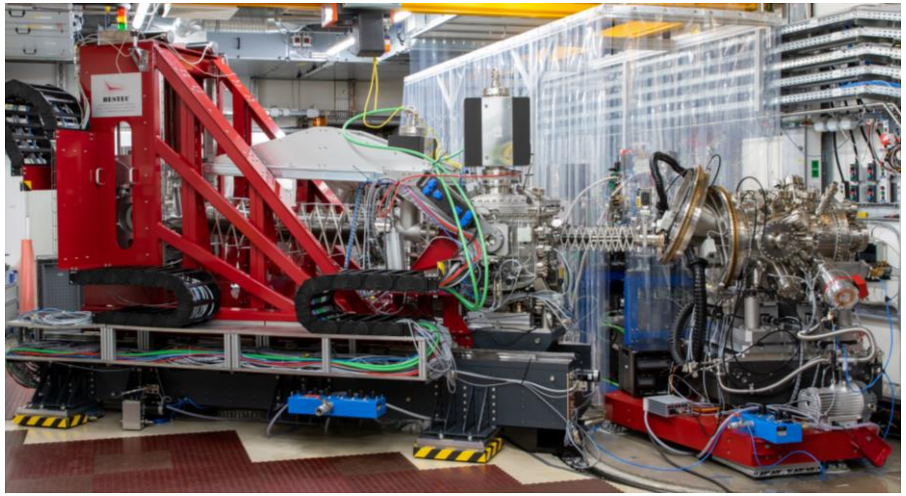
hRIXS@SCS: first commissioning results from cuprate and nickelate Mott insulator samples

January 26, 2022

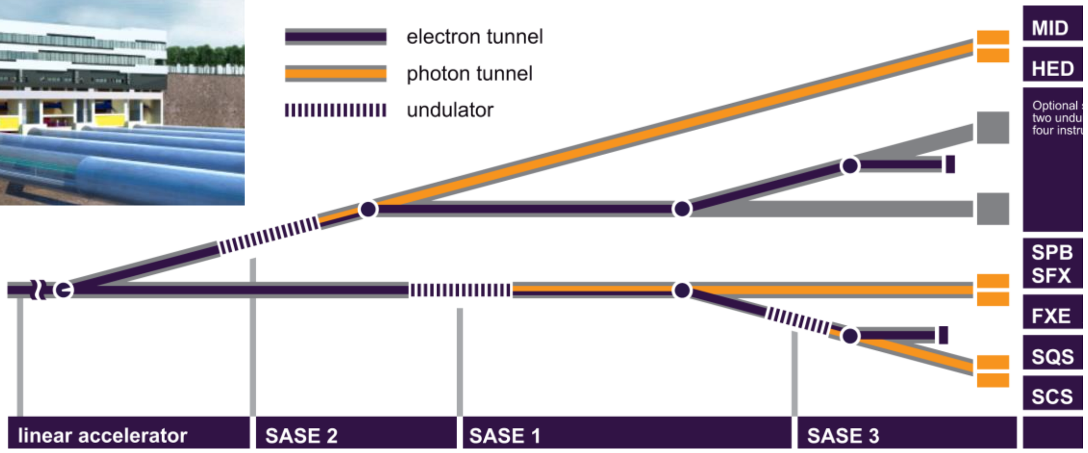
Justine Schlappa
SCS instrument, European XFEL

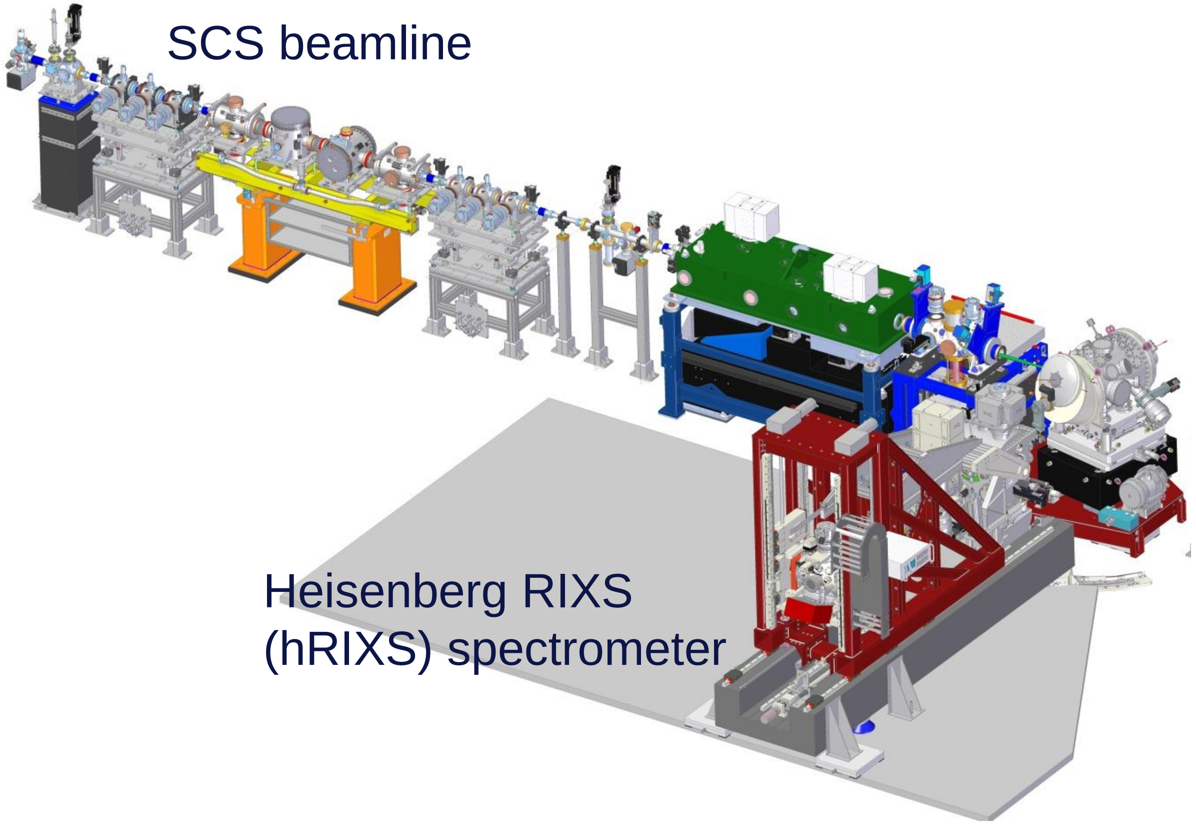


Scope:

- hRIXS @ SCS: motivation and overview
- Results from hRIXS commissioning

SCS Instrument & SASE3, European XFEL



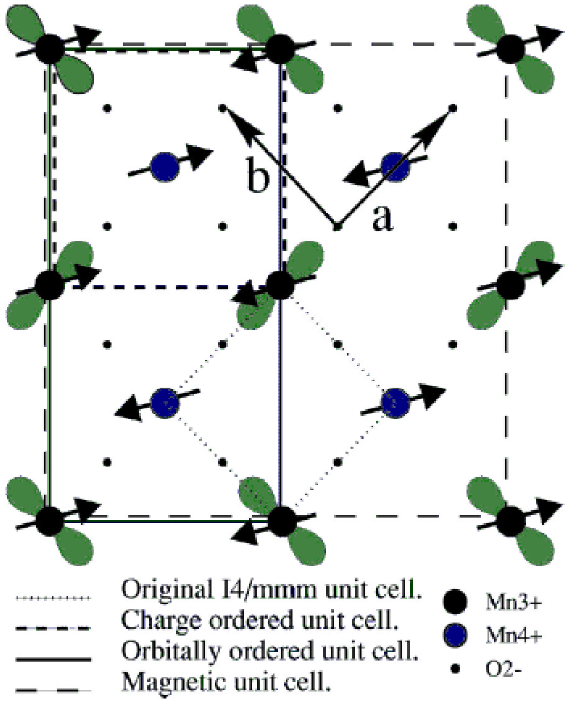


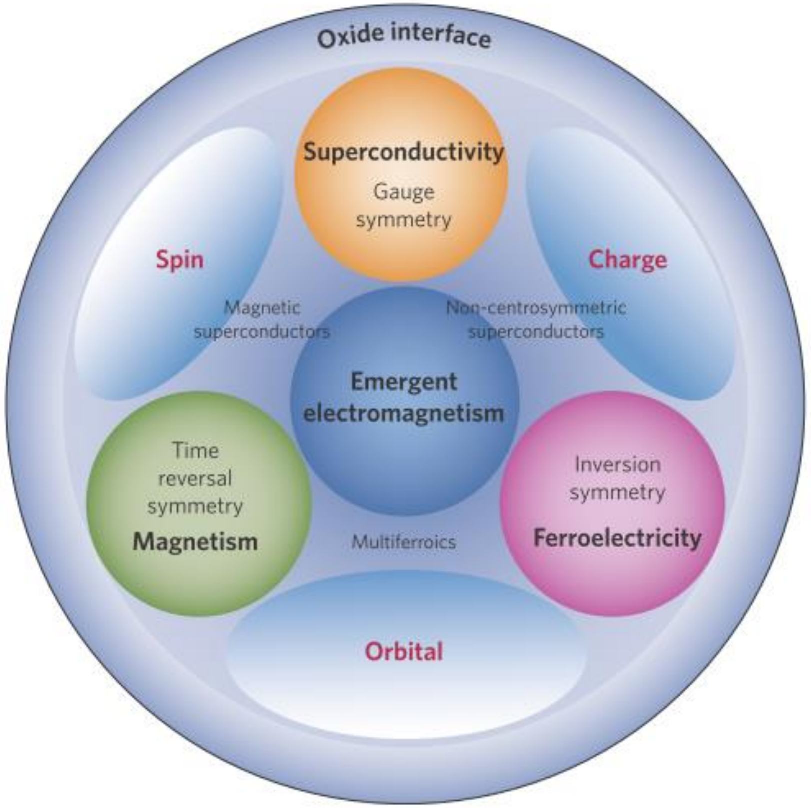
Spectroscopy and Coherent Scattering (SCS):

- Soft x-ray beamline
- Time-resolved/ non-linear x-ray spectroscopies
- Time-resolved/ non-linear x-ray diffraction
- Forward- / small-angle scattering geometries
- Reflection- / backscattering geometries
- RIXS
- Solid samples
- Liquid-jet samples

Physics of complex materials by nuclear, charge, spin, and orbital degrees of freedom and their interplay

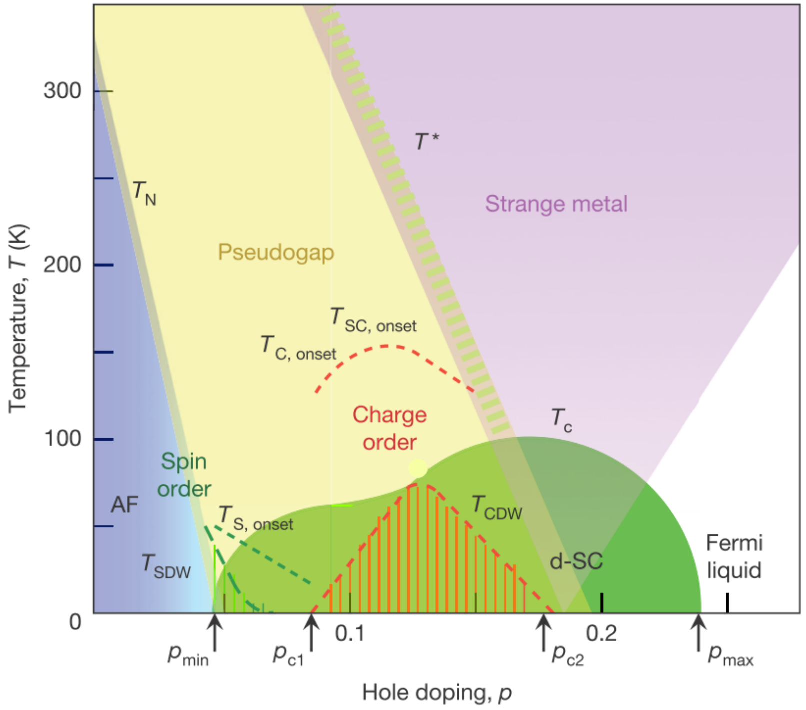
C.W.M. Castleton and M Altarelli,
Phys. Rev. B 62, 1033 (2000)





