	1.80	1.50	1.64	1.56	1.52
D_s (eV)	0.29	0.33	0.42	0.43	0.32
D_t (eV)	0.11	0.13	0.19	0.19	0.14

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

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  - ✓ the case of $CaIrO_3$
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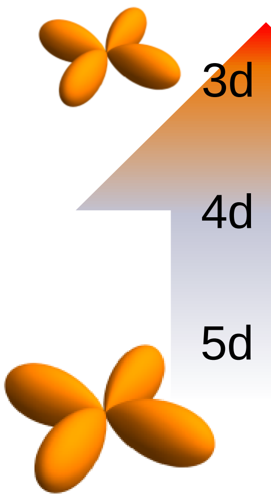
Introduction to the physics of iridates

Electronic correlation is strongest for 3d *TM*

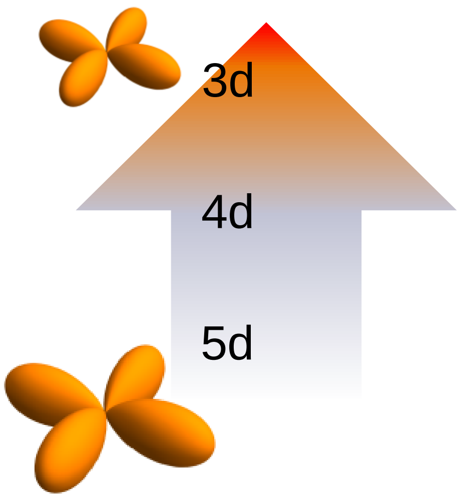


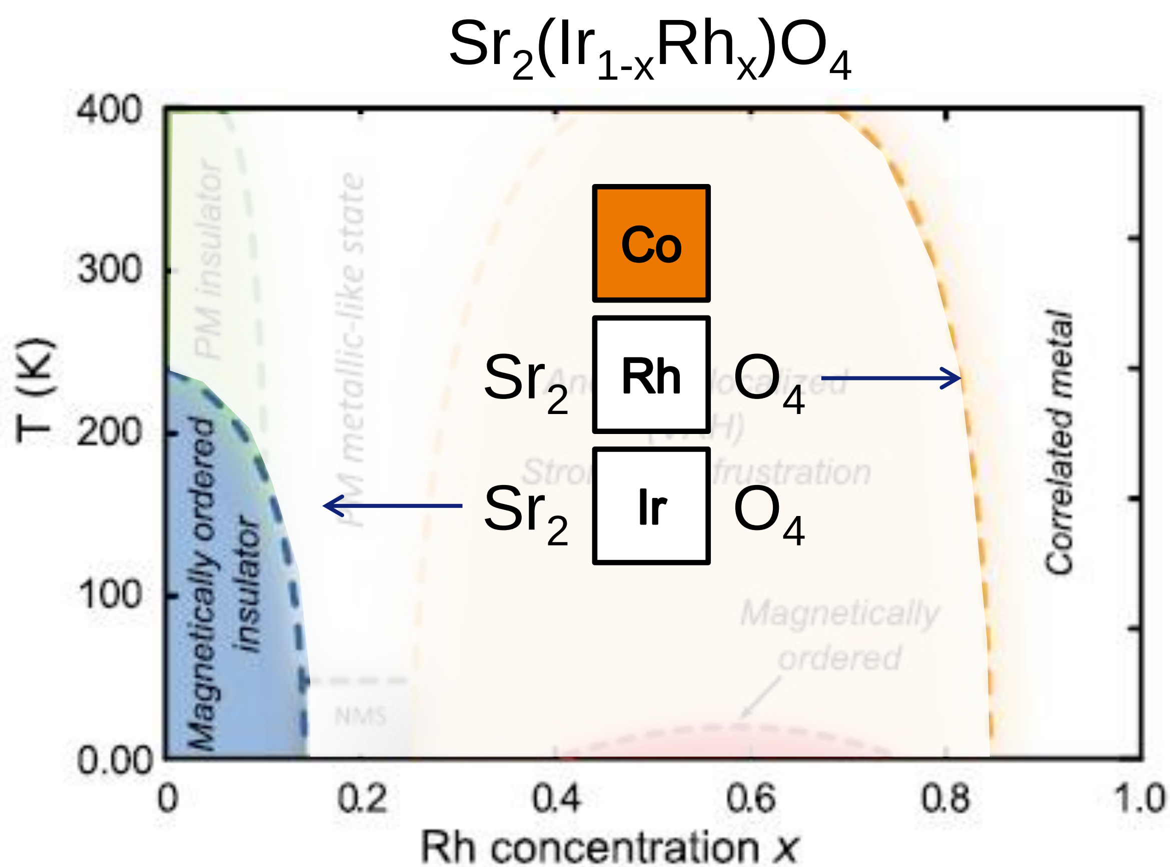
3d	Sc	**Ti**	**V**	**Cr**	**Mn**	**Fe**	**Co**	**Ni**	**Cu**	Zn
4d	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd
5d	Lu	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg

