State of hRIXS and the case for chemical dynamics

Alexander Föhlisch, Helmholtz-Zentrum Berlin and University of Potsdam

Momentum-Time resolved resonant inelastic x-ray scattering at the European XFEL: Heisenberg RIXS

Alexander Föhlisch, Helmholtz-Zentrum Berlin and University of Potsdam
Alexei Erko, Helmholtz-Zentrum Berlin
Giacomo Ghiringhelli, Dip. Fisica, Politecnico di Milano
Rafael Abela, 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
Marco Grioni, Ecole Polytechnique Fédérale de Lausanne
Hao Tjeng, Max-Planck-Institut für Chemische Physik fester Stoffe Dresden
Jeroen van den Brink, IFW Dresden
Maurits W. Haverkort, Max-Planck-Institut für Festkörperforschung Stuttgart
Frank de Groot, Utrecht University
Stefan Eisebitt, Technische Universität Berlin
Marc Simon, Laboratoire Chimie-Physique-Matière et Rayonnement, SOLEIL, Paris

Bringing together the European RIXS and FEL expertise

	Instrument design	FEL	Experiment	Theory
Alexander Föhlisch	●	●	●	
Alexei Erko	●			
Giacomo Ghiringhelli	●		●	
Rafael Abela		●	●	
Bernhard Keimer			●	
Simone Techert		●	●	
Jan-Erik Rubensson	●		●	
Joseph Nordgren	●		●	
Marco Grioni	●		●	
Hao Tjeng			●	
Jeroen van den Brink				●
Maurits W. Haverkort				●
Frank de Groot				●
Stefan Eisebitt	●	●	●	
Marc Simon			●	

Grand Challenges:
Functionality of matter

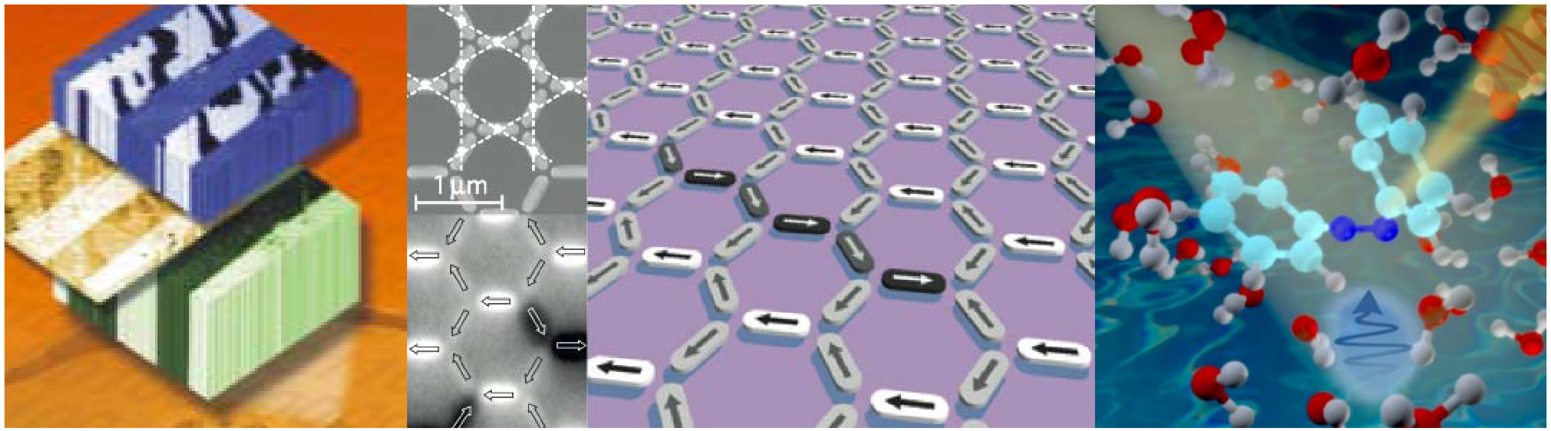
How do we communicate and archive knowledge ?

How do we harvest, convert and store energy ?

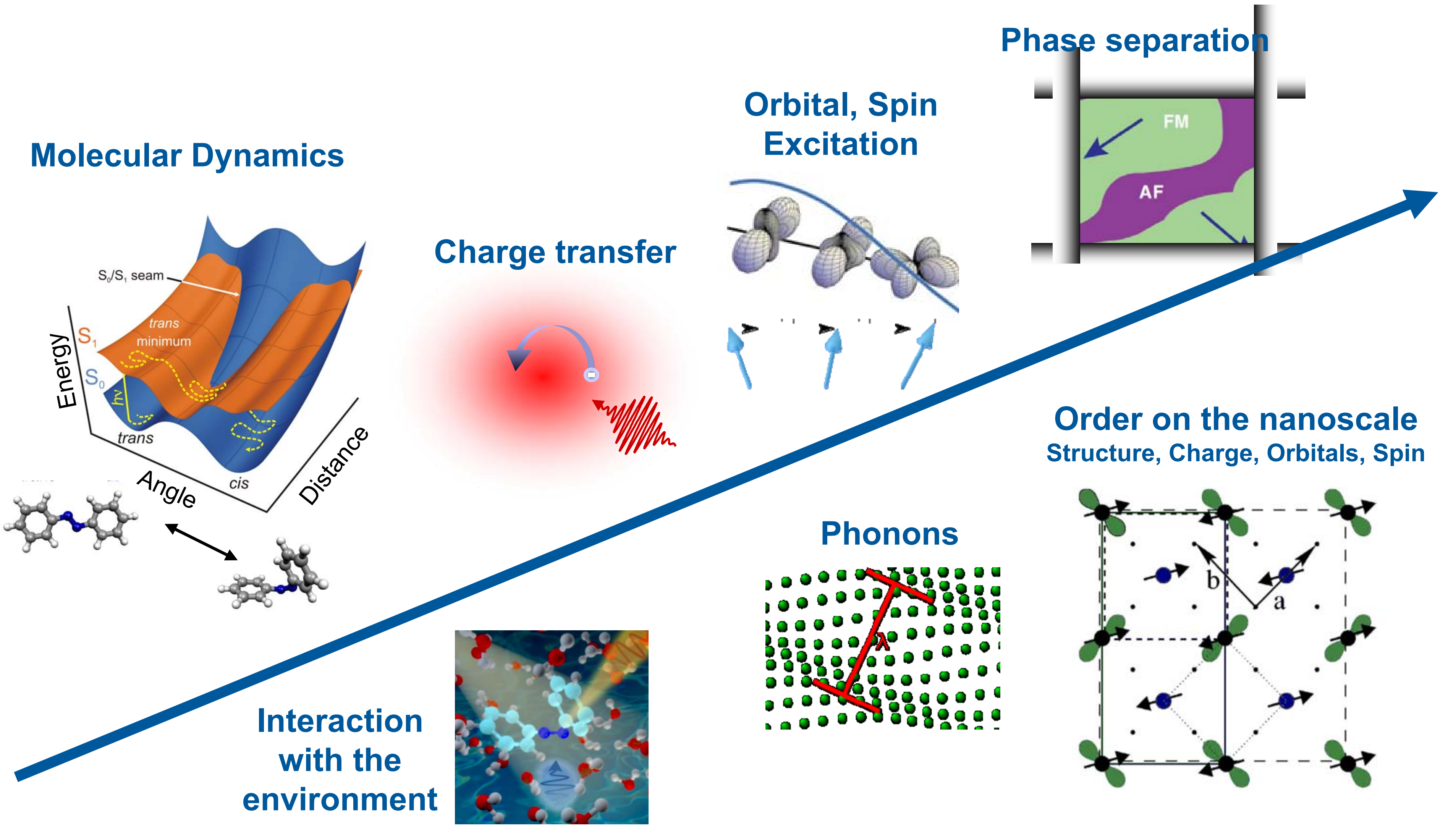
How do we govern selectivity and rate of chemical processes ?

Utilize and govern functionality

Governing principles of materials function

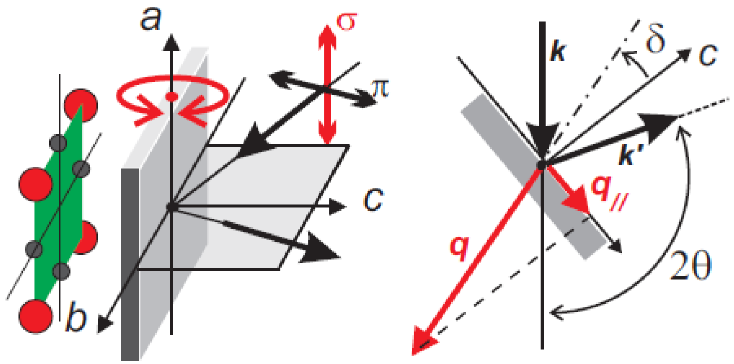


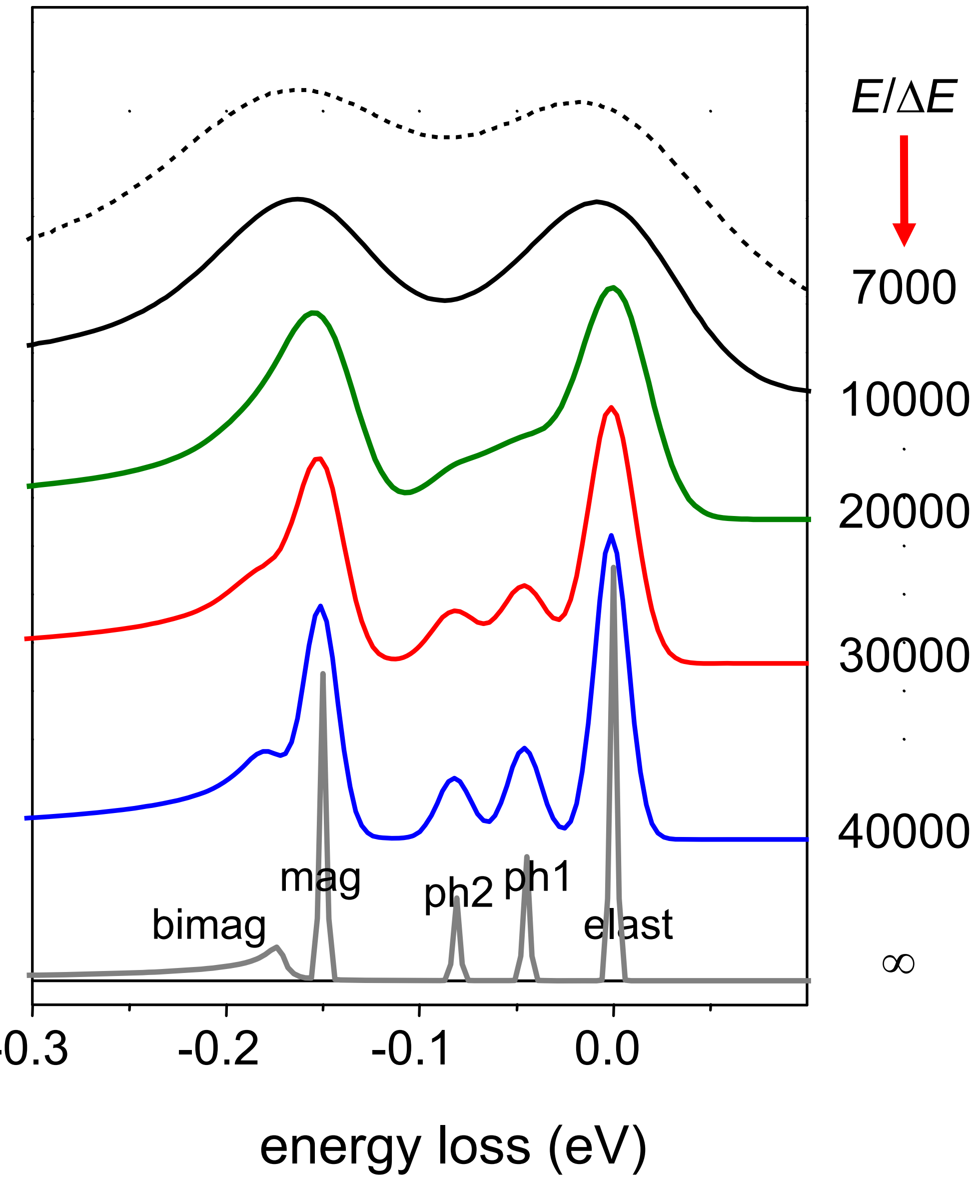
Governing principles of materials function : Coupled degrees of freedom from local to global