H.Y. Hwang et al.,
Nature materials 11, 103 (2012)

B. Keimer et al.,
Nature 518, 179 (2015)

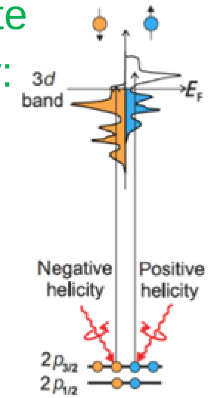


Microscopic probes

Synchrotrons:

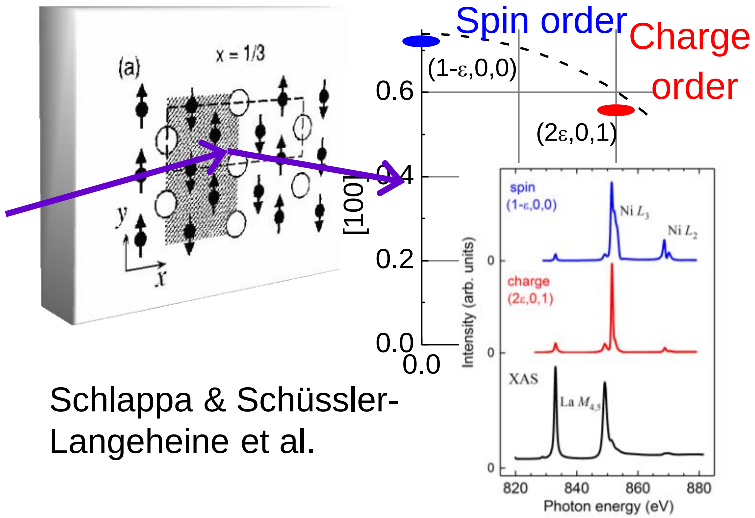
- **See the ultra small**

Atomic-site selectivity:



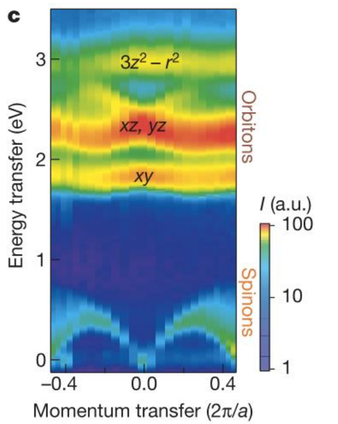
Stöhr & Siegmann, Magnetism Springer

Length scales of nm:



Schlappa & Schüssler-Langeheine et al.

Inter-particle correlations:

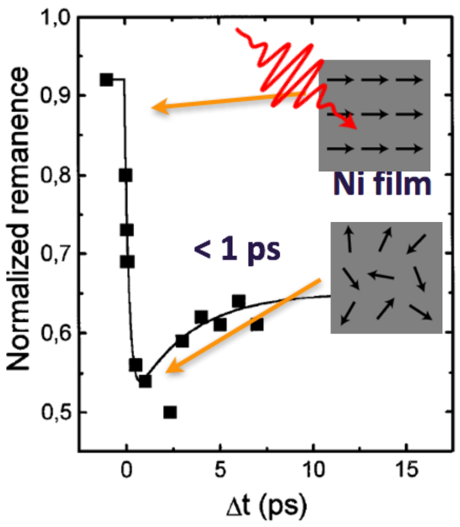


Schlappa et al., Nature (2012)

Optical Lasers:

- **See the ultra fast**

Time scales of sub-ps:



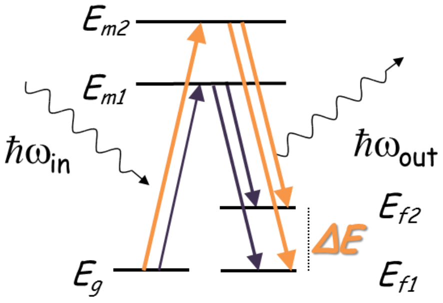
Beaurepaire et al. PRL (1996)

X-ray Lasers:
- **See the ultra small**
- **See the ultra fast**

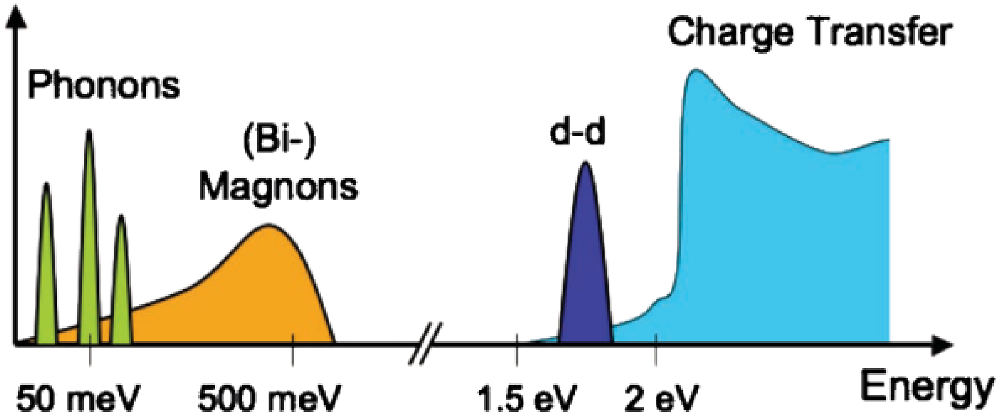
Resonant inelastic x-ray scattering (RIXS)

- Two-photon scattering process
- Inelastic process: energy transfer from photons on the sample
- Resonance: selective excitation (spectroscopy)
- Momentum transfer from the photons on the sample → study of collective excitations or quasi particles

Energy conservation



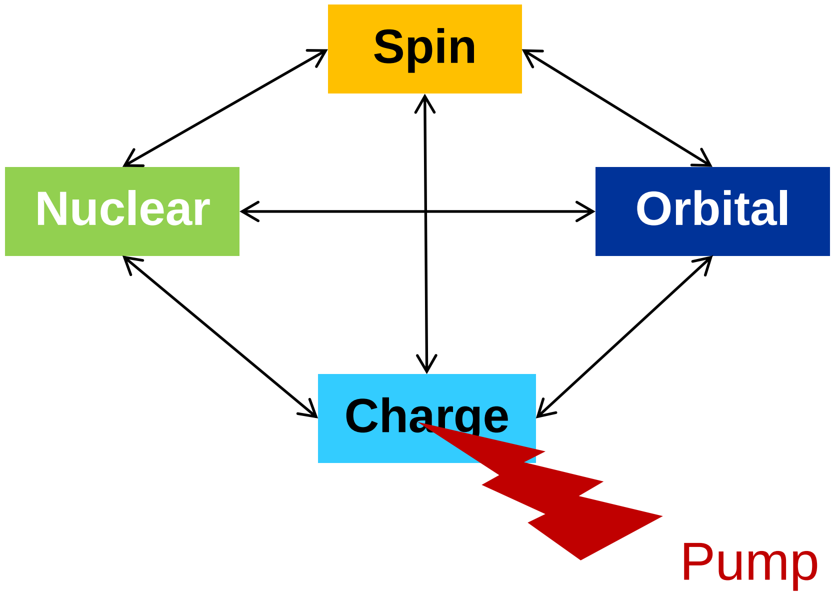
RIXS:



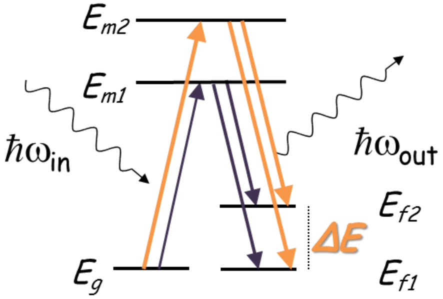
Rev. Mod. Phys. 83, 705 (2011).

Resonant inelastic x-ray scattering (RIXS)

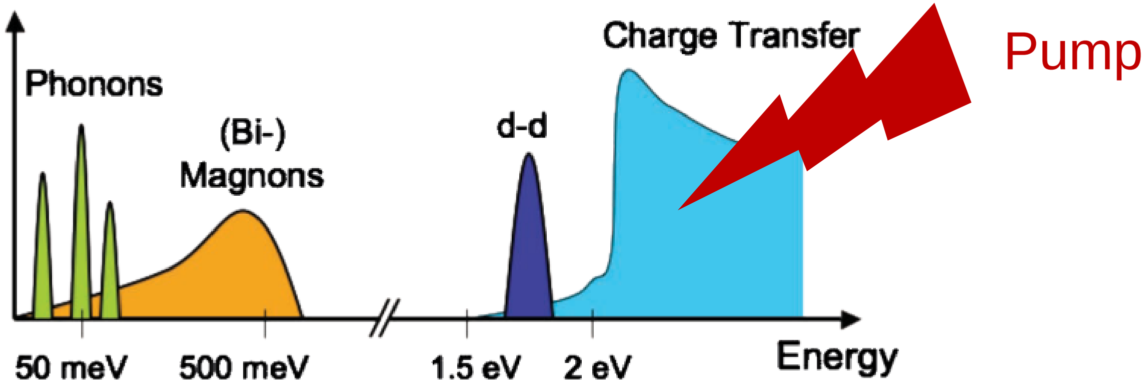
- Two-photon scattering process
- Momentum transfer from the photons on the sample
- Inelastic process: energy transfer from photons on the sample



Energy conservation

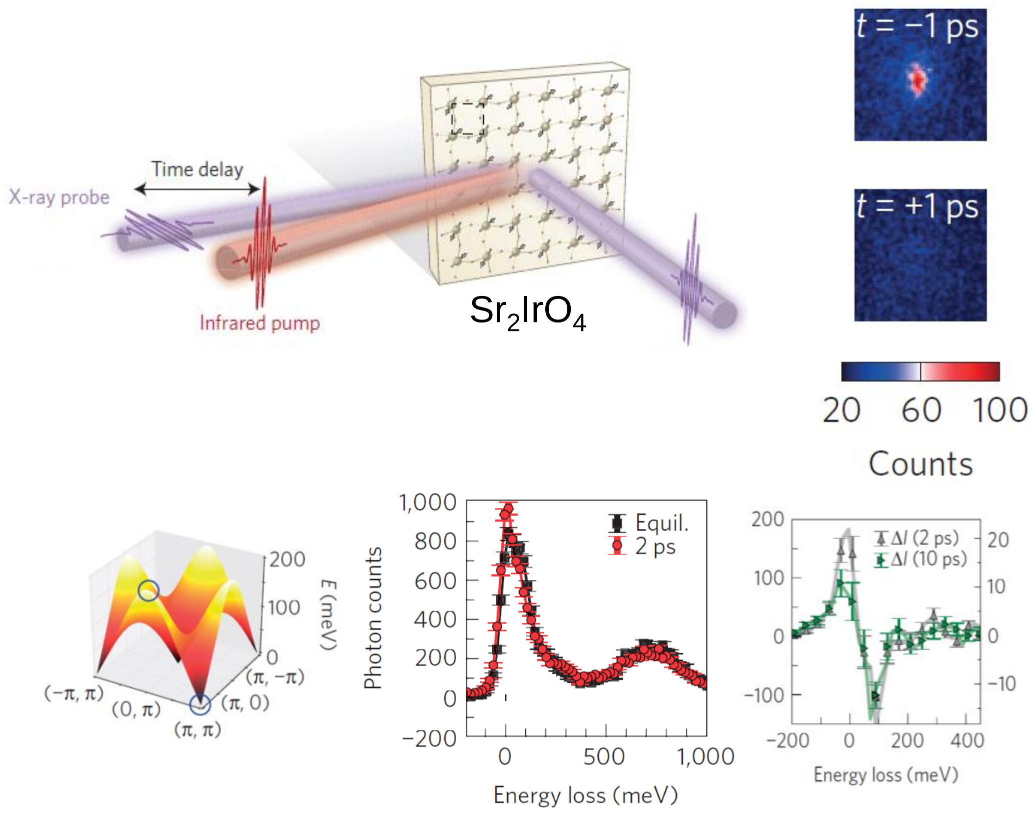


time-resolved RIXS:



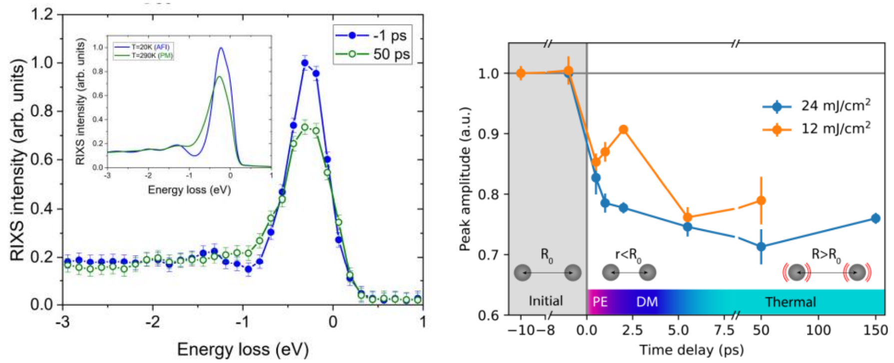
Scientific motivation: time-resolved RIXS in photoexcited complex materials

Magnetic correlation dynamics in Sr_2IrO_4:



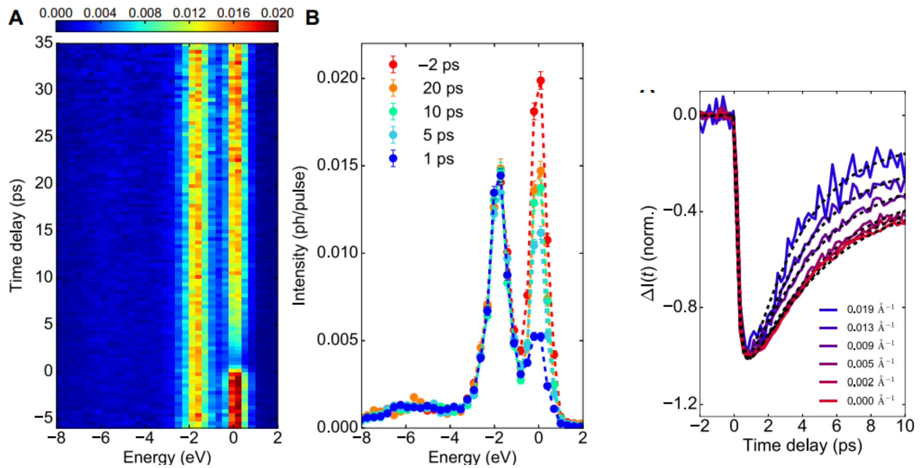
M. Dean et al., Nature Mat 15, 601 (2016).

Orbital dynamics through insulator to metal transition in V_2O_3:



S. Parchenko et al., Phys. Rev. Research 2, 023110 (2020).

Charge dynamics in $La_{1.875}Ba_{0.125}CuO_4$:

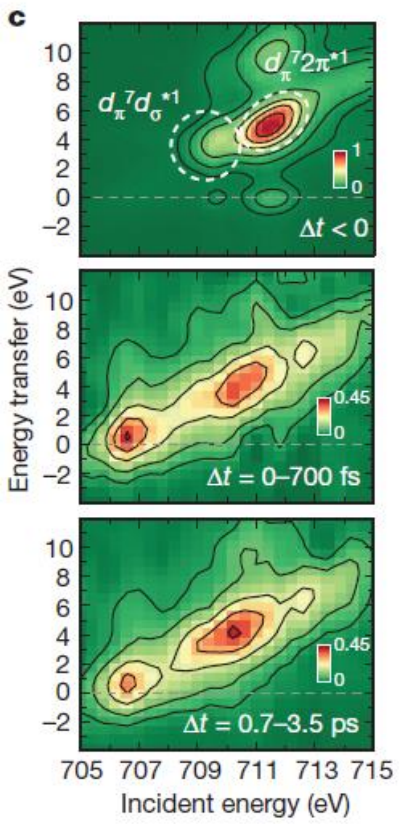


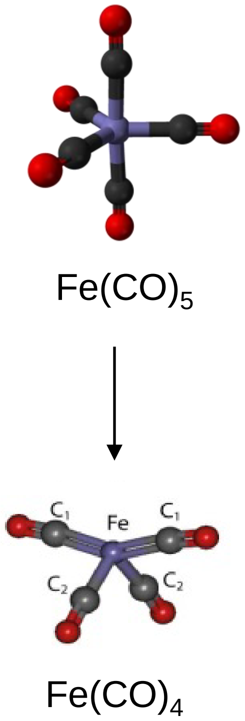
M. Mitrano et al., Sci. Adv. 2, eaax3346 (2019).

Scientific motivation: time-resolved RIXS in chemical systems

Metal L-edge RIXS spectroscopy gives a direct measure of the d-d excitations which report on the chemically active electrons

Bond Breaking and Formation

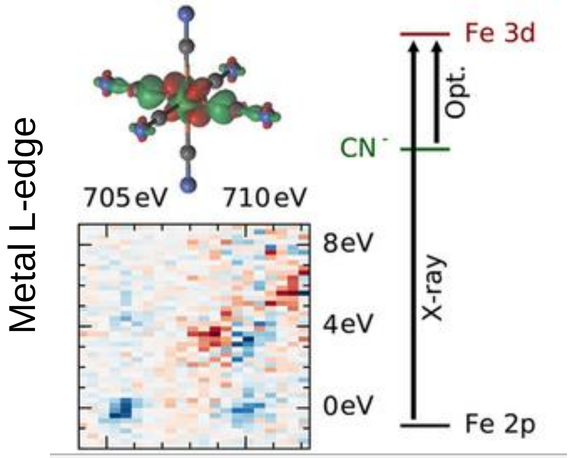


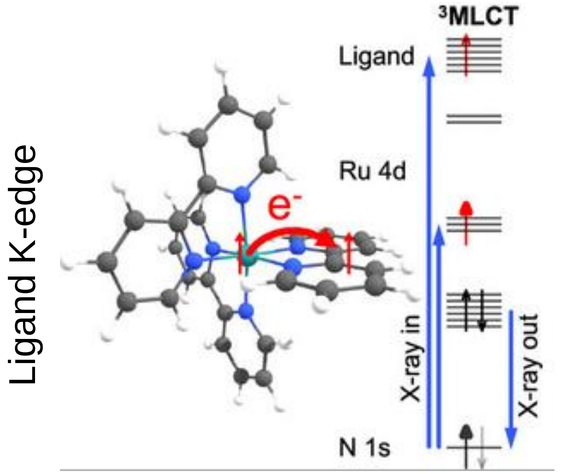


P. Wernet et al. Nature 520, 78 (2015)

Presentation by
B. van Kuiken, SCS
Satellite Workshop

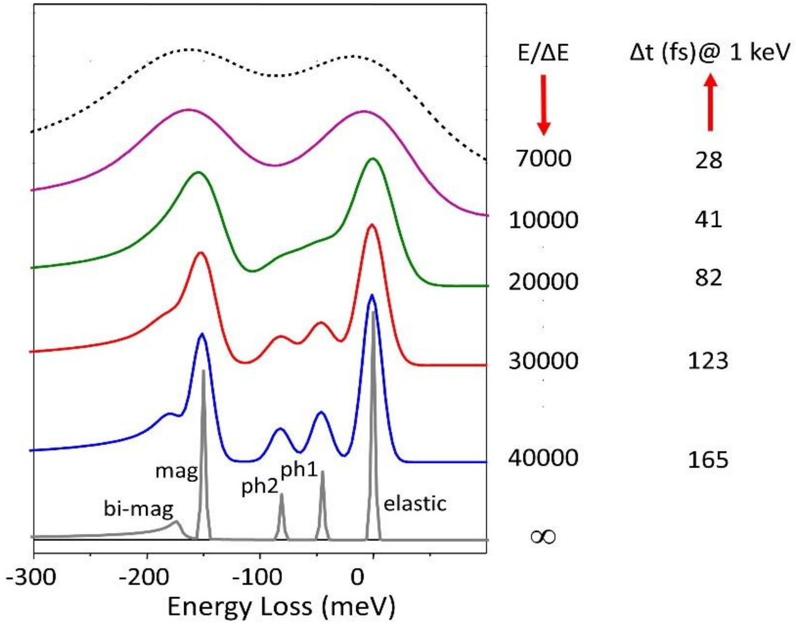
Charge Transfer Reactions

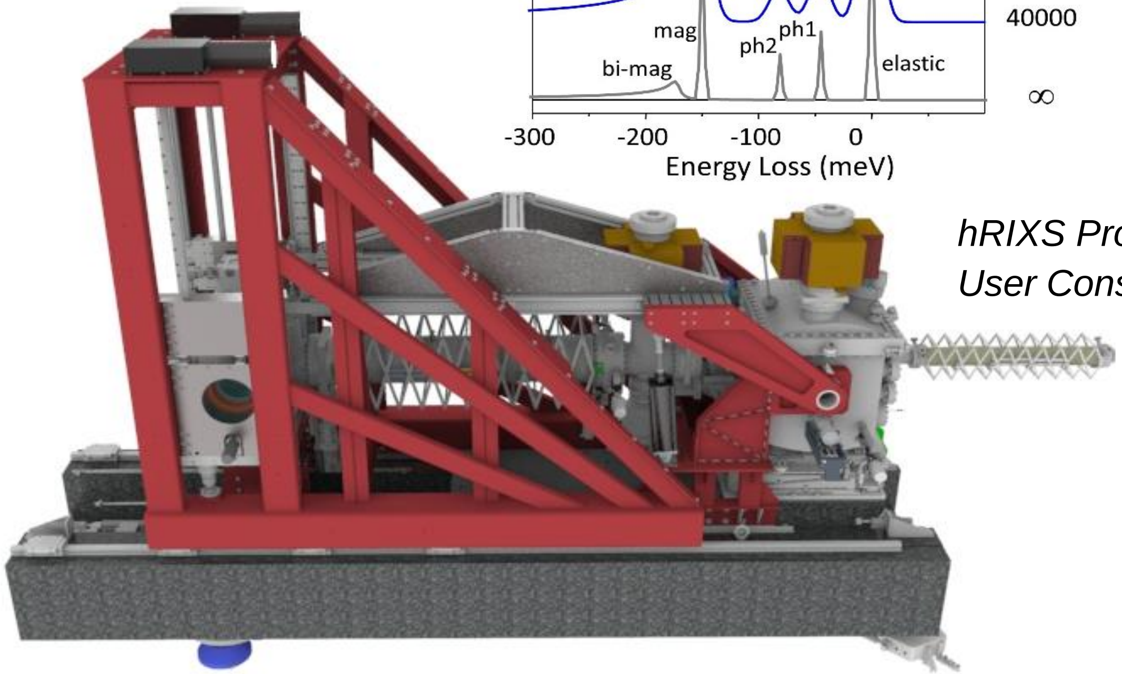




Jay *et al. J. Phys. Chem. Lett 9, 3538 (2018)*
Jay *et al. J. Phys. Chem. Lett.* 12, 6676 (2021)