Ferroelectricity
Magnetoresistance
High temperature superconductivity
…

Introduction to the physics of iridates

However…

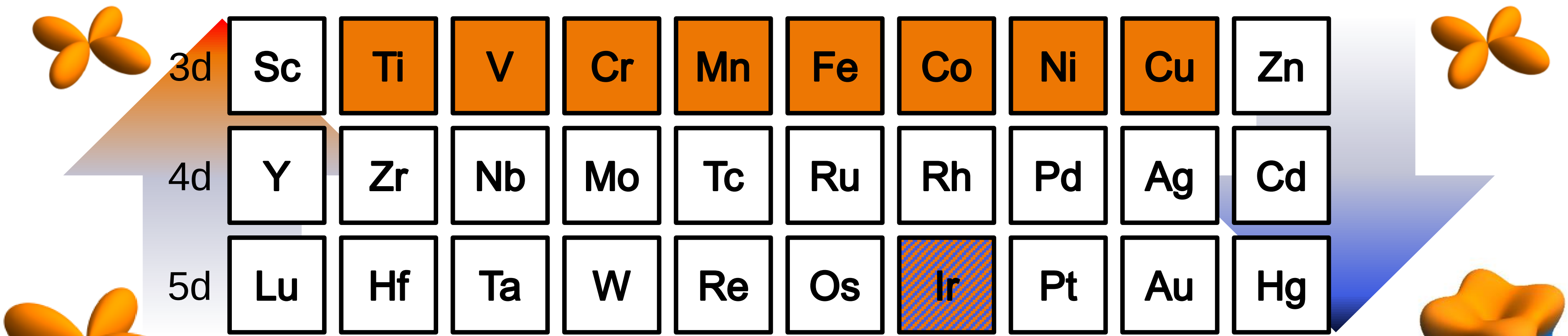




T. F. Qi et al., Phys. Rev. B, 86, 125105 (2012)

Introduction to the physics of iridates

…spin-orbit coupling is strongest for 5d *TM*