The role of high resolution RIXS: low energy excitations in solids

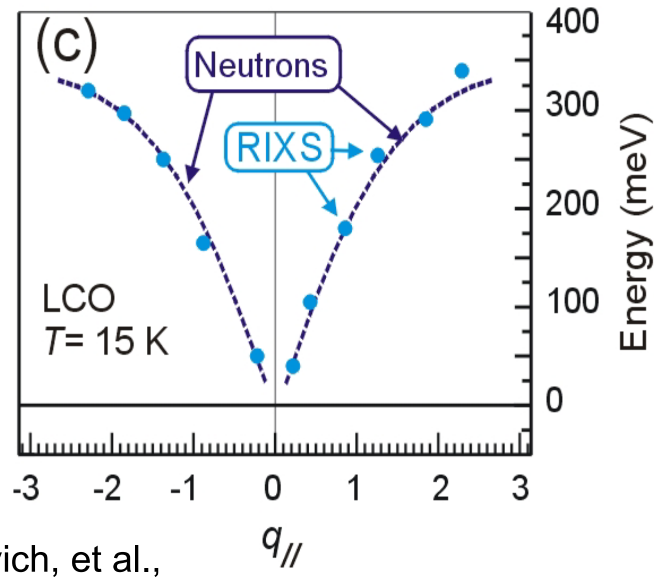
Resonant inelastic X-ray scattering



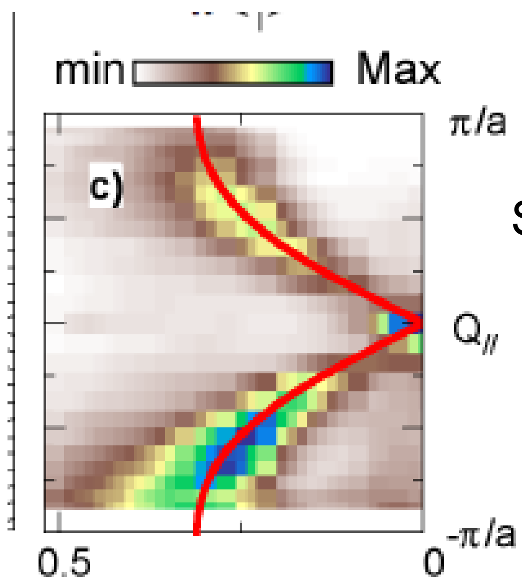


The role of high resolution RIXS: low energy excitations in solids

La_2CuO_4

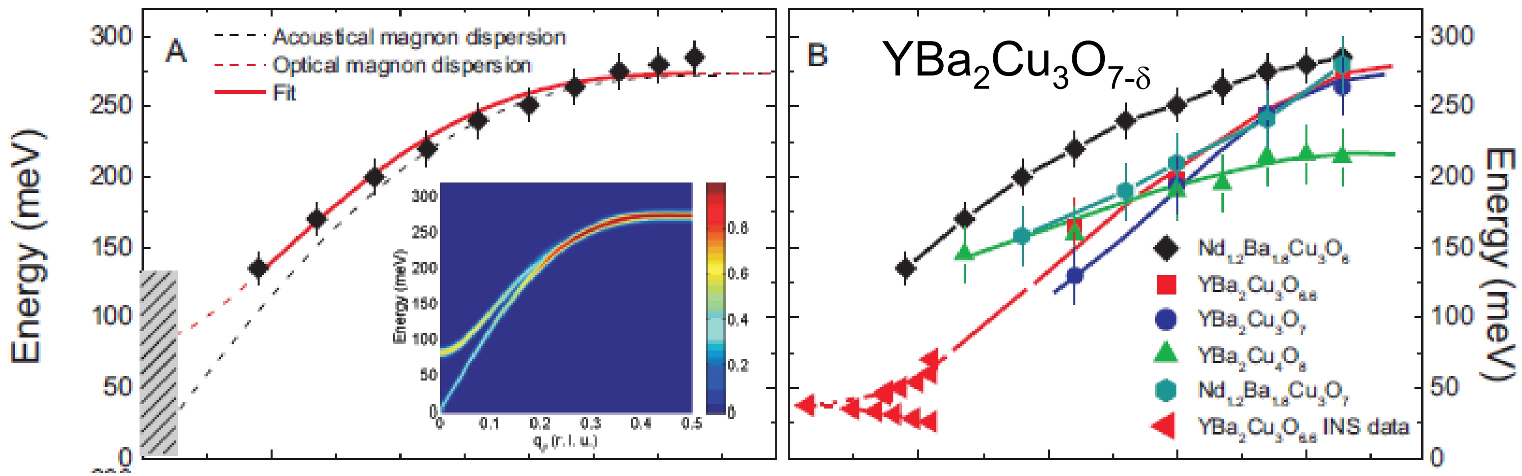


L. Braicovich, et al.,
Phys. Rev. Lett. **104**, 077002 (2010)



$Sr_2CuO_2Cl_2$

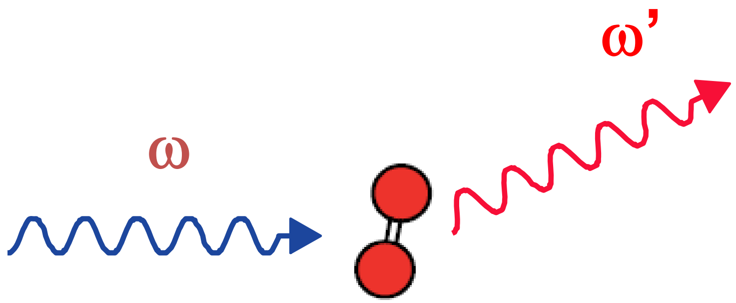
M. Guarise, et al.,
Phys. Rev. Lett. **105**, 157006 (2010)



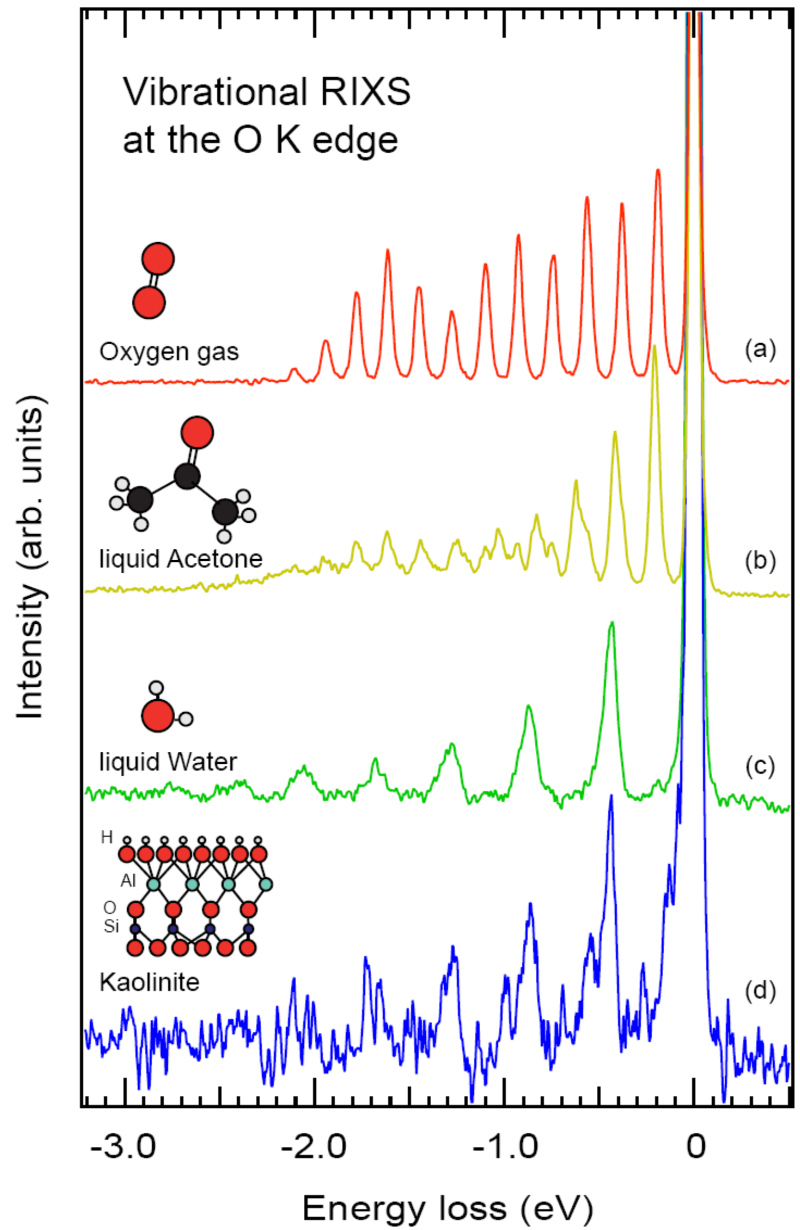
M. Le Tacon, et al., Nature Phys. **7**, 725 (2011)

Sub-natural line width RIXS for molecular systems: Mapping Potential energy surfaces and fs wave packet dynamics

PRL 106 153004 (2011). **PRL 104, 193002 (2010)**, PRA 2011
HZB, UP: J.Schlappa, M. Berglund, A. Föhlisch
MaxLab: B. Kennedy, A. Pietzsch, F. Hennies
U Uppsala: J.E. Rubensson
U Stockholm: Y-P Sung, F. Gel'mukhanov,
M. Odelius, H. Agren
SLS: T. Schmitt, V. Strokov