Heisenberg RIXS User Consortium instrumentation (hRIXS) @ SCS



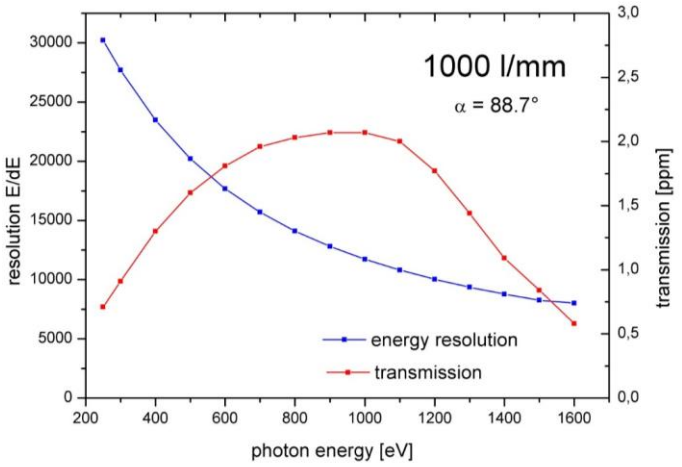


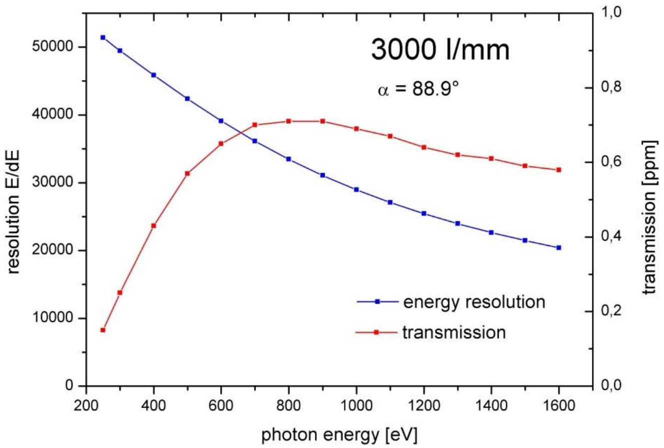
hRIXS Proposal, User Consortium

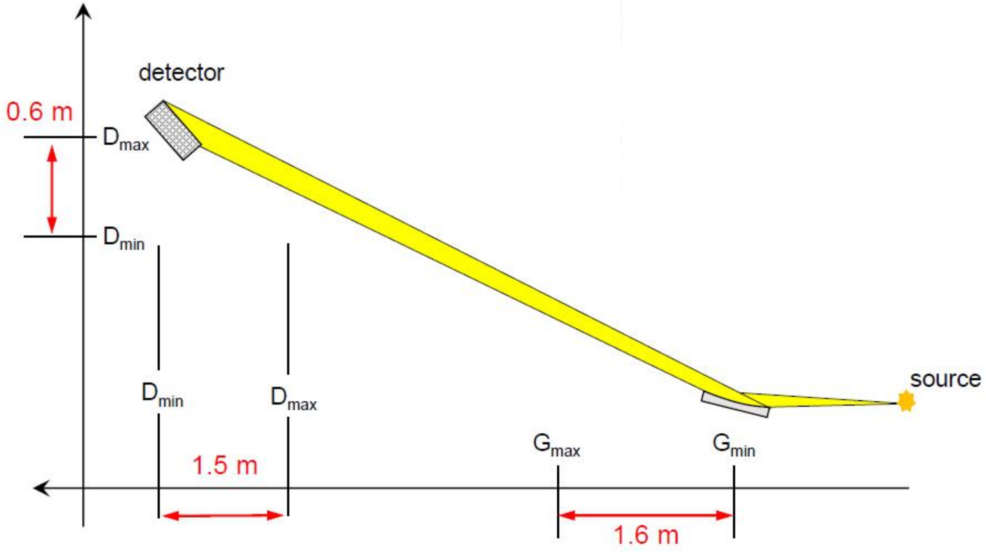
- time-resolved, soft x-ray Resonant Inelastic X-ray Scattering
- tr-RIXS close to the transfer limit
  - High resolving power (≥ 10,000)
  - High temporal resolution (below 100 fs)
- Variable momentum transfer
- UHV sample environment
- Liquid-jet sample environment

First high-performance instrument for RIXS at an FEL

hRIXS User Consortium spectrometer



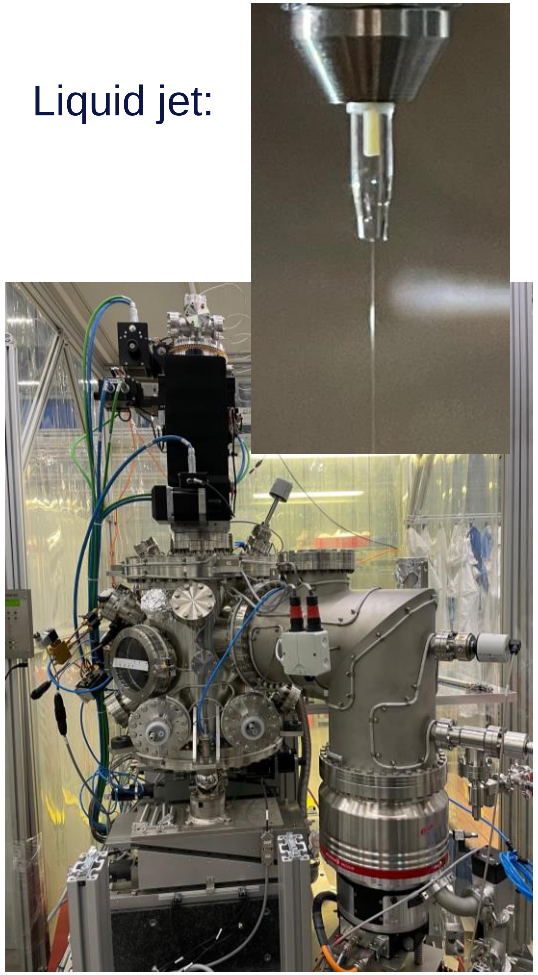




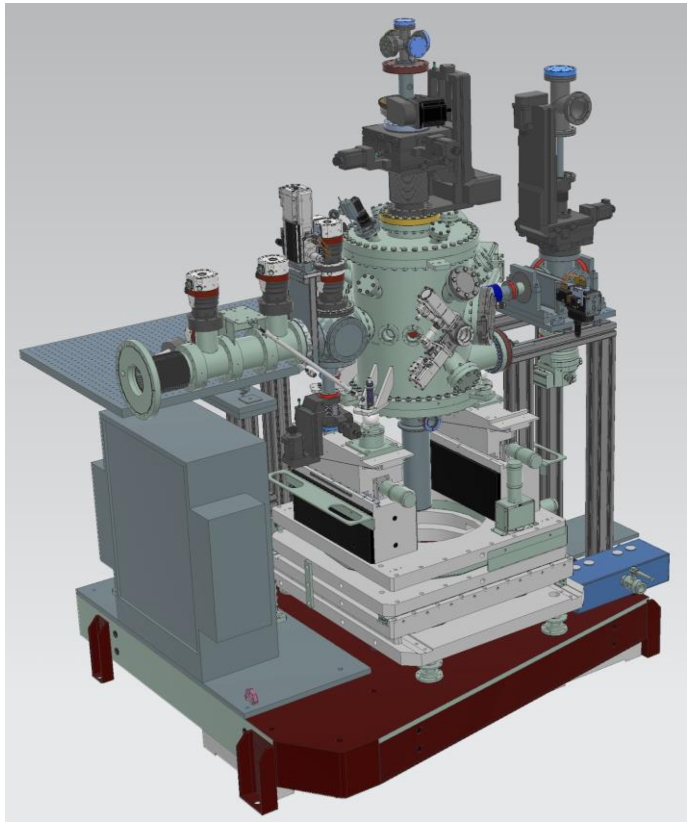
- Single optical element, spherical VLS
  → ease of alignment
- Large working range with a single grating: 270 – 1500 eV
- three grating slots, two occupied: high-resolution and high-transmission grating

Chemistry Chamber (CHEM) by the hRIXS UC

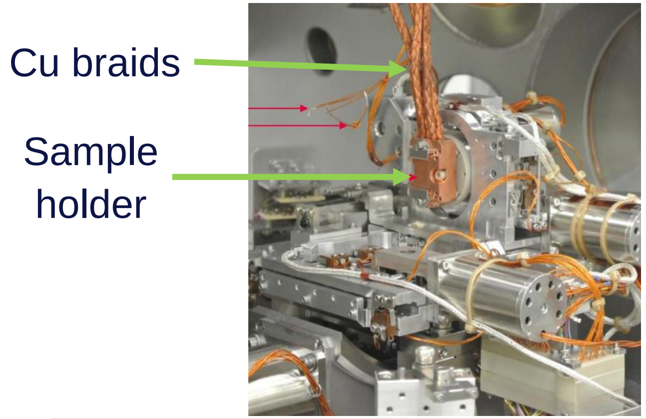
Liquid jet:

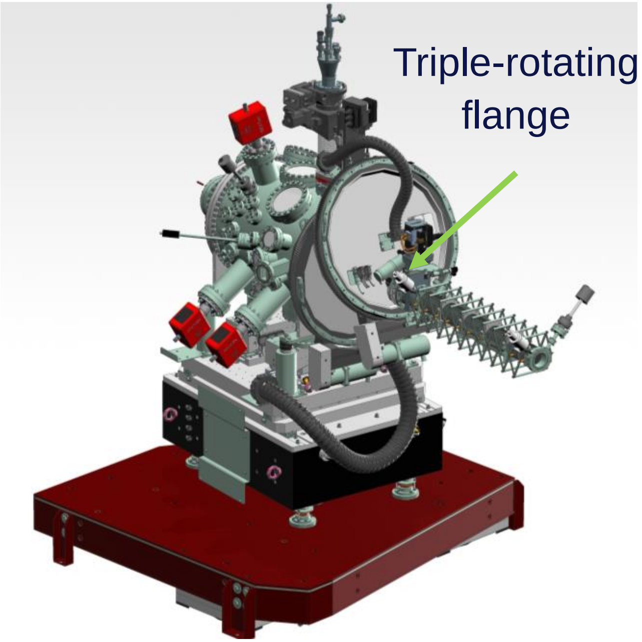


- Time-resolved RIXS studies of chemical systems
- Solid sample holder
- Liquid-jet: cylindrical, diameter 20 – 50 µm
- Running jet for bio-chemical relevant solvents, i.e. liquid water, ethanol, iso-propanol ~10^{-3} mbar
- Renewable sample, up to MHz repetition rate
- Switching channel device for up to 6 liquid samples
- Three-step differential pumping stage, pressure difference ~10^{6} mbar
- XAS in TFY