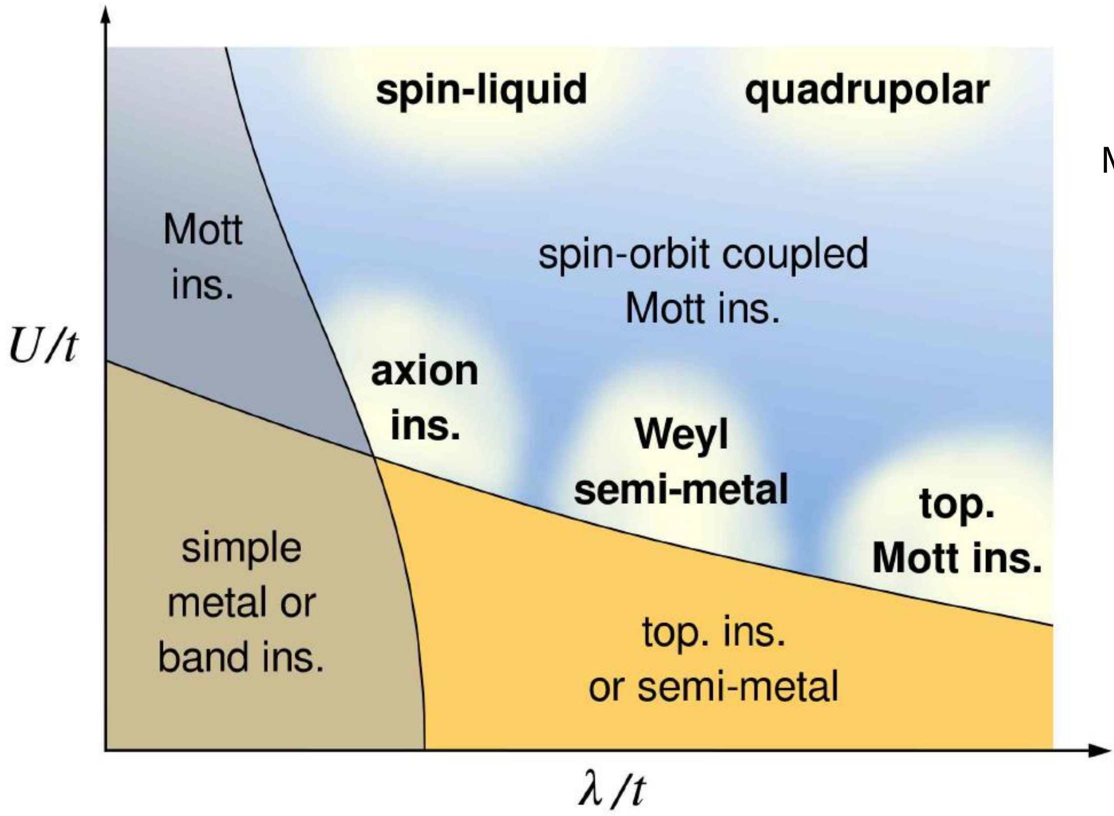


Novel quantum phases
Quantum spin-liquid
Unconventional superconductivity?

Spin-orbit coupling

Introduction to the physics of iridates

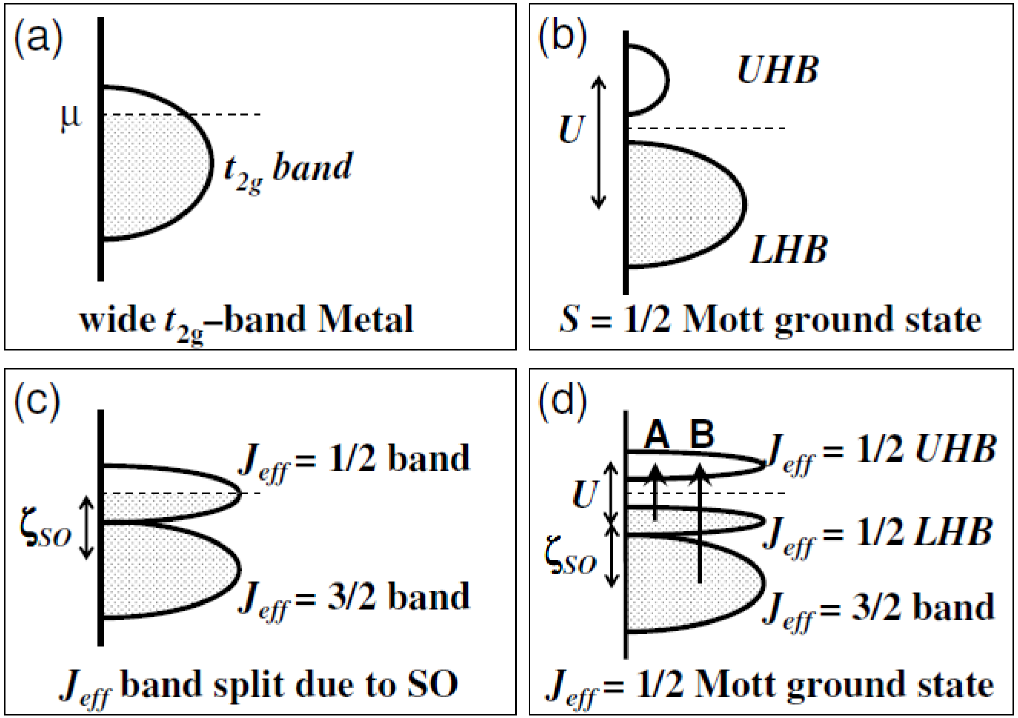
Spin-orbit coupling renormalizes the effect of electron correlation