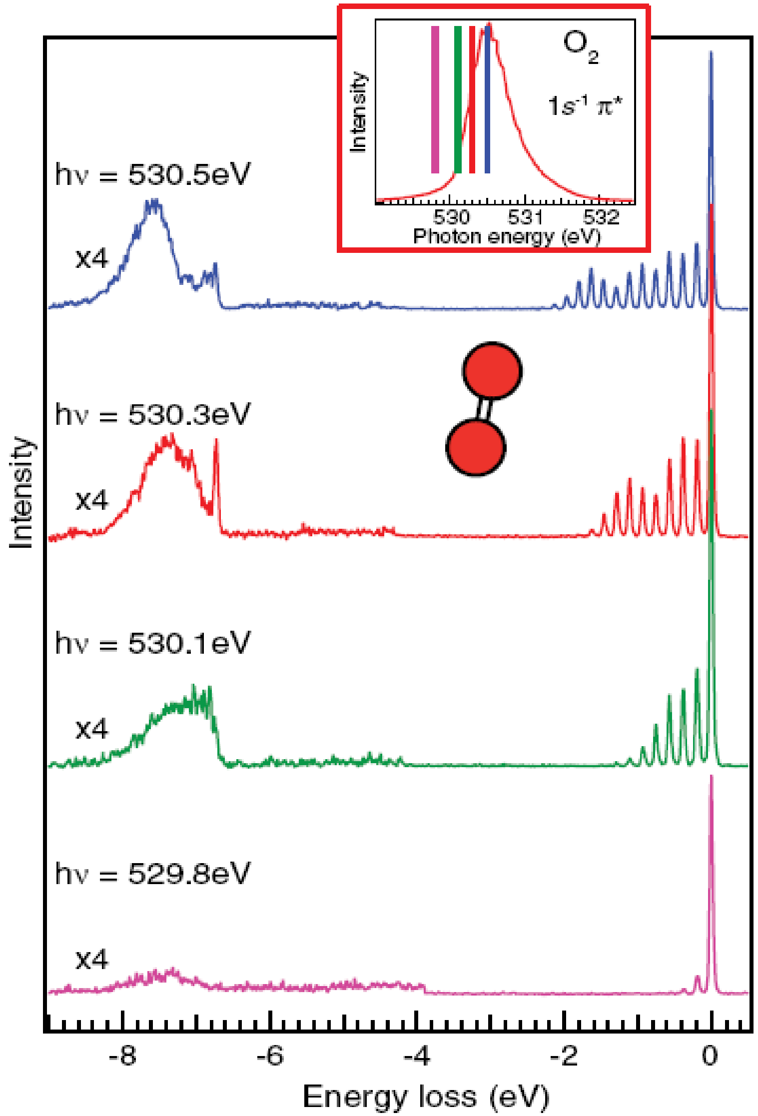


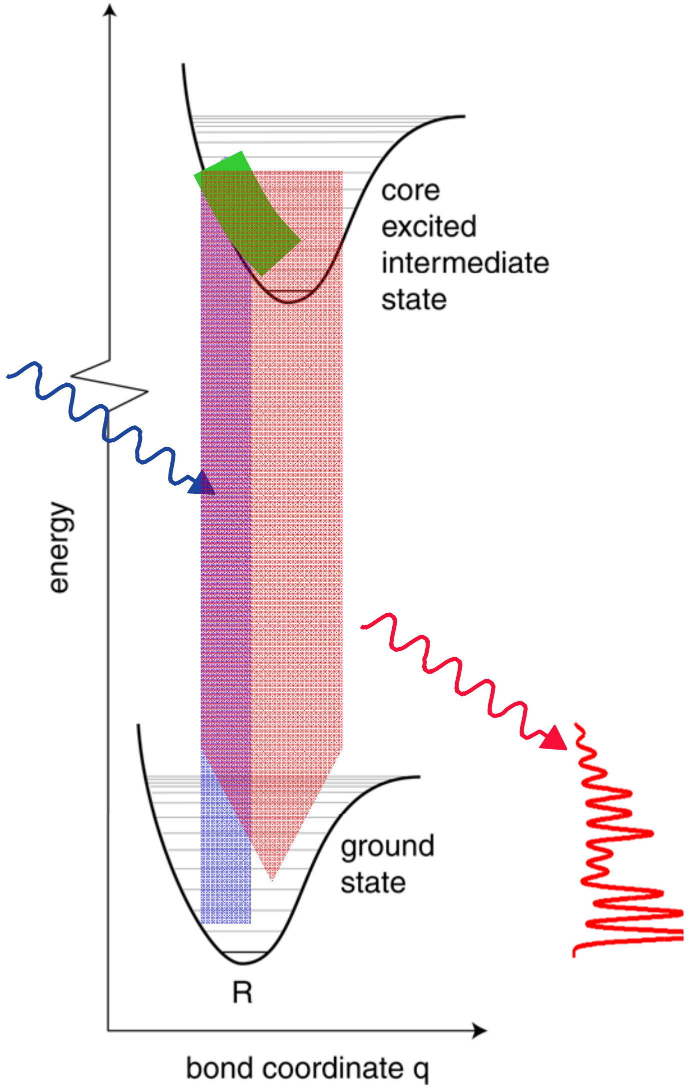
$$\Delta\varepsilon = \varepsilon_{vib} + \varepsilon_{elec}$$



Molecular dynamics from sub natural linewidth RIXS

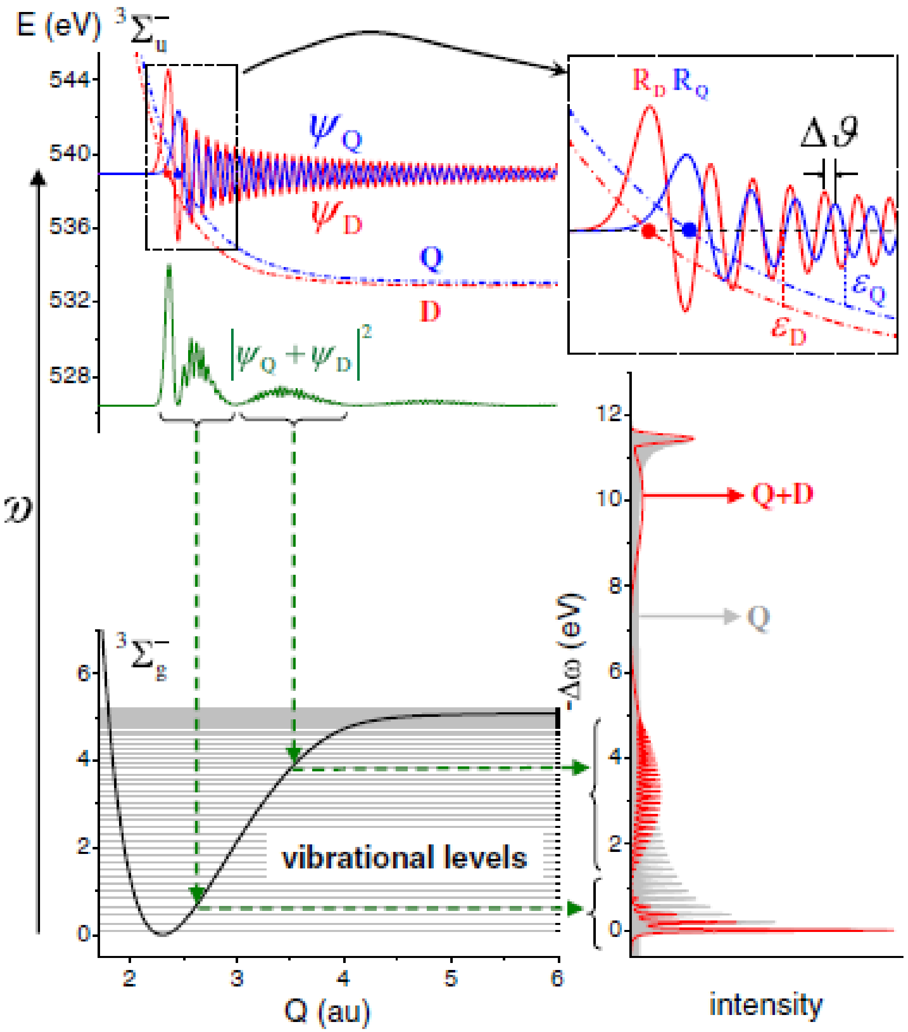
Molecular O_2 O-K edge RIXS at SAXES/Adress





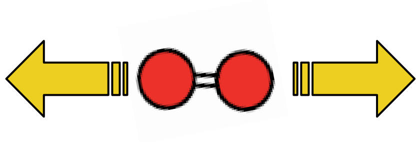
PRL **104**, 193002 (2010)

A nice example: The discontinuous nature of breaking the O-O bond!



Molecular O_2 O-K edge

RIXS at SAXES/Adress



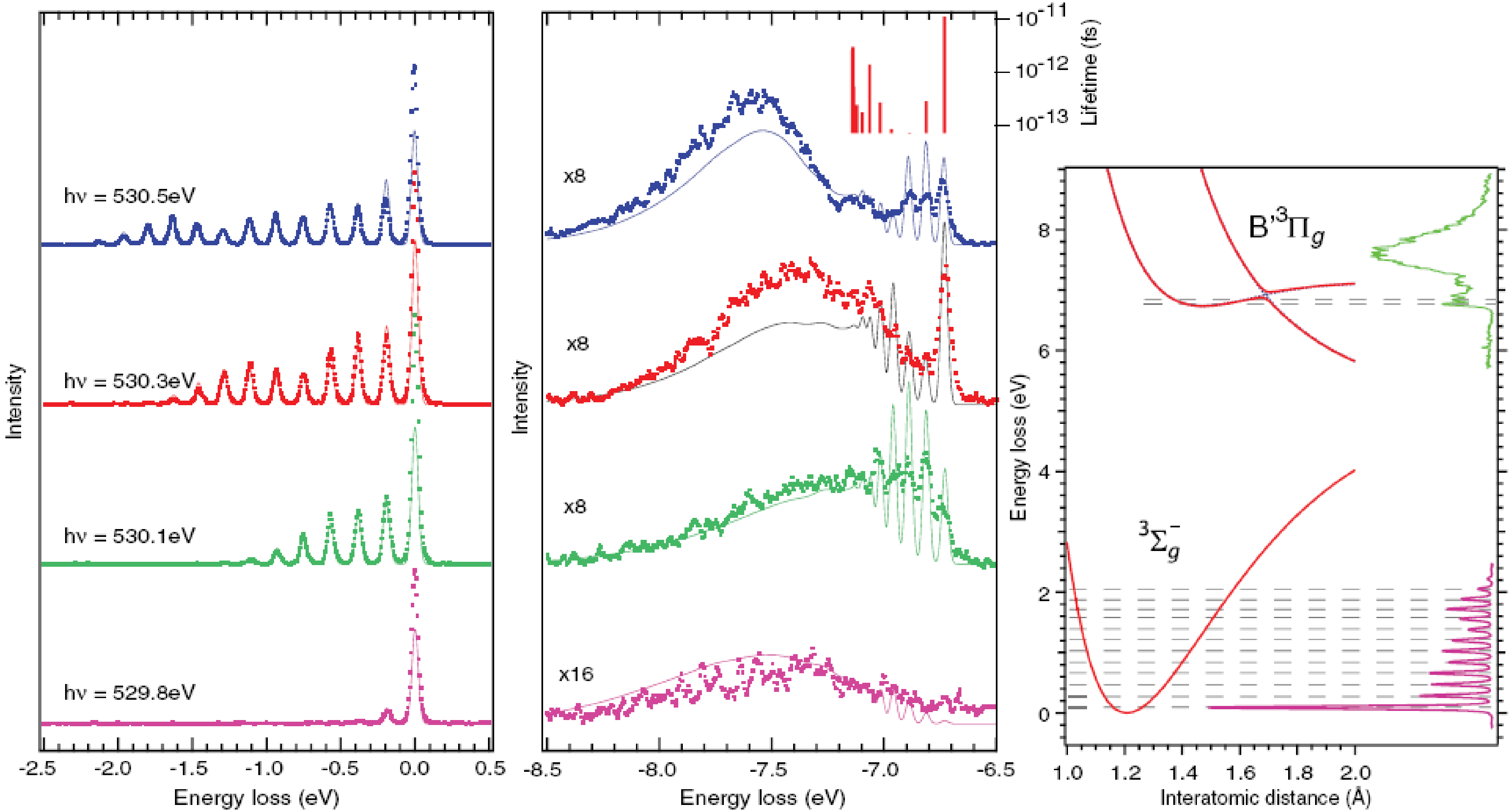
- Nuclear wave packet in dissociation creates amplitude only at selected atomic distances:

- Chemistry can only happen there!