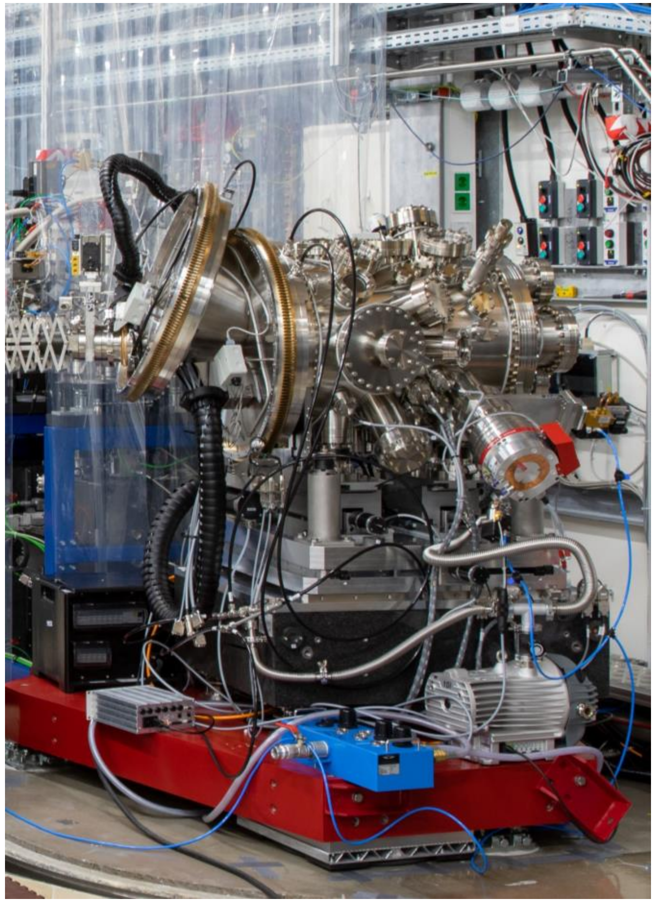


XRD setup for solid samples: baseline SCS



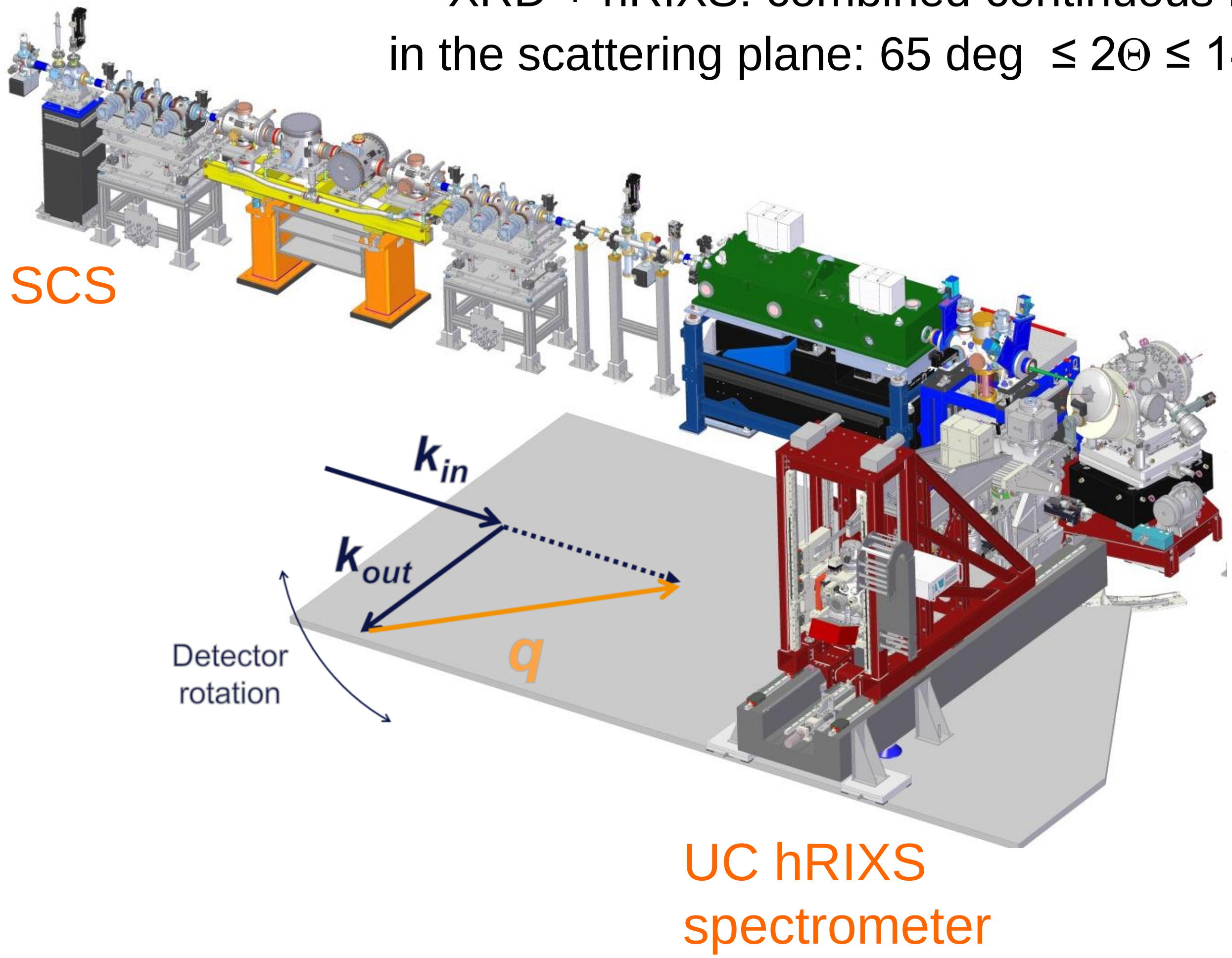


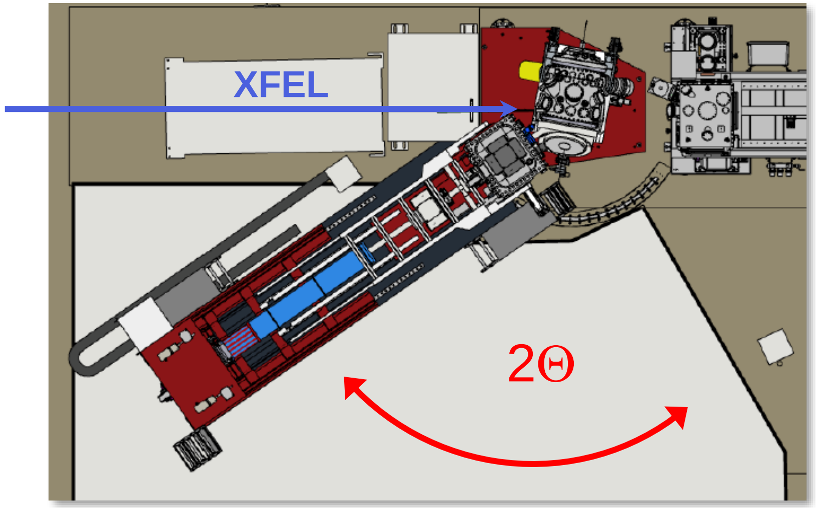
- Dedicated to time-resolved spectroscopy from solid samples:
- UHV ($p < 10^{-9}$ mbar)
- Cryogenic temperatures: RT – 20 K (specification)
- Maximum sample size: ~ 1 cm^2
- Sample degrees of freedom: 6
- In-vacuum diffractometer
- Triple-rotating flange to change scattering angle :65 deg $\leq 2\Theta \leq$ 145 deg
- Sample transfer system
- Technical offline commissioning starting in April



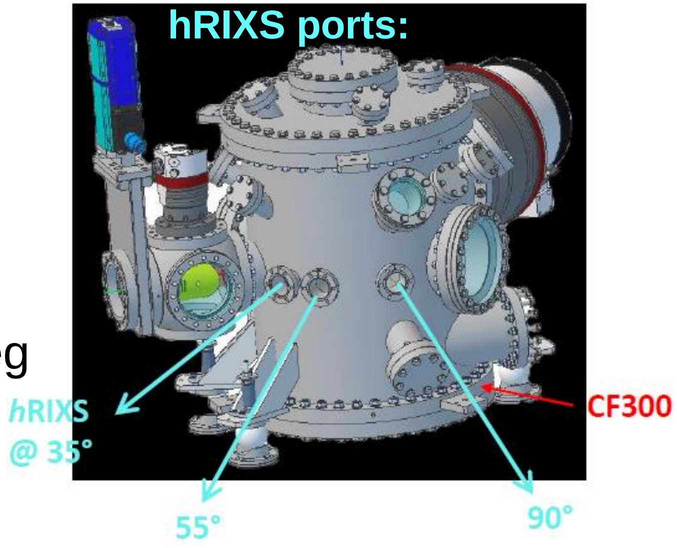
Momentum-resolved RIXS at SCS instrument:

- XRD + hRIXS: combined continuous rotation in the scattering plane: 65 deg ≤ 2Θ ≤ 145 deg





- CHEM + hRIXS: fixed geometry. Three 2Θ angles: 2Θ = 90, 125, 145 deg



hRIXS: where are we?

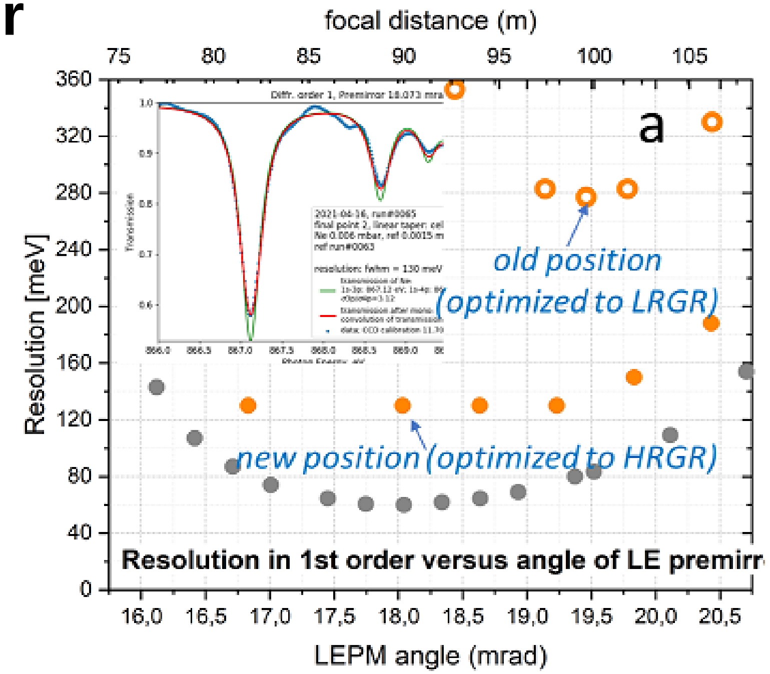
- New mono grating installed in January 2021
- hRIXS spectrometer OSAT finished in February 2021
- X-ray commissioning in May 2021
- Last proposal call: hRIXS open for the first time for user proposals → user operation in 2022-II !