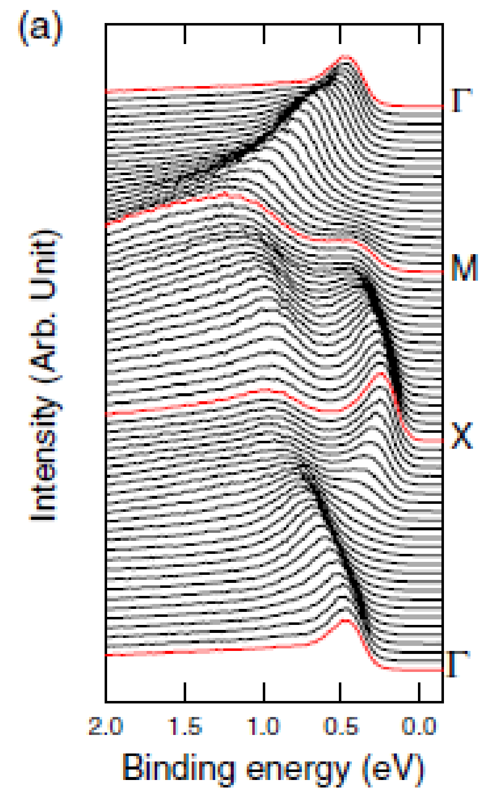


W. Witczak-Krempa et al., Annual Review of Condensed Matter Physics 5, 57-82 (2014)

Introduction to the physics of iridates

Spin-orbit induced Mott insulating state in Sr_2IrO_4



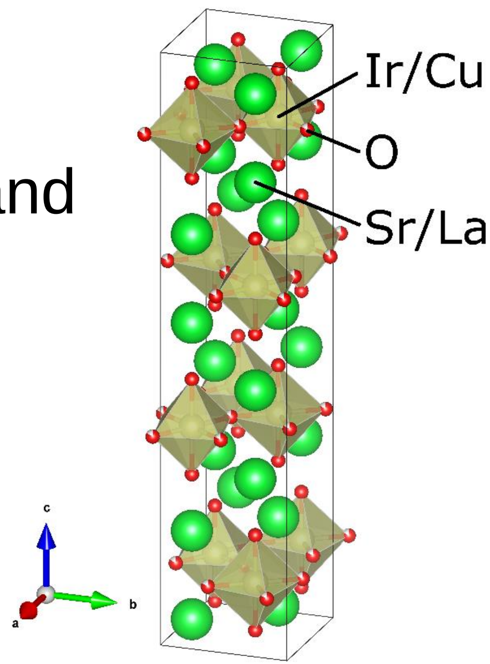


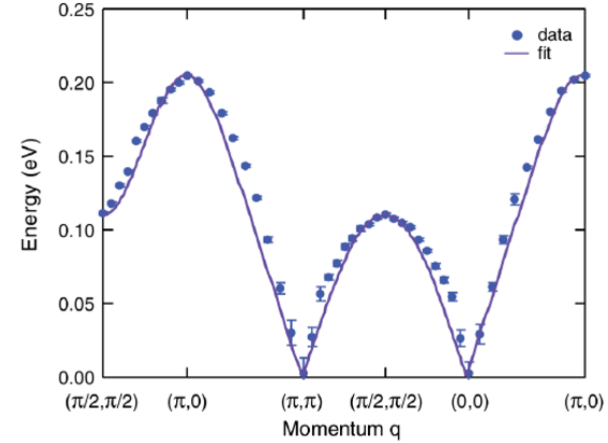
B.J. Kim et al., Phys. Rev. Lett., 101, 076402 (2008)