Spatial Quantum Beats in Vibrational Resonant Inelastic Soft X-ray Scattering at dissociating States of Oxygen

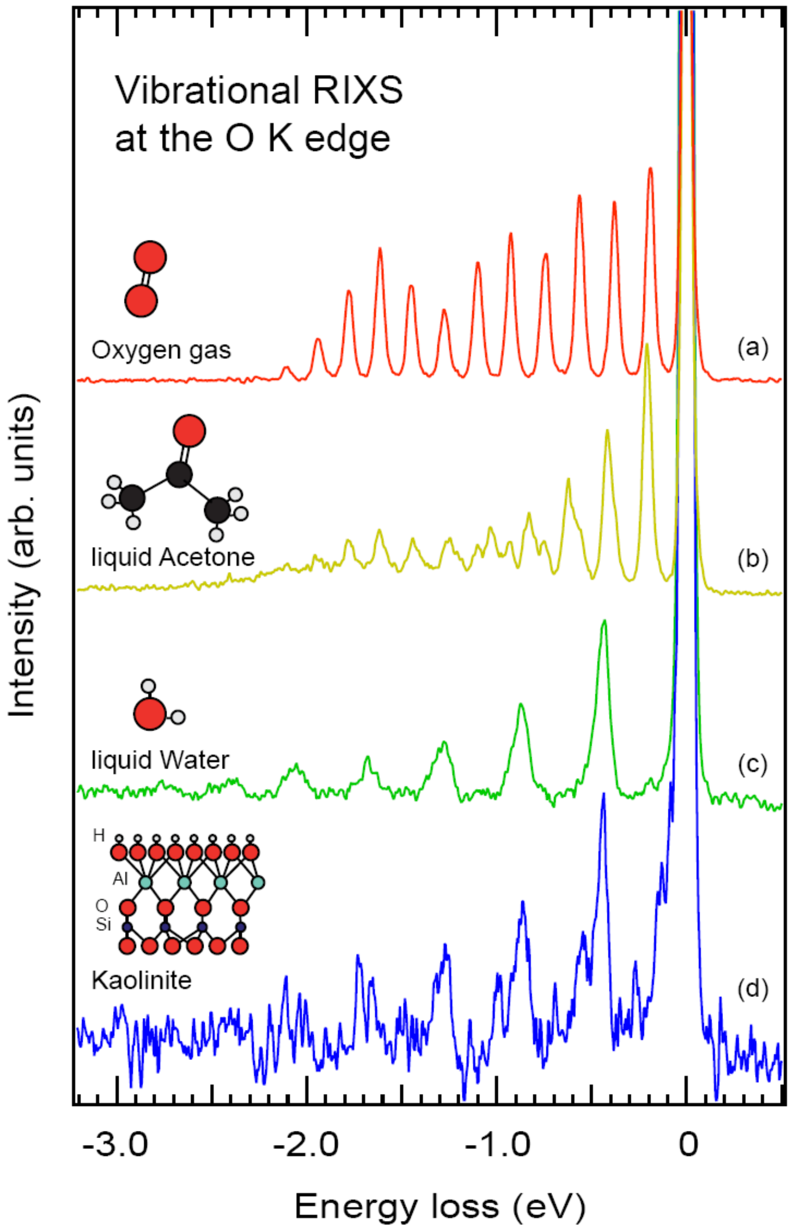
A. Pietzsch et al, Phys. Rev. Lett. 153004 (2011). DOI: 10.1103/PhysRevLett.106.153004

Approaching curve crossings!



PRL **104**, 193002 (2010)

Sub Natural line width RIXS is robust! Dipolar interaction (broadening)



The role of dipolar interaction (broadening)

$$\gamma \rightarrow \gamma + \gamma_S$$

$$\gamma_S \approx \frac{4}{3}\sqrt{\pi \ln 2}|\Delta\mu_f||\mu_S|\sqrt{\rho/a^3}.$$

Change in solute dipole moment in RIXS

$$\Delta\mu_f = \mu_f - \mu_0$$

The solvent dipole moment

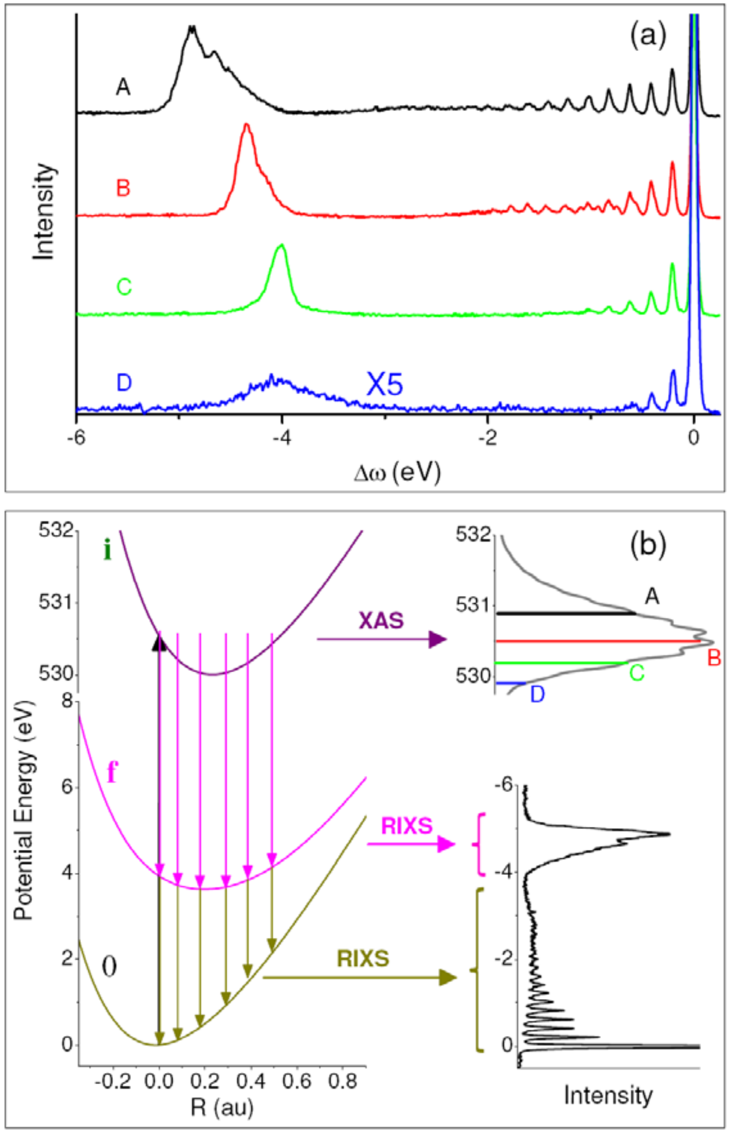
$$\mu_S$$

ρ is the concentration of the solvent molecules

α is the radius of the solute (Weisskopf radius)

Sub Natural line width RIXS is robust! Dipolar interaction (broadening)

Liquid acetone O K-edge



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Change in solute dipole moment in RIXS

$$\Delta\mu_f = \mu_f - \mu_0$$

The solvent dipole moment

$$\mu_S$$

Acetone in Acetone

$$\gamma_S \approx 0.04 \text{ eV}$$

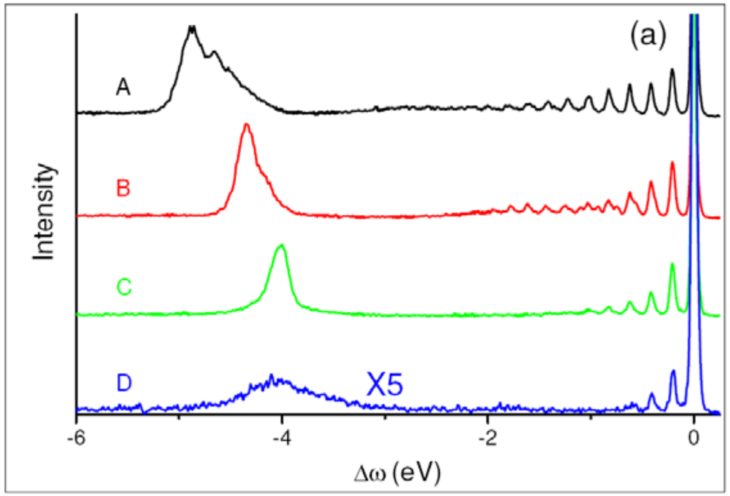
$$\mu_0 = 3.038 \text{ D}$$
$$\mu_f = 1.958 \text{ D}$$

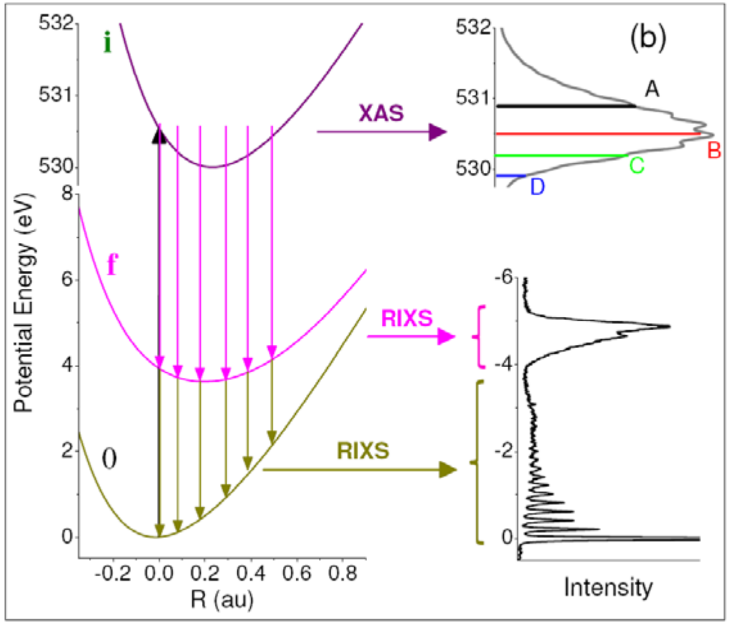
Acetone in Water

$$\gamma_S \approx 0.11 \text{ eV}.$$

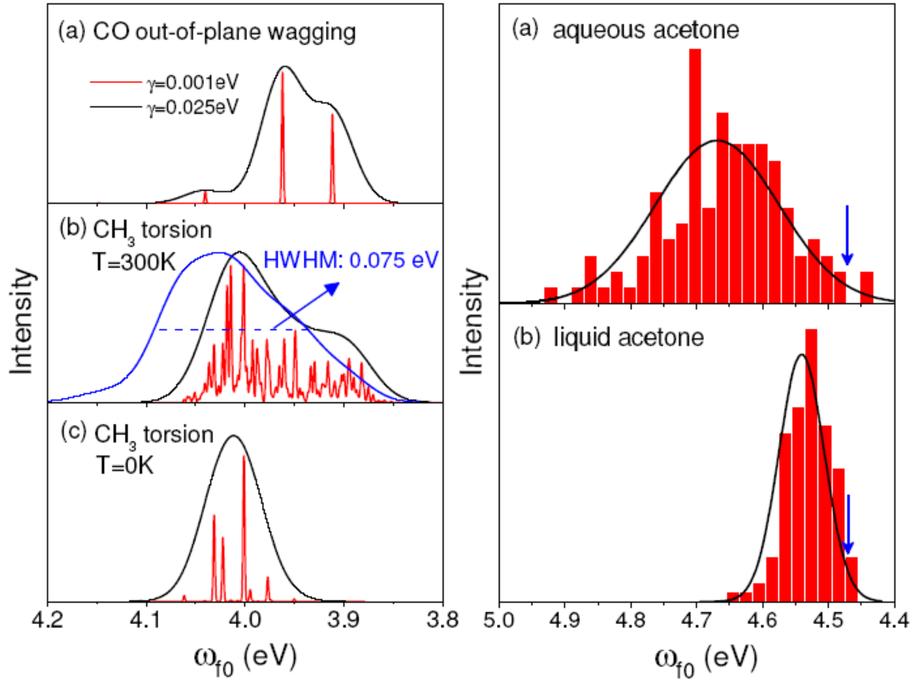
Sub Natural line width RIXS is robust! Low energy degrees of freedom