Next steps:

- X-ray commissioning of time-resolved RIXS in February 2022
- User operation in 2022-II: we received 29 user proposals on hRIXS

Results from x-ray commissioning of monochromator

- High-resolution 150 l/mm monochromator grating:
  - Required for high-resolution RIXS
  - Funded by hRIXS project
  - Installed in January 2021
  - Commissioned in April: realignment of pre-mirror angle
  - Resolving power > 7,000 from Ne 1s-3p absorption lines
  - Resolving power > 10,000 confirmed with hRIXS at Cu L_3 edge and O K edge



Two working points for mono at SCS:

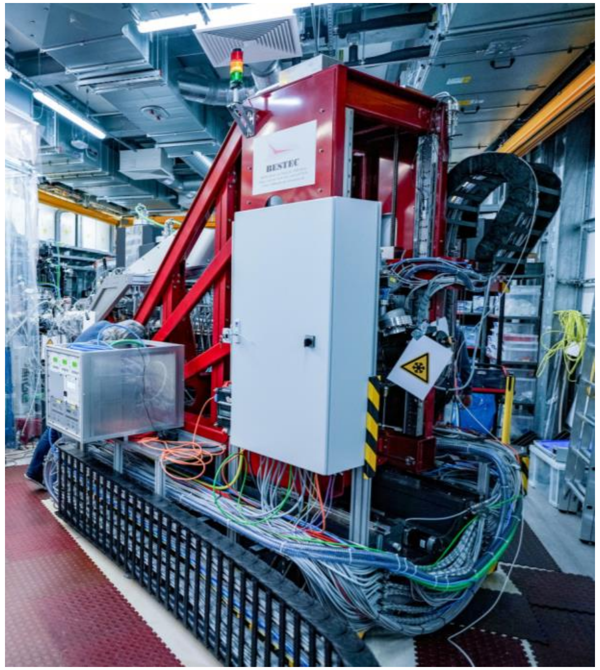
HR grating: 150 l/mm	
Resolving power	**10.000** (1^{st} order)
Pulse stretching	**80-150 fs**
X-ray pulse energy	up to 5 µJ

- High energy resolution
- Moderate temporal resolution

LR grating: 50 l/mm	
Resolving power	**3.000** (1^{st} order)
Pulse stretching	**30-50 fs**
X-ray pulse energy	up to 30 µJ

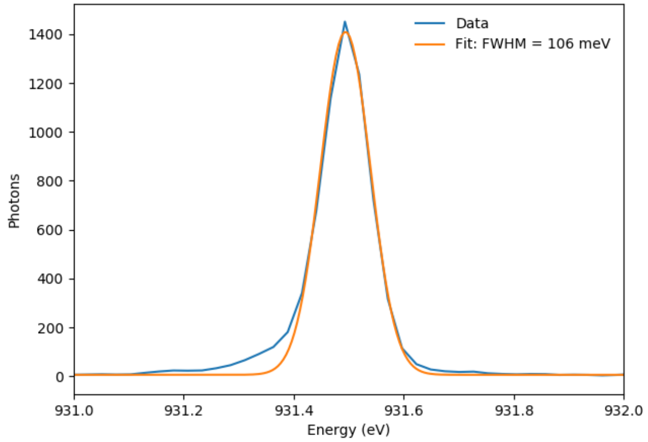
- Moderate energy resolution
- High temporal resolution

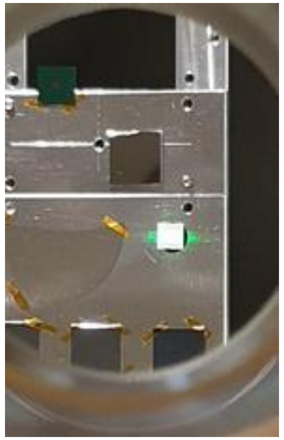
X-ray commissioning of hRIXS spectrometer: Overview



- Commissioned for static RIXS at Cu L3, Ni L3 and OK edge with 3000 l/mm grating -> achieved resolving power better than 10,000
- FEL repetition rate of 1.1 MHz, 400 pulses/train
- Spot size on sample: 15 micron (vertical), 500 micron (horizontal)
- CHEM station with solid sample environment was used
- Princeton CCD detector in integrating mode (1 – 10 min acquisition time) was used

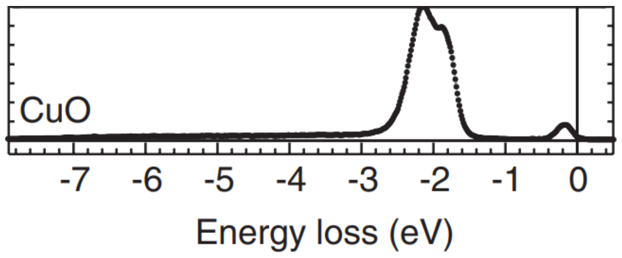
Elastic line at 931.5 eV
FWHM = 106 meV





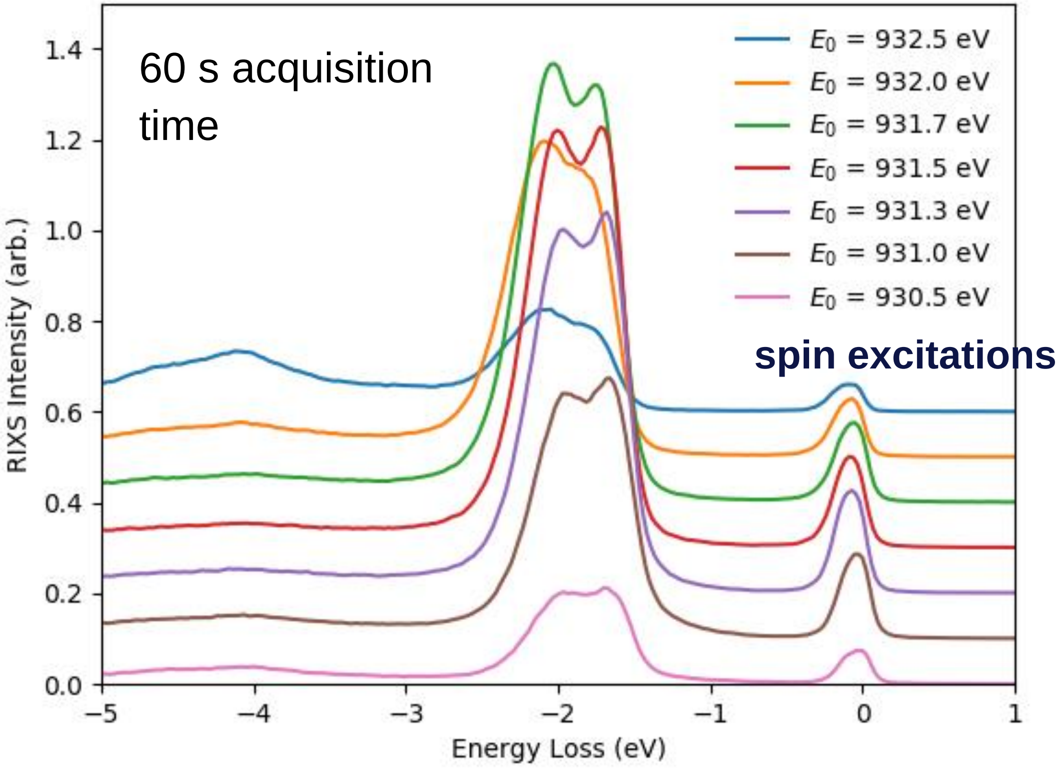
Static Cu L_3-edge RIXS: polycrystalline CuO

Transmission GATT: 100%
Photon flux at sample: 1.9 mW



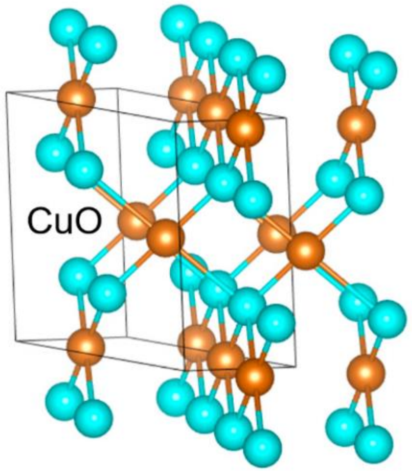
Eur. Phys. J. Special Topics **169**, 199–205 (2009)

***dd* excitations**



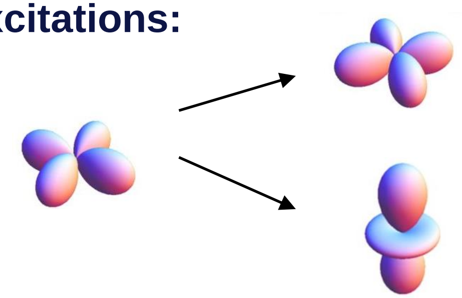
spin excitations

- Cu 3d^9 (1 hole/ site)
- co-planar coordination
- in the ground state the hole occupies 3d_{x2-y2} orbital
- *dd* excitation: in the final state the hole occupies 3*d* orbital of different symmetry e.g. 3d_{xy} or 3d_{z2}



Materials **2020**, *13*(12), 2878

***dd* excitations:**

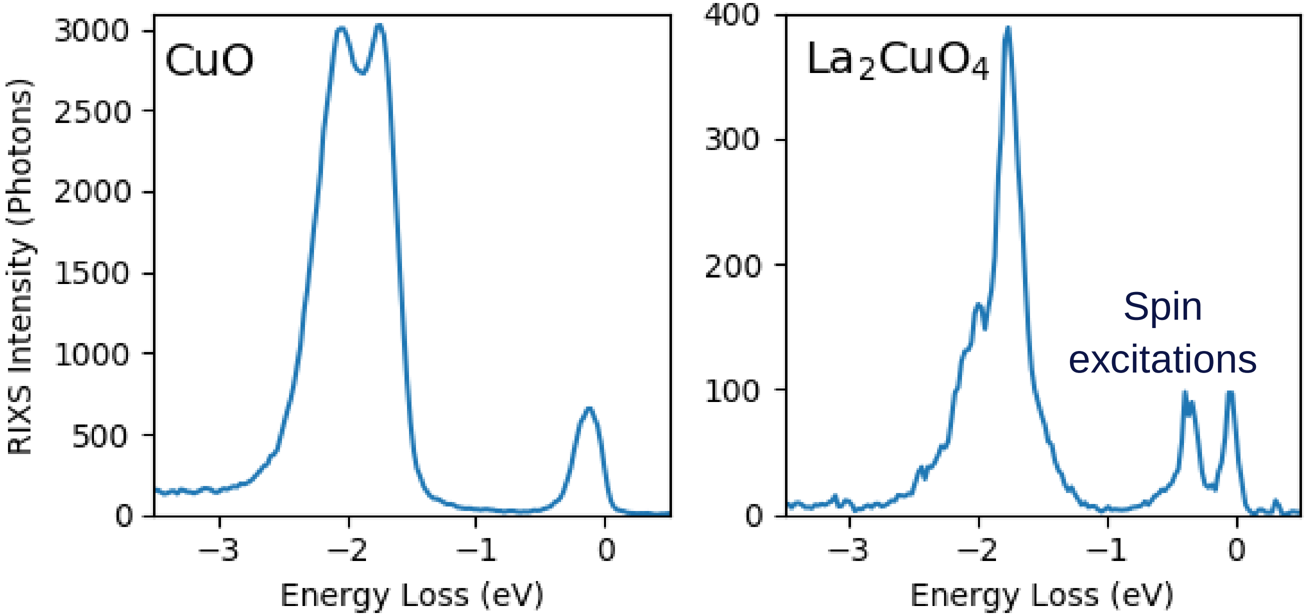


Comparison of RIXS intensity between CuO and La2CuO4:

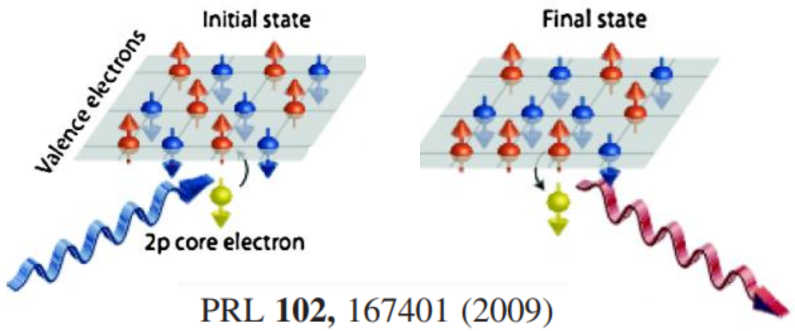
- Polycrystalline CuO
- measured with 100% GATT transmission (~2 mW), 1 min acquisition

- Thin-film La_2CuO_4
- measured with 10% GATT (~0.2 mW), 10 min acquisition

dd excitations



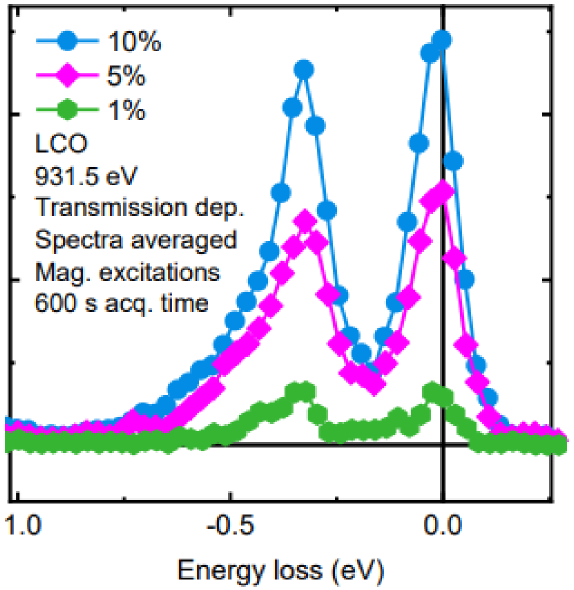
Spin excitations in La_2CuO_4:



PRL **102,** 167401 (2009)

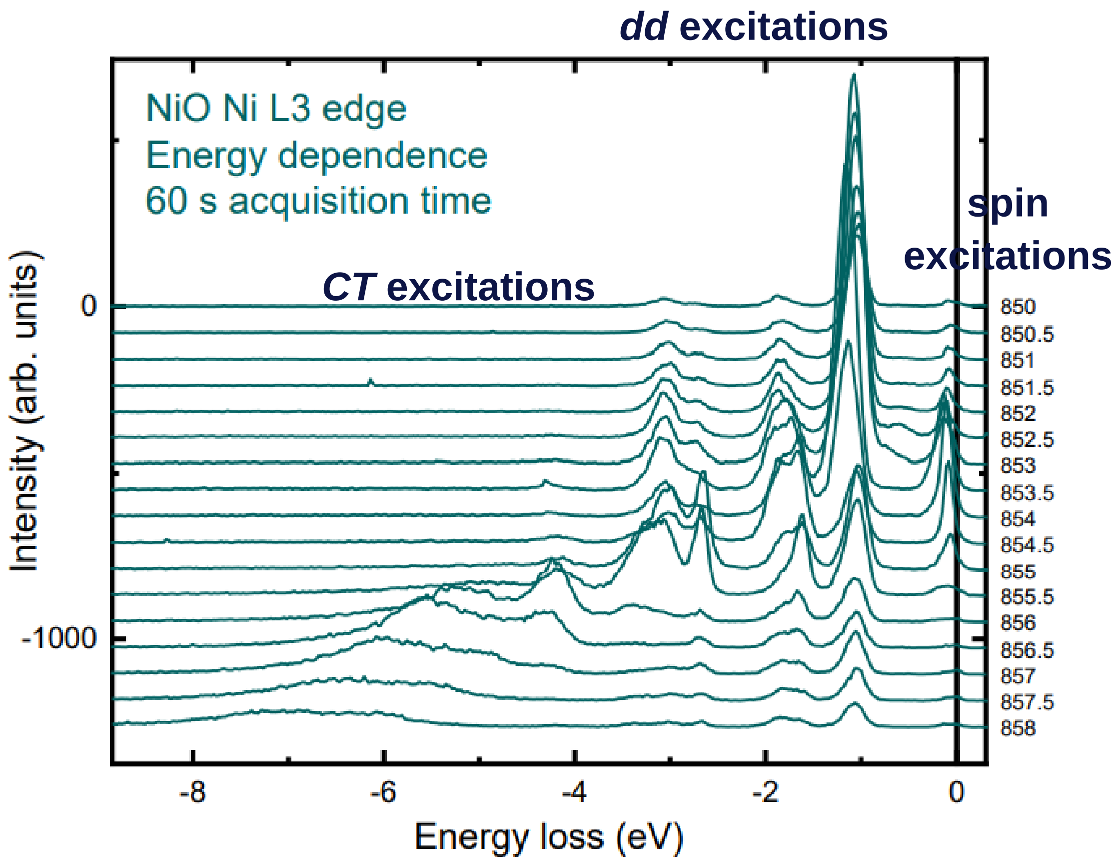
PHYSICAL REVIEW B **103**, 224427 (2021)

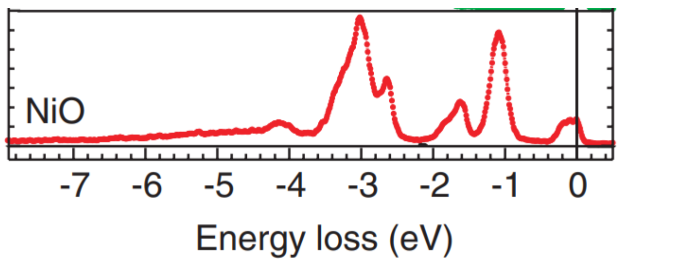
- RIXS can probe $\Delta S = 1$ magnetic excitations



Static Ni L_3-edge RIXS: single crystal bulk NiO

Transmission GATT: 100%
Photon flux at sample: 1.4 mW

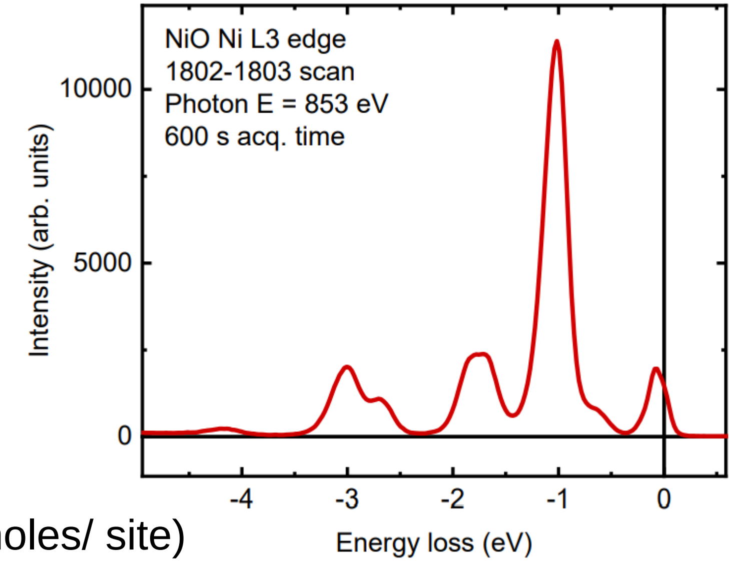




Eur. Phys. J. Special Topics **169**, 199–205 (2009)

PHYSICAL REVIEW B **96**, 020409(R) (2017)

PHYSICAL REVIEW LETTERS **124**, 067202 (2020)

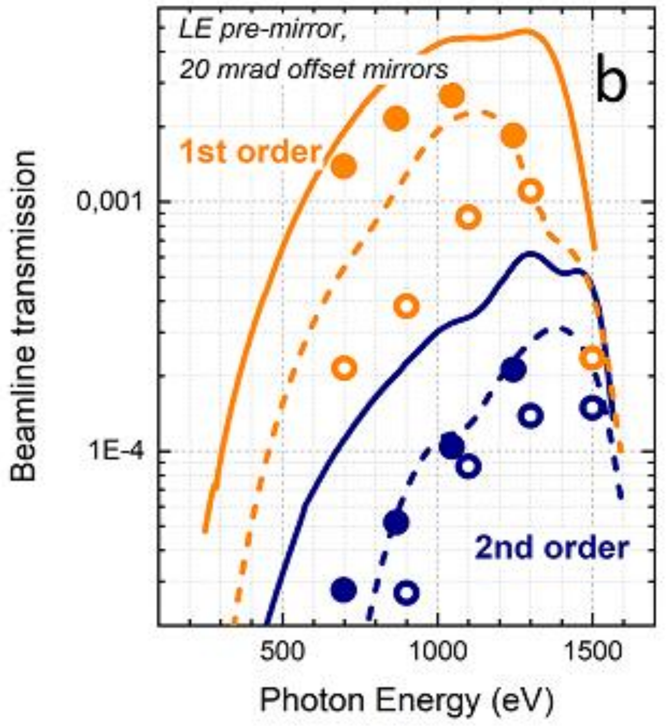


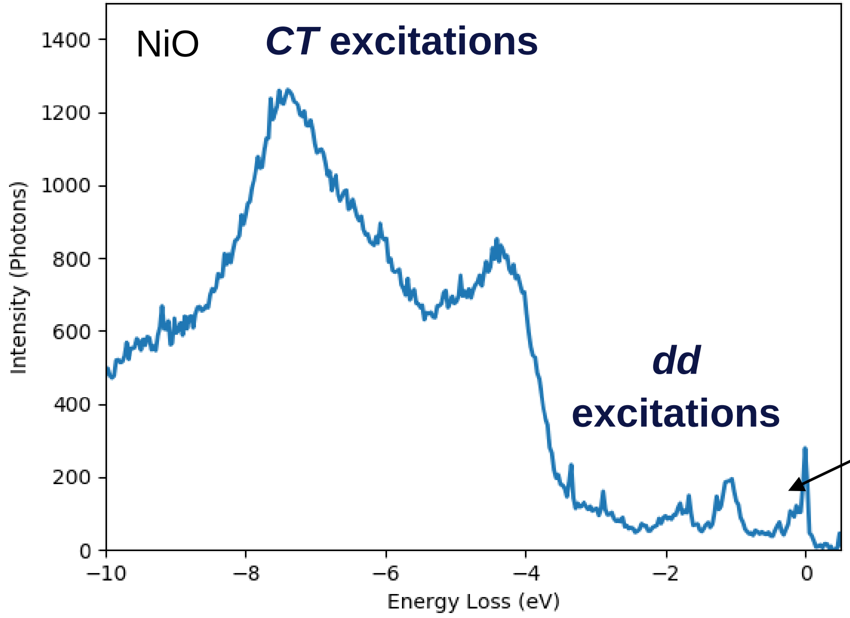
- Cu $3d^8$ (2 holes/ site)
- octahedral coordination
- *dd* excitations more complex due to multiplet effects