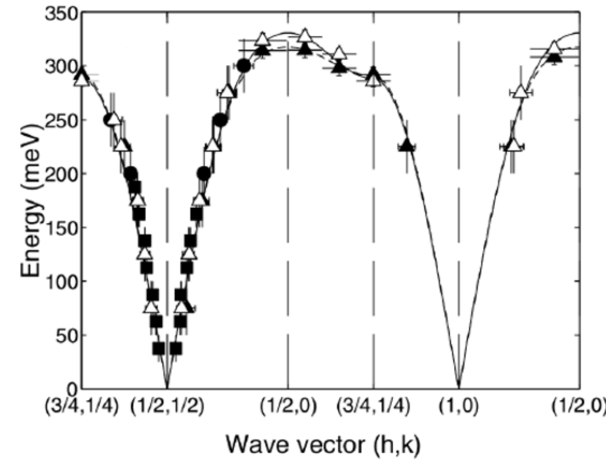
Introduction to the physics of iridates

Sr_2IrO_4 is characterized by a cuprate-like behaviour

- Mott-insulating phase
  - ✓ crystal structure
  - ✓ electronic structure
  - ✓ magnetic structure and dynamics







R. Coldea et al., Phys. Rev. Lett. 86, 5377 (2001)
J. Kim et al., Phys. Rev. Lett. 108, 177003 (2012)

Introduction to the physics of iridates

Sr_2IrO_4 is characterized by a cuprate-like behaviour

- Mott-insulating phase
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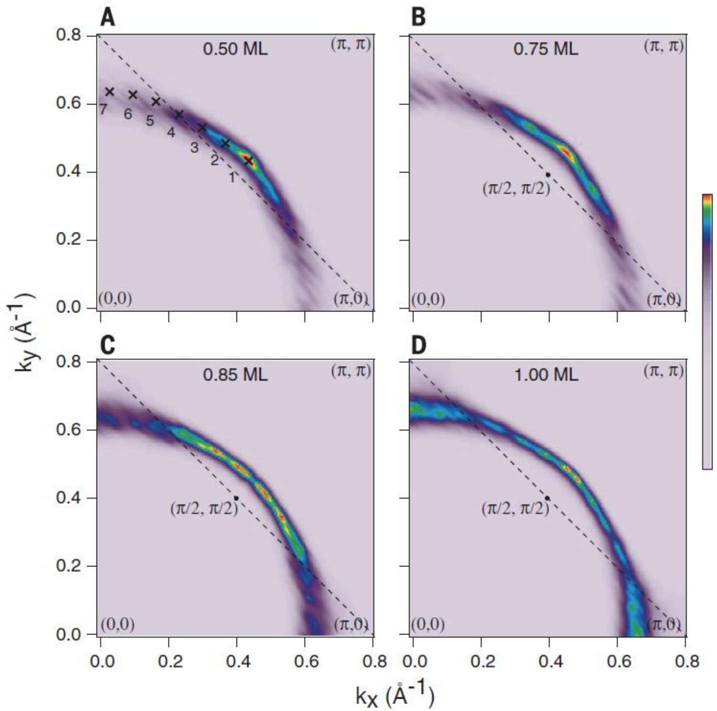
  - ✓ electronic structure
  - ✓ magnetic structure and dynamics
- Doped metallic phase
  - ✓ “fermiology”