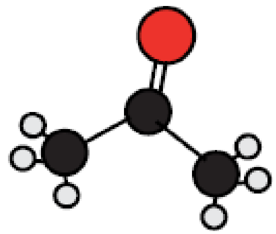
Liquid acetone O K-edge





The role of low energy degrees of freedom

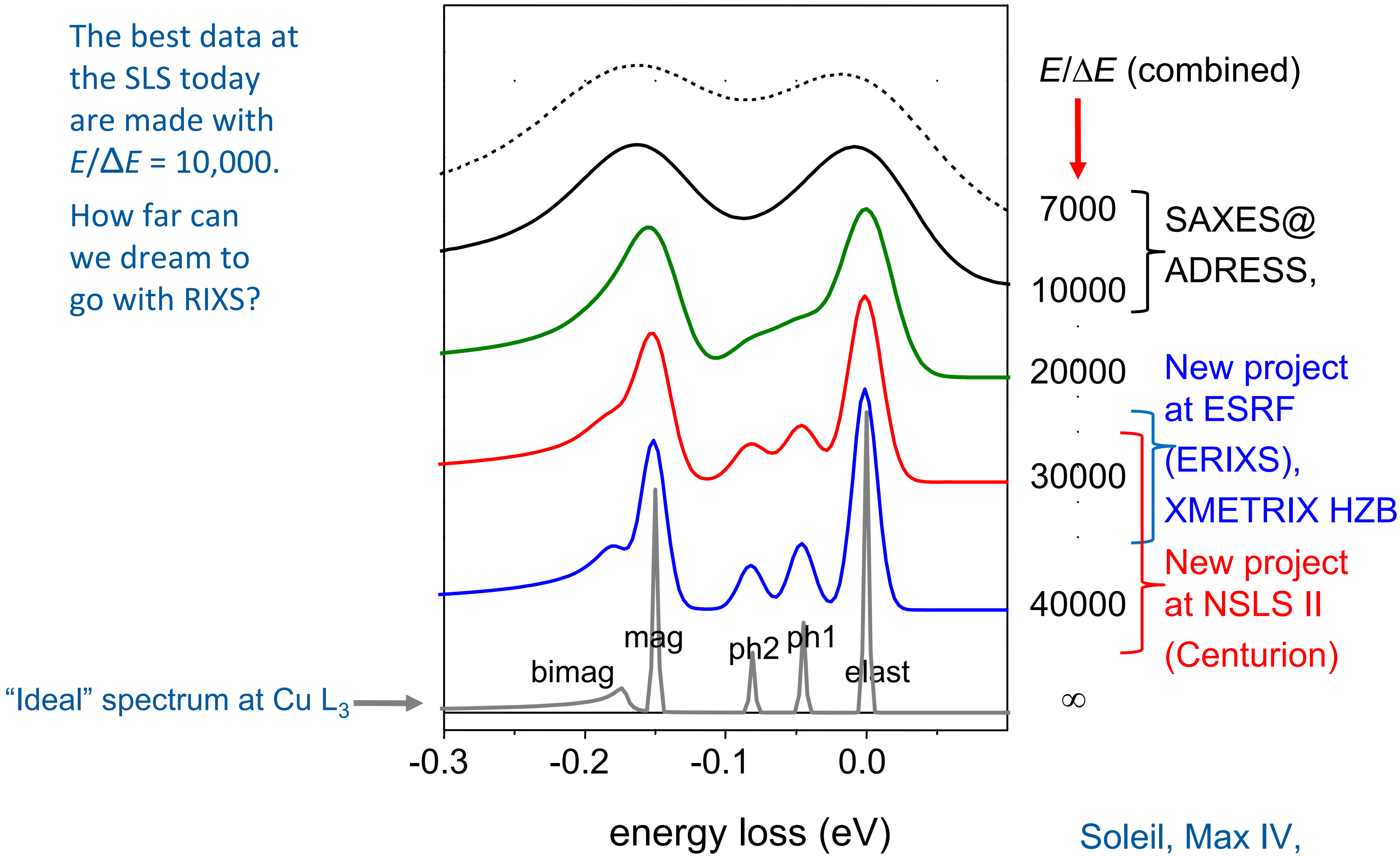




The race for RIXS at relevant energy scales

The best data at the SLS today are made with $E/\Delta E$ = 10,000.

How far can we dream to go with RIXS?



Soleil, Max IV, NSRRC, Spring-8

At XFEL: Get RIXS to the Heisenberg limit in time and energy

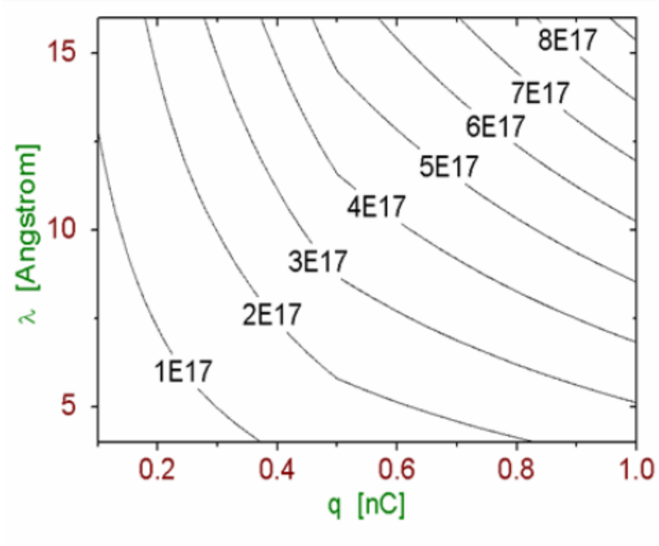
+ Average brilliance/brightness:

SASE3: $5x10^{17}$ ph/s

ESRF ID8 after upgrade: $5x10^{15}$ ph/s
NSLS II: 6-$8x10^{15}$ ph/s

→ two orders of magnitude gain in photon flux
→ improve energy resolution
→ polarization analysis
→ short data acquisition time

→ 3D-momentum-transfer mapping , dilute samples



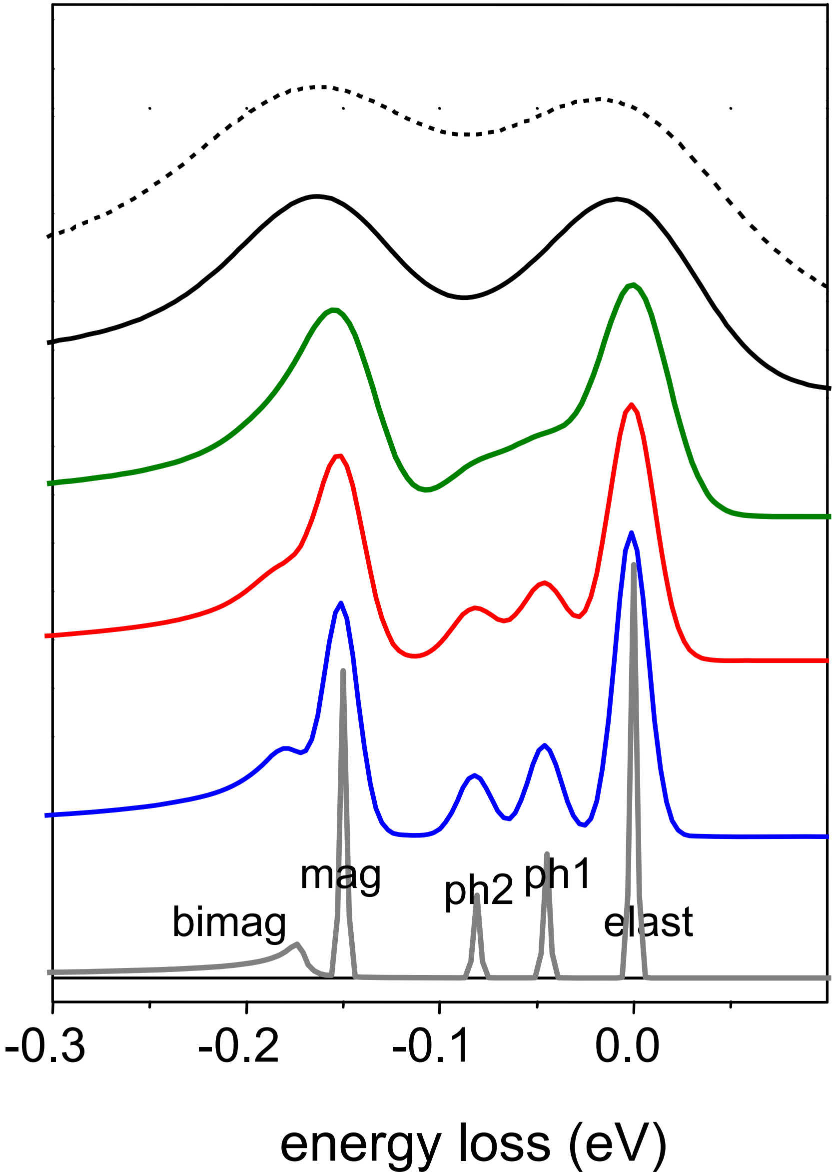
Average number of photons

Data courtesy by M.Yurkov et.al.