O *K*-edge RIXS

Transmission GATT: 100%
Photon flux at sample: 0.14 mW

Beamline transmission through 100μm exit slit

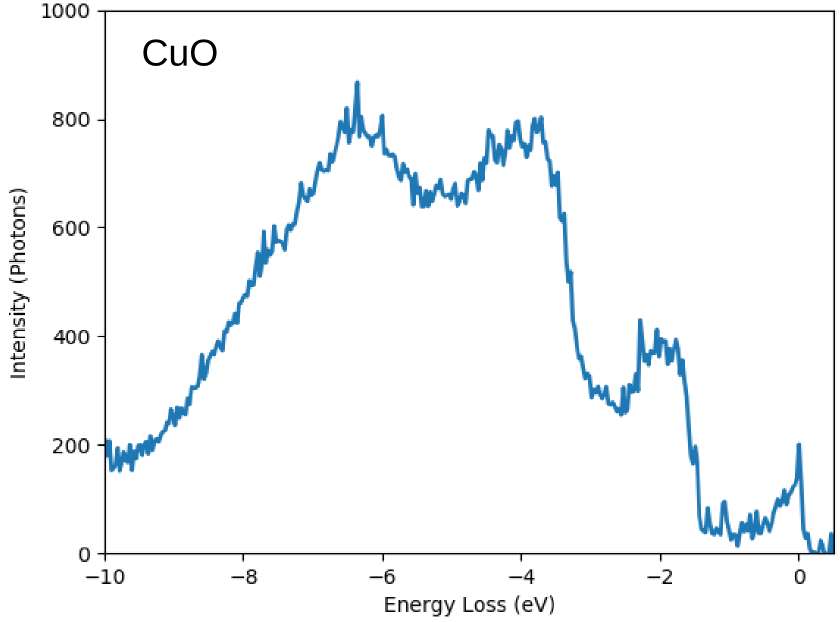




Sum of 15
10 min spectra
E0 = 531.6 eV

bi-magnon

RIXS can probe
ΔS = 0 magnetic excitations

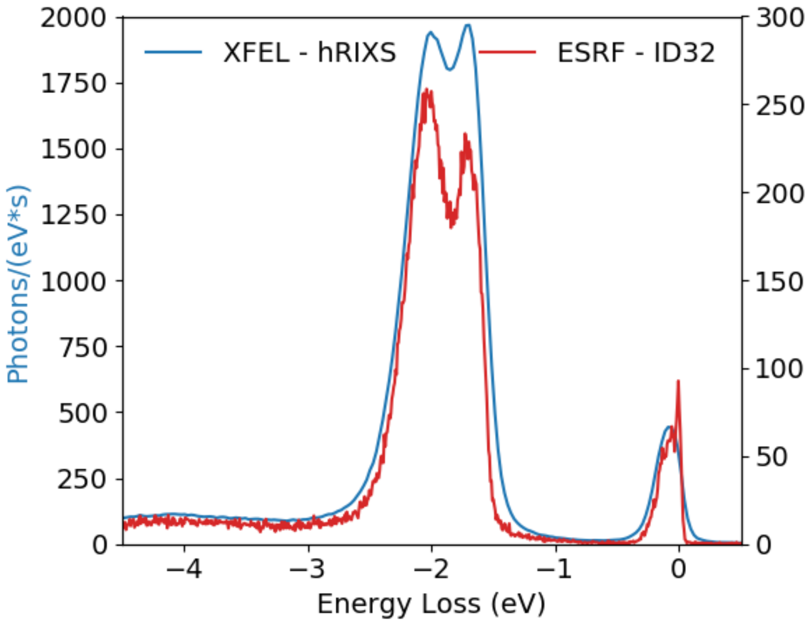


Sum of 14
10 min spectra
E0 = 530.1 eV

hRIXS: measured count rates from the samples

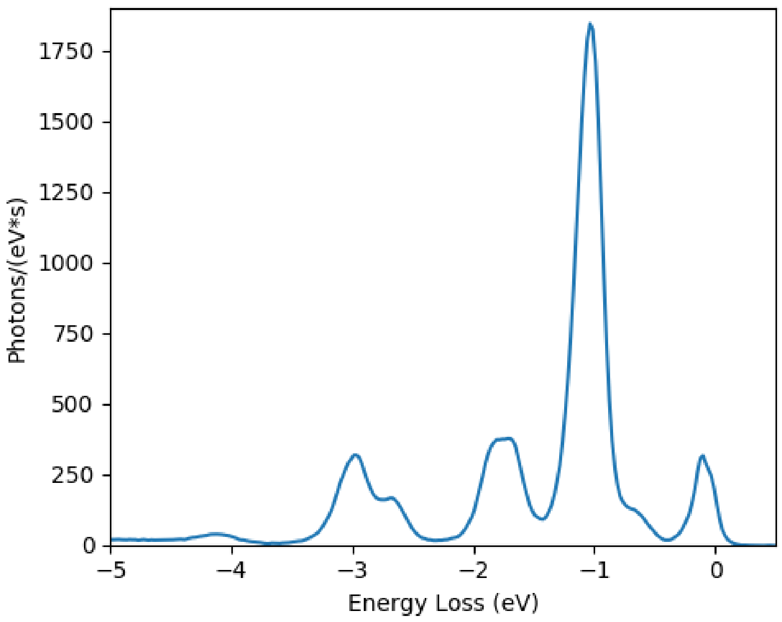
Cu L_3-edge

Incident Beam:
1.3 x 10^{13} ph/s



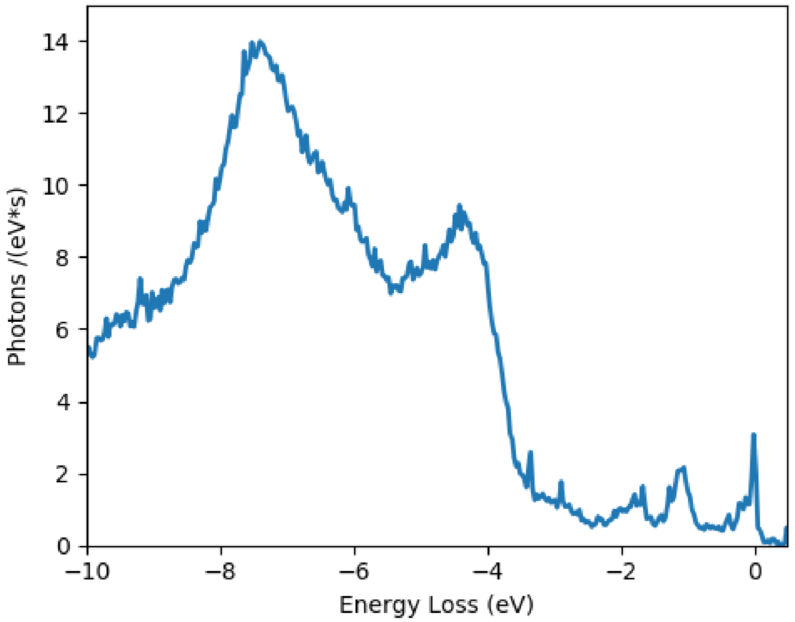
Ni L_3-edge

Incident Beam:
1.0 x 10^{13} ph/s



O K-edge

Incident Beam:
1.6 x 10^{12} ph/s



Summary hRIXS x-ray commissioning:

- hRIXS Spectrometer Commissioning
  - Commissioning of Cu L_3, Ni L_3 and O *K* edge
  - Static RIXS from prototypical Mott insulators, bulk samples and thin films
  - Achieved combined resolving power better than 10,000 at O *K* edge
  - Count rates with 400 pulses/train similar or higher than at 3rd generation synchrotron instruments

Edge	Energy (eV)	Flux on sample (ph/s)	ΔE (meV)	E/ΔE
Cu L_3	930	1.3×10^{13}	106	8 700
Ni L_3	853	1.0×10^{13}	122*	6 900*
O K	530	1.6×10^{12}	49	10 400

* not enough time to optimize

User-assisted commissioning in Q1 2022: enabling optical pump – RIXS probe at SCS for solids and liquids

- Pump-probe on solid-state samples
  - KW6 will be used to commission the spectrometer, laser in-coupling and time-resolved RIXS
  - KW8 will be dedicated to pump-probe measurements on photoexcited La_2CuO_4 and NiO

- Pump-probe measurements on Solution Samples
  - KW11 will commission the cylindrical liquid jet system at O K-edge
  - KW13 will be dedicated to pump-probe at Fe L-edge

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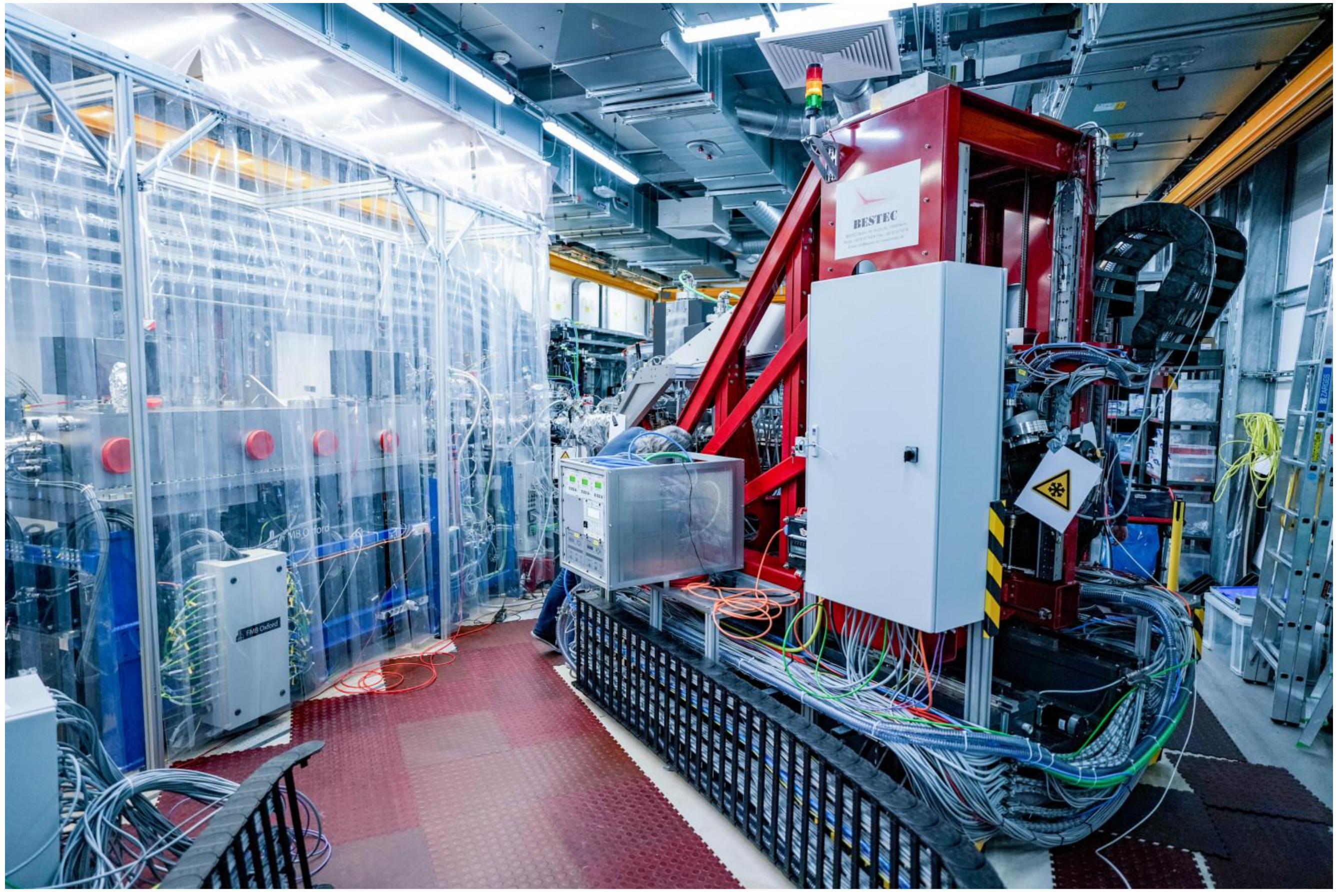
Thank you!



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