Y. K. Kim et al., Science 345, 187-190 (2014)
Y. K. Kim et al., Nature Physics (2015)

Introduction to the physics of iridates

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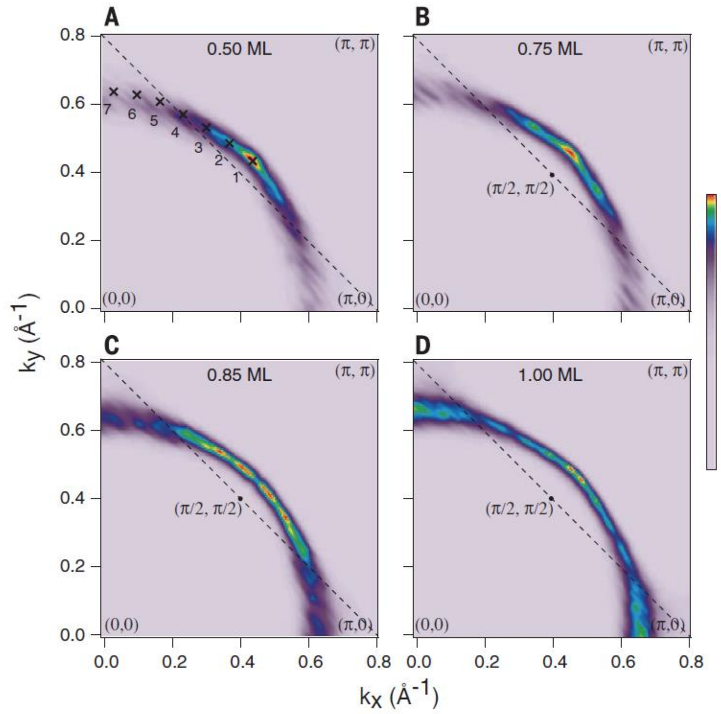
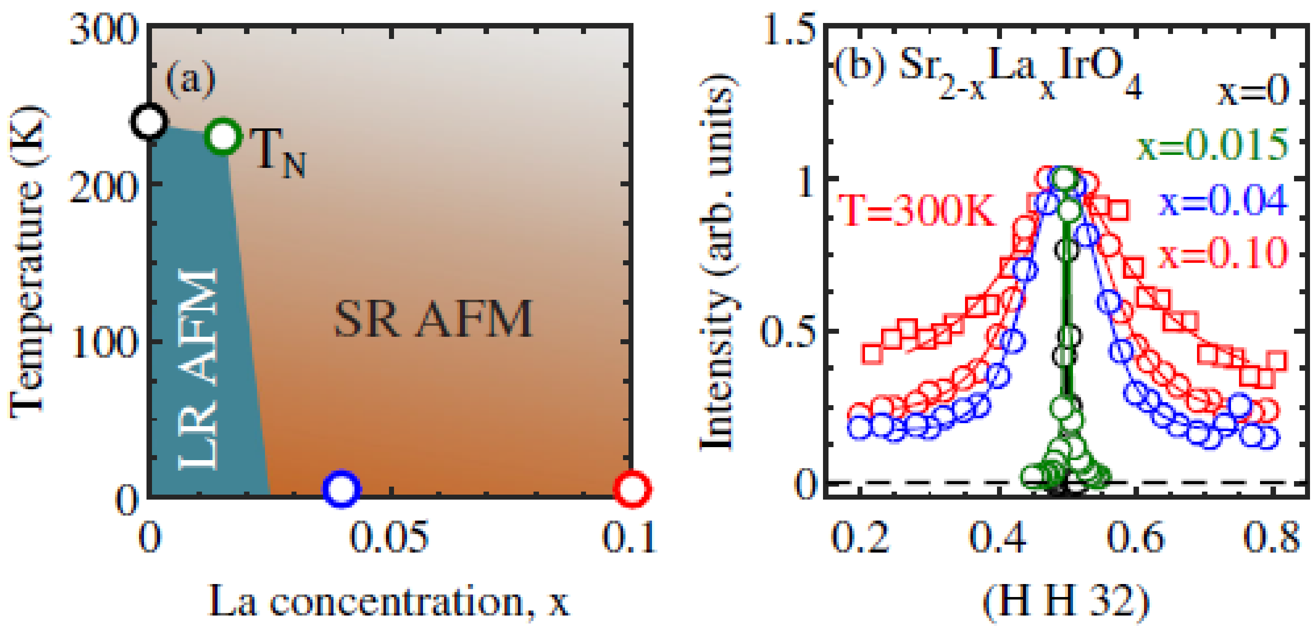
  - ✓ magnetic structure and dynamics
- Doped metallic phase
  - ✓ "fermiology"
  - ✓ magnetic dynamics?
  - ✓ superconducting?