+ femtosecond time structure:

→ high brightness pump-probe experiments
→ transient phases
→ photochemistry

At XFEL: Get RIXS to the Heisenberg limit in time and energy

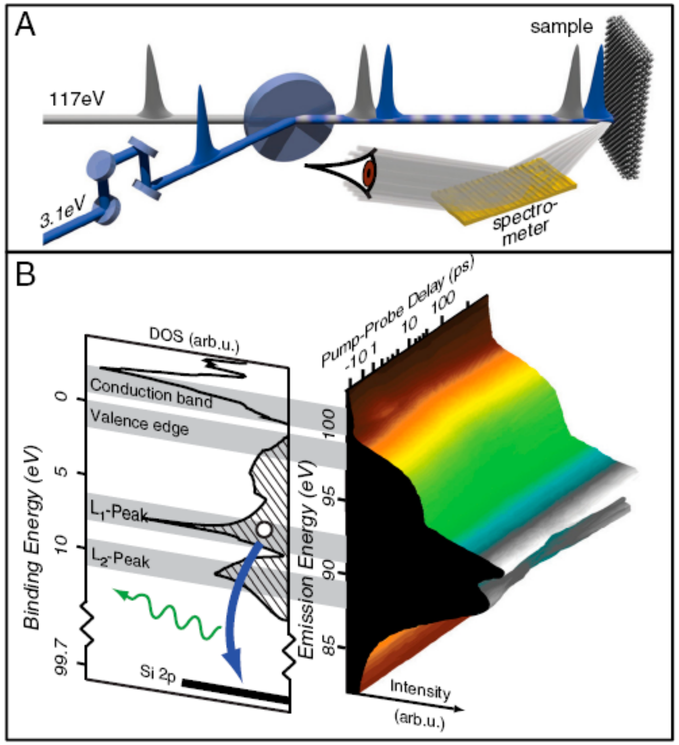


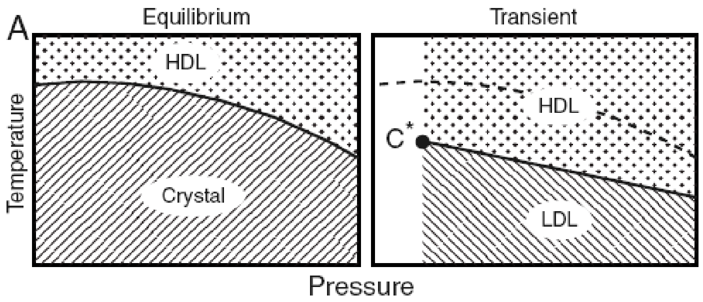
$E/\Delta E$ ↓	Δt (fs) @ 1 keV ↑	Δt (fs) @ 300 eV ↑
7000	28	93
10000	41	137
20000	82	273
30000	123	410
40000	165	550
∞		

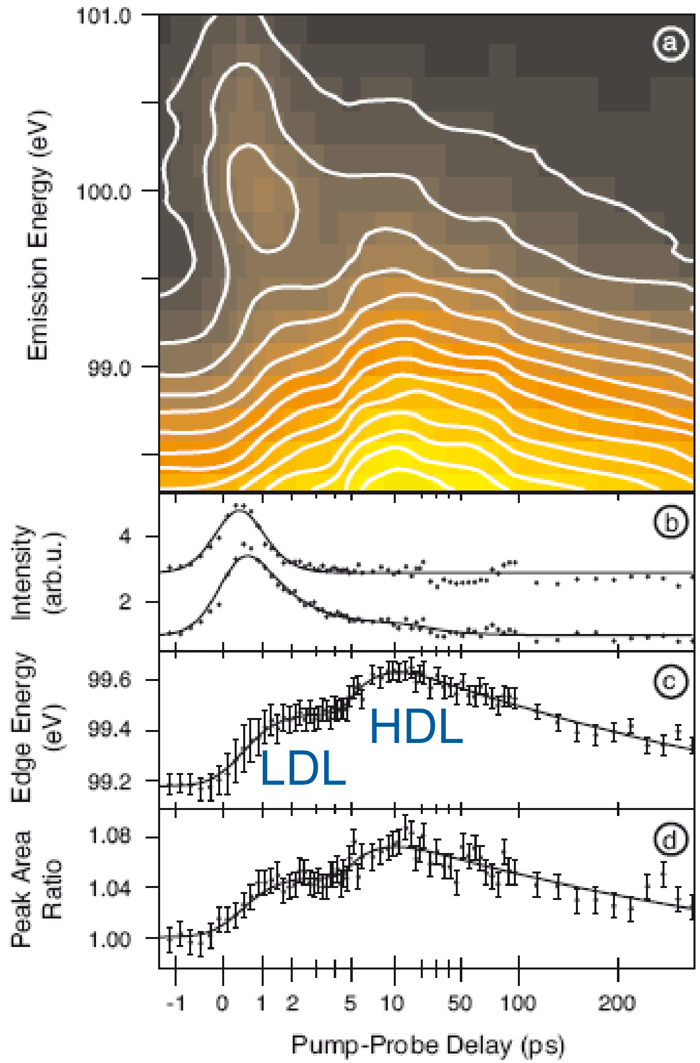
$\Delta t = h/\Delta E = E/\Delta E * \lambda/c$

The role of time resolved RIXS at FELs: Transient phases

The liquid-liquid phase transition in silicon revealed by snapshots of valence electrons
M. Beye, F. Sorgenfrei, W. F. Schlotter, W. Wurth, A. Föhlisch,
PNAS 2010 107 (39) 16772-16776







Transient photoinduced phases: a made table for hRIXS

nature materials — LETTERS

PUBLISHED ONLINE: 16 JANUARY 2011 | DOI:10.1038/NMAT2929

Transient photoinduced 'hidden' phase…

PRL 107, 036403 (2011) PHYSICAL REVIEW LETTERS week ending 15 JULY 2011

…Melting of a Charge Density Wave in $TiSe_2$

E.

PRL 106, 217401 (2011) PHYSICAL REVIEW LETTERS week ending 27 MAY 2011

…of Antiferromagnetic Order in $La_{0.5}Sr_{1.5}MnO_4$ Measured Using Ultrafast …Diffraction

…abhakaran,[1] …leri[1,3,†]

" las… exp…

nature

Cont… mod…

Matteo Ri… Robert W.

" …resonar… system … phase."

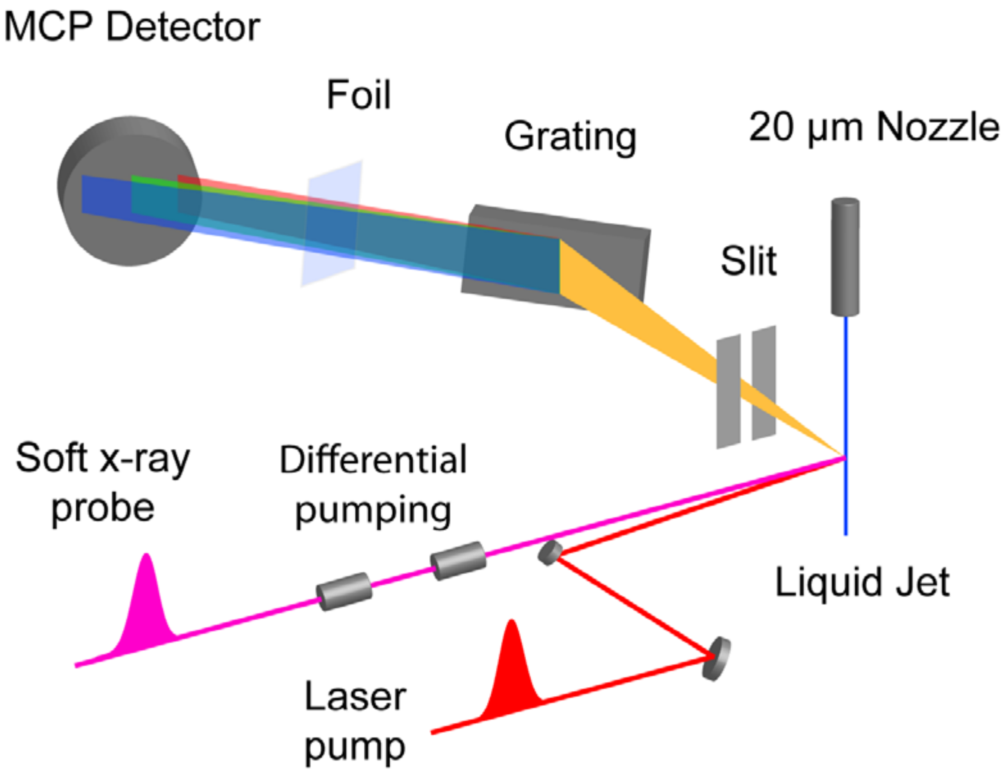
APPLIED PHYSICS LETTERS 98, 182504 (2011)

Time-resolved resonant soft x-ray diffraction with free-electron lasers: Femtosecond dynamics across the Verwey transition in magnetite

N. Pontius,[1,a)] T. Kachel,[1] C. Schüßler-Langeheine,[1,2] W. F. Schlotter,[3,4] M. Beye,[1,3] F. Sorgenfrei,[3] C. F. Chang,[2] A. Föhlisch,[1,3,5] W. Wurth,[3] P. Metcalf,[6] I. Leonov,[7] A. Yaresko,[8] N. Stojanovic,[9] M. Berglund,[1,3,5] N. Guerassimova,[9] S. Düsterer,[9] H. Redlin,[9] and H. A. Dürr[4,10]

Resonant soft x-ray diffraction (RSXD) with femtosecond (fs) time resolution is a powerful tool for disentangling the interplay between different degrees of freedom in strongly correlated electron materials. It allows addressing the coupling of particular degrees of freedom upon an external selective perturbation, e.g., by an optical or infrared laser pulse. Here, we report a time-resolved RSXD experiment from the prototypical correlated electron material magnetite using soft x-ray pulses from the free-electron laser FLASH in Hamburg. We observe ultrafast melting of the charge-orbital order leading to the formation of a transient phase, which has not been observed in equilibrium. © 2011 American Institute of Physics. [doi:10.1063/1.3584855]

Liquid phase femtosecond chemistry from time resolved RIXS
HZB, UP: K. Kunnus, P. Wernet, M. Beye, S. Schreck, A. Föhlisch
MPIbpC: S. Grübel, W. Quevedo, I. Rajkovic, M. Scholz, C. Schmidt, S. Techert
MaxLab: B. Kennedy, F. Hennies (Martensson/Nordgren/Rubensson)
SLAC: D. Nordlund, K. Gaffney, R. Harsock, W. Zhang, B. Schlotter, J. Turner
Stockholm U: Ida Josefsson, Michael Odelius (Theory)
Utrecht U: Frank de Groot (Theory)