Y. K. Kim et al., Science 345, 187-190 (2014)
Y. K. Kim et al., Nature Physics (2015)

Magnetic excitations in pure and doped Sr_2IrO_4

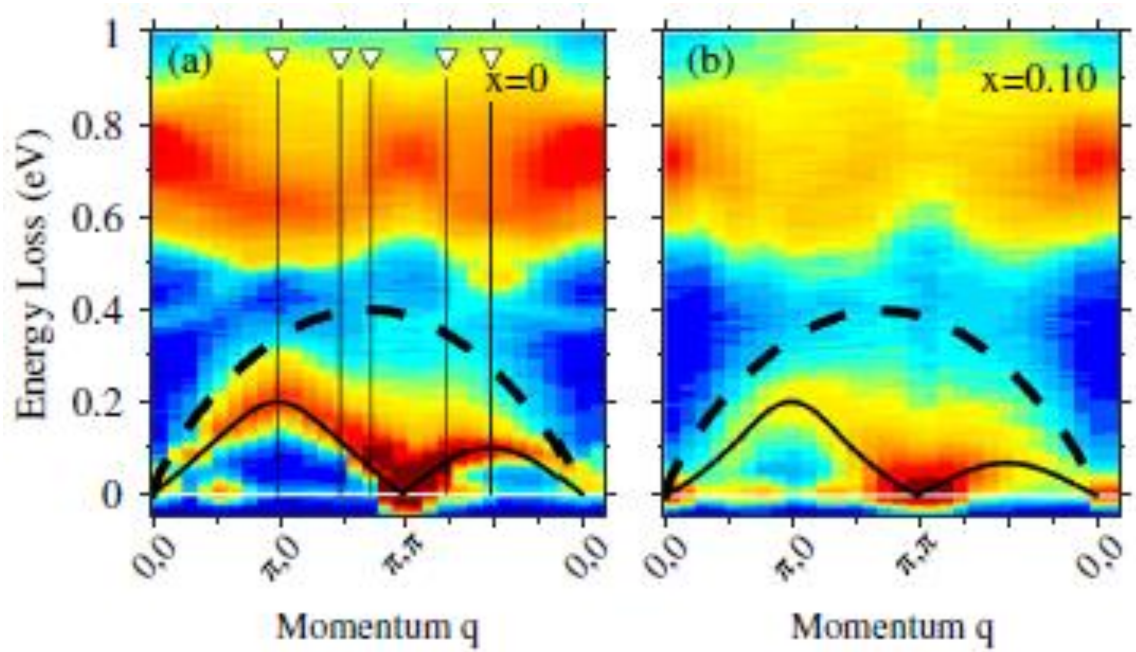
Sr_2IrO_4 probably the best system to look for superconductivity



H. Gretarsson et al., Phys. Rev. Lett. 117, 107001 (2016)

Magnetic excitations in pure and doped Sr_2IrO_4

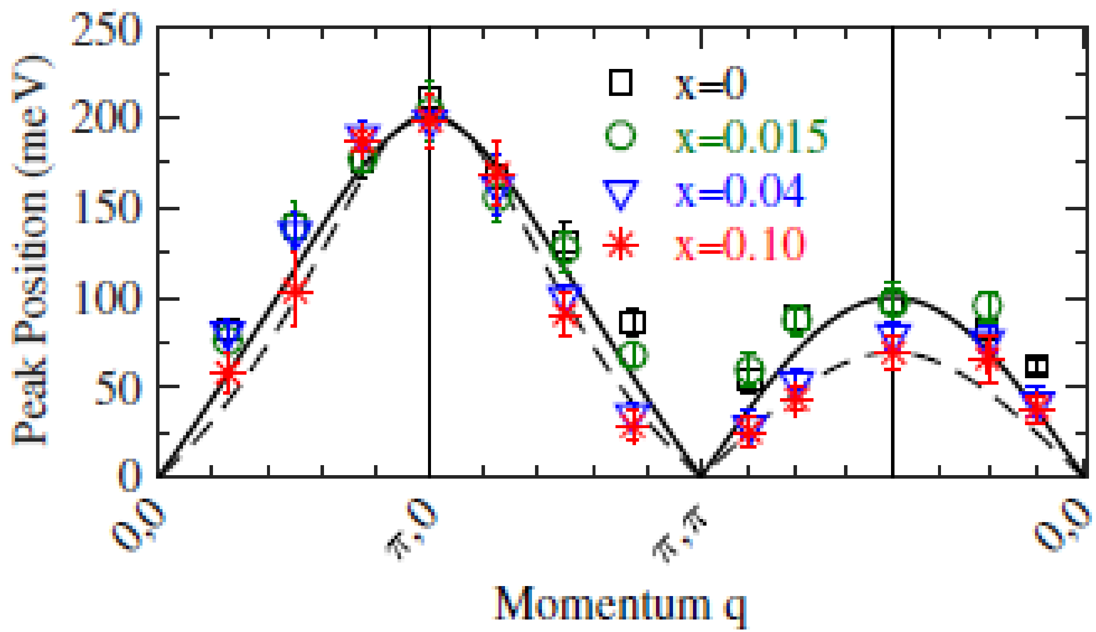
Persistence of magnetic excitations in the doped phase described by a $j_{\mathrm{eff}} = 1/2$ Heisenberg model