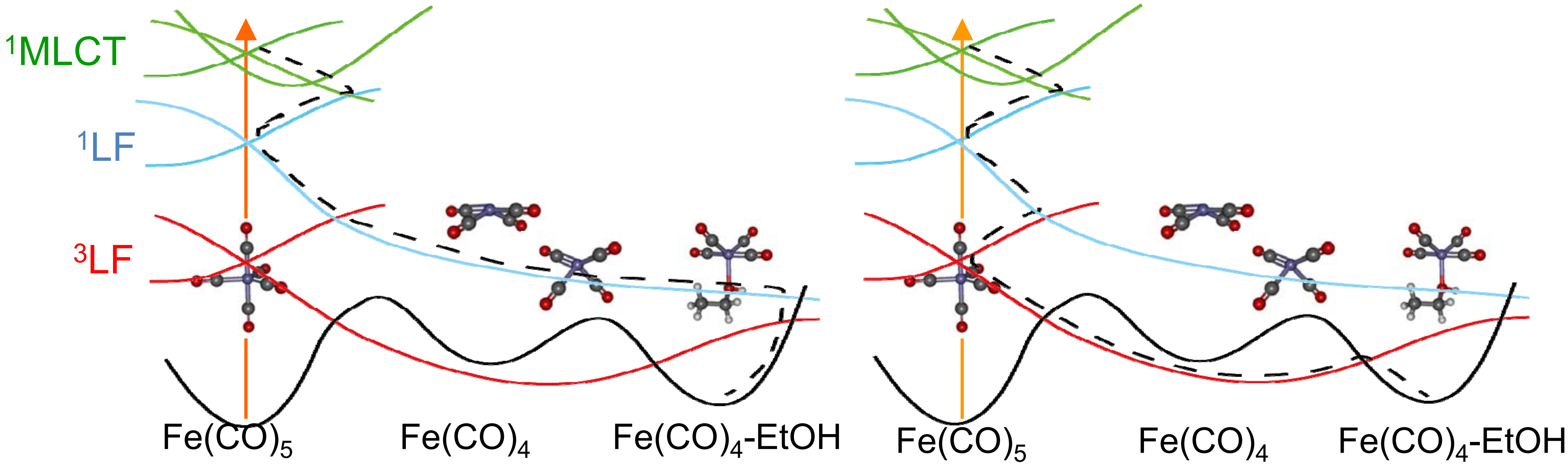
At HZB BESSY:
• FlexRIXS users operation

At LCLS:
• Liquid Jet Endstation SXR endstation with users operation

Scenarios of $Fe(CO)_5$ photodissociation in solution

Concentred ligand substitution reaction via singlet pathway
Ahr, Rose-Petruck, et al.,
Phys. Chem. Chem. Phys. 2011, 13, 5590.
Trushin, Fuss, et al.,
J. Phys. Chem. A 2000, 104, 1997.

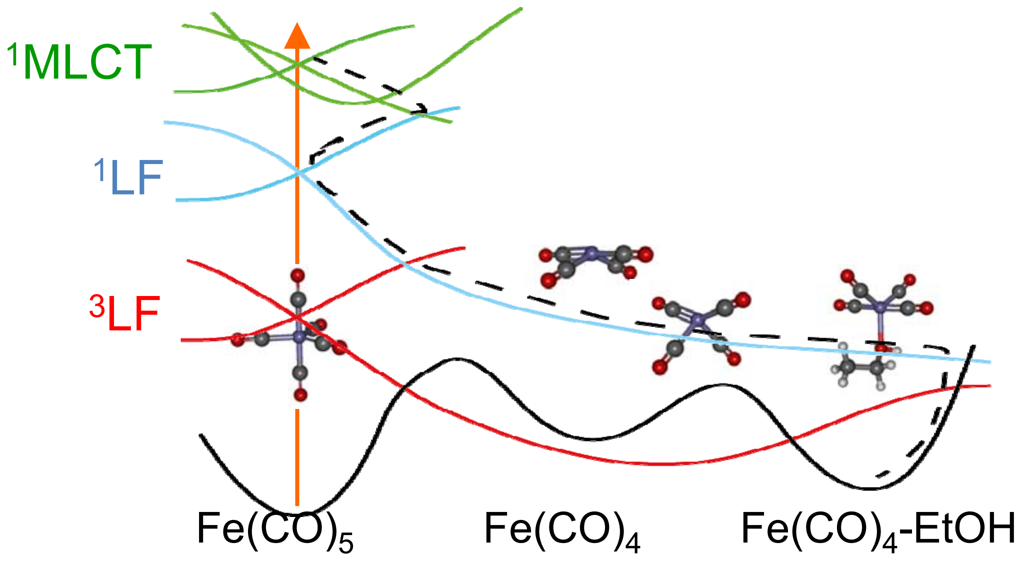
Reaction via triplet intermediate and diffusion limited complexation
Snee, Harris, et al., JACS. 2001, 123, 6909.
and JACS 2001, 123, 2255.



Simplified schemes!
More then one nuclear coordinate involved!

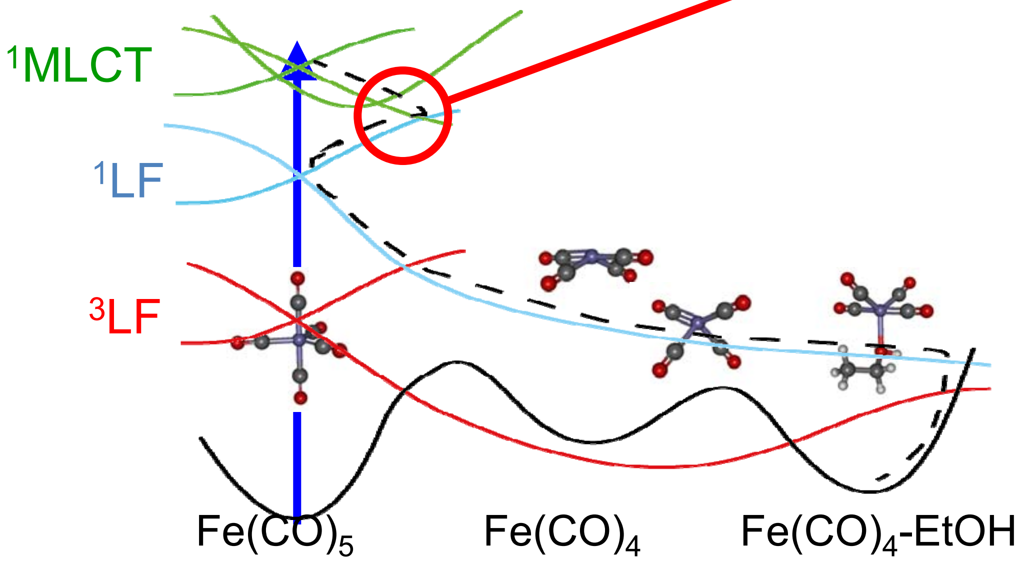
Time-resolved $Fe(CO)_5$ Fe-L-edge RIXS in a nut-shell

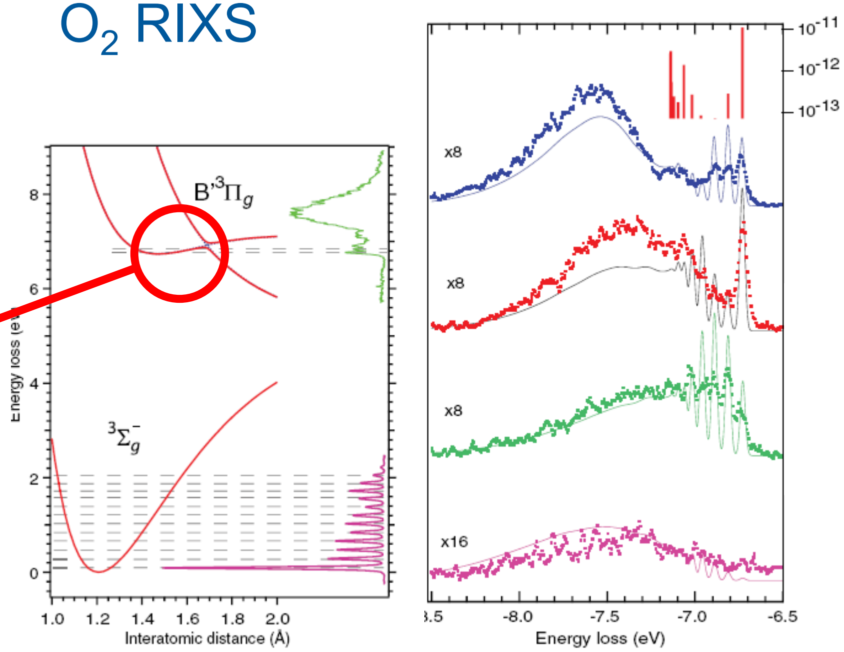
Concentred ligand substitution reaction via singlet pathway
Ahr, Rose-Petruck, et al.,
Phys. Chem. Chem. Phys. 2011, 13, 5590.
Trushin, Fuss, et al.,
J. Phys. Chem. A 2000, 104, 1997.



At hRIXS: get to the (photo-) chemically relevant curve crossings/ transition states!

PRL **104**, 193002 (2010)
O_2 RIXS





Proposed hRIXS at XFEL.EU

Spectrometer

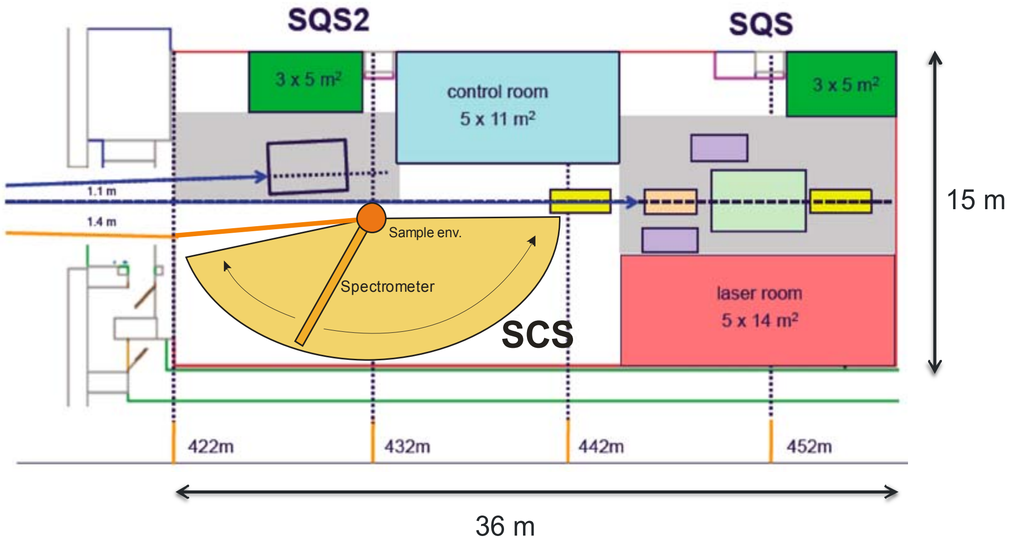
- Large acceptance (to avoid radiation damage)
- Multi-color RIXS option (Strocov, Chen or RZP layout)
- Minimum 5 m arm length
- combined resolution: 30,000 @ Cu-L3
- detection angle continuously variable

Sample environment flexible

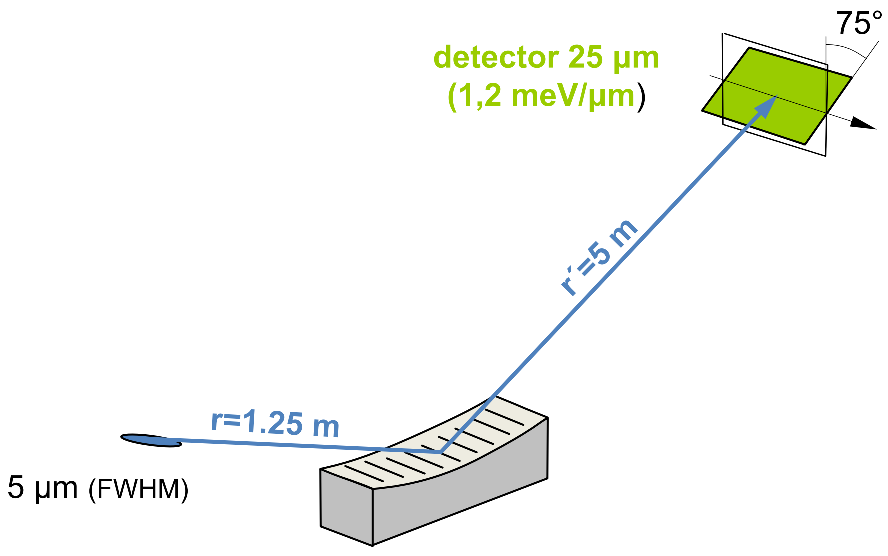
- UHV with ARPES-like cryo-manipulator for variable momentum transfer
- Liquid phase and gas phase for chemistry