H. Gretarsson et al., Phys. Rev. Lett. 117, 107001 (2016)

Magnetic excitations in pure and doped Sr_2IrO_4

Damping and softening at (π/2,π/2) like in hole-doped cuprates



Magnetic excitations in pure and doped Sr_2IrO_4

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  - ✓ magnetic structure and dynamics
- Doped metallic phase
  - ✓ “fermiology”
  - ✓ magnetic dynamics
  - ✓ superconducting?

If not, why?

Magnetic excitations in pure and doped Sr_2IrO_4

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  - ✓ electronic structure
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- Doped metallic phase
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  - ✓ magnetic dynamics
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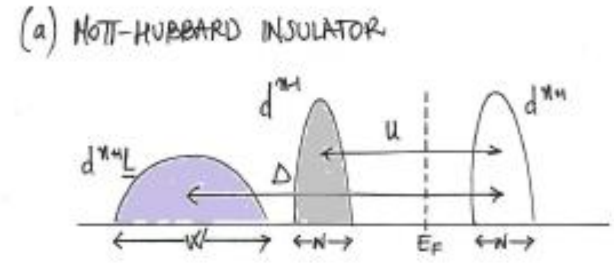
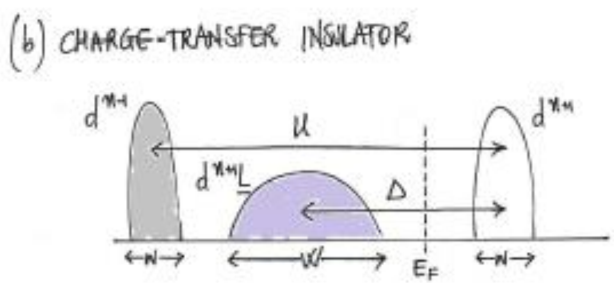
If not, why?

Pseudo-spins vs. spins
No charge ordering (so far)
Mott-Hubbard vs. charge transfer ins.



Sr_2IrO_4



La_2CuO_4

Conclusions and acknowledgements

RIXS probes spin, orbital (and lattice) degrees of freedom

Lot of things can be understood at the single ion level

Orbital excitations: symmetry of the ground and excited states

Magnetic excitations: persistence in the metallic phase of both cuprates and iridates

Conclusions and acknowledgements

Politecnico di Milano

- L. Braicovich
- G. Ghiringhelli

ESRF, Grenoble

- Nick Brookes
- Giulio Monaco
- Michael Krisch
- Matteo Rossi

Various collaborators

- D. McMorrow, S. Boseggia (UCL)
- J. Van den Brink (IFW, Dresden)
- K. Wohlfeld (Warsaw Univ.)
- B. J. Kim, H. Gretarsson, M. Le Tacon (MPI, Stuttgart)
- A. Gubanov (Russian Academy of Sciences)
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Thank you for your attention!