Beamline extension

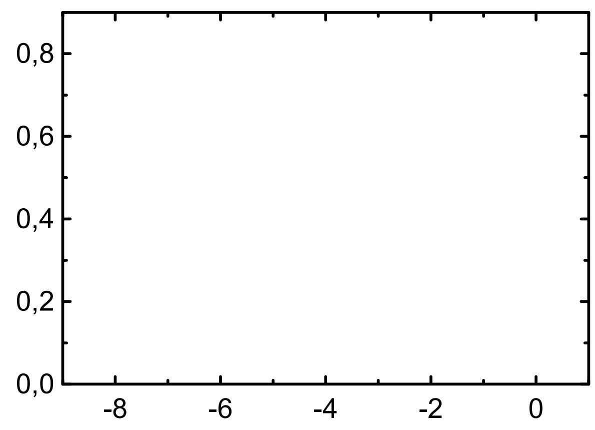
- Timing-conservation mono: Low line-density grating, various RZPs?
- Zero-order option (highest temporal resolution with XES)

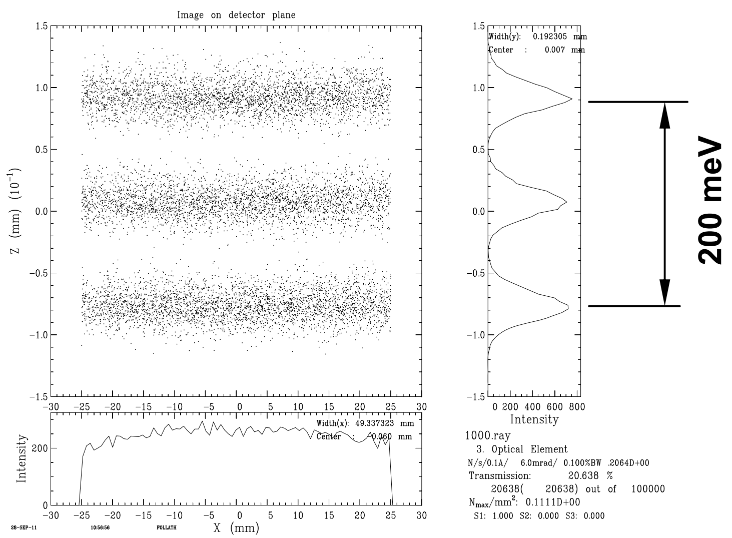


What is the ideal design for XFEL: A compact VLS-spectrometer?



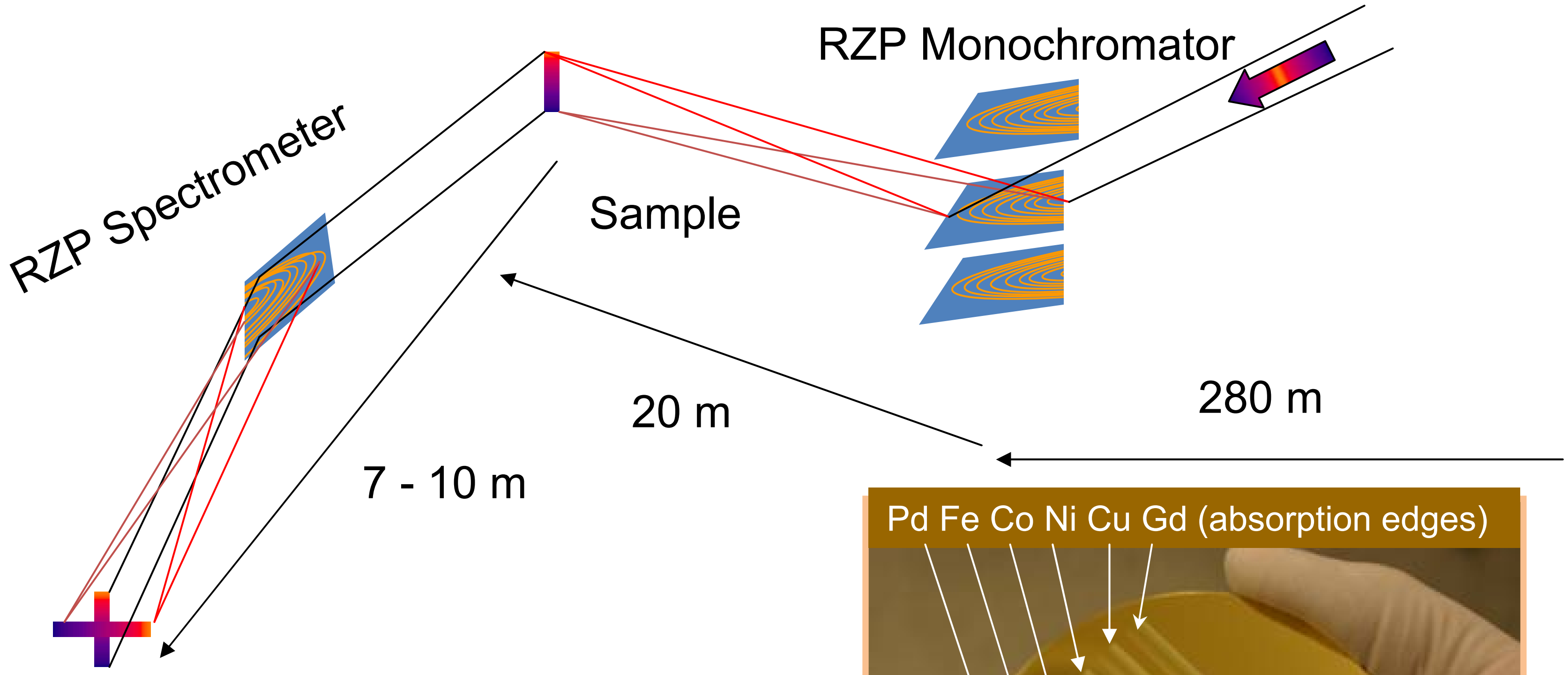
R_G = 108 m, L=150 mm
N = 3200 l/mm
B_2 = 1,14e-4 1/mm
B_3 = 1.06e-8 1/mm^2
σ = 0.25 µrad

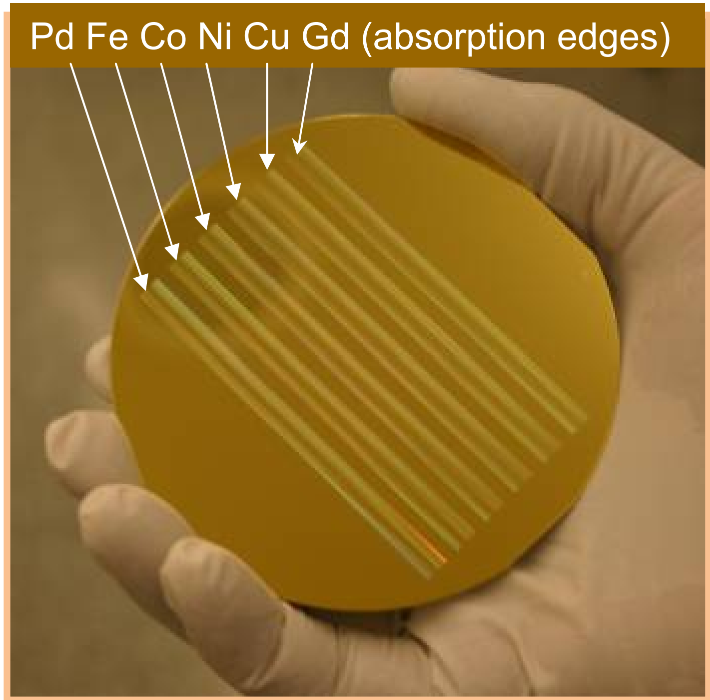




Bandwidth ≈ 30 meV @ 1000 eV
E/ΔE=33000
Acceptance 4 x 2 mrad^2

What is the ideal design for XFEL : Double dispersive?



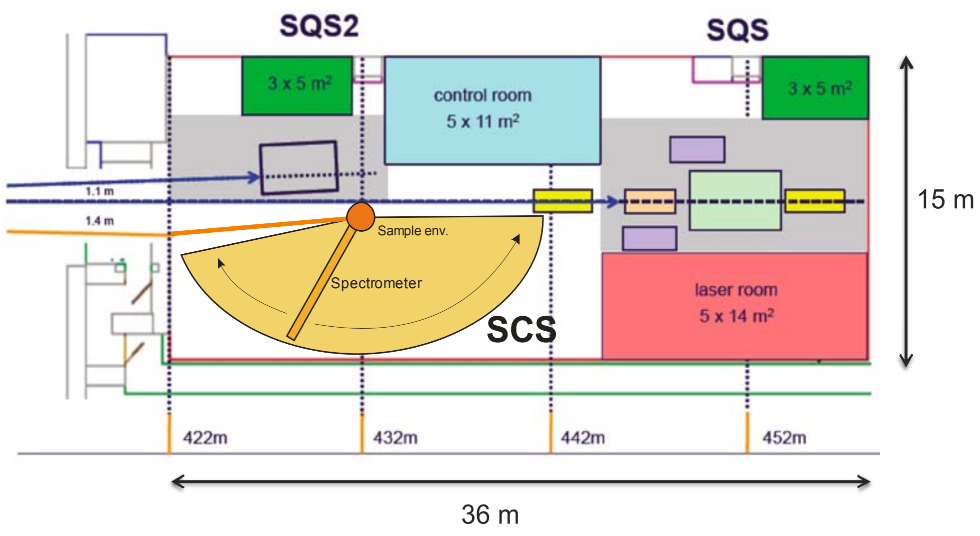


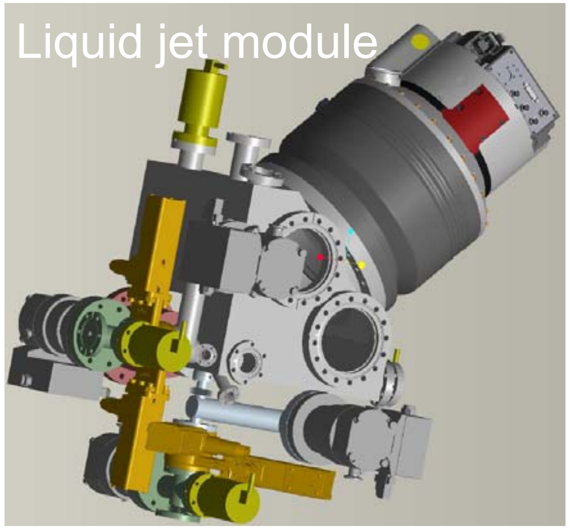
Monochromator + Spectrometer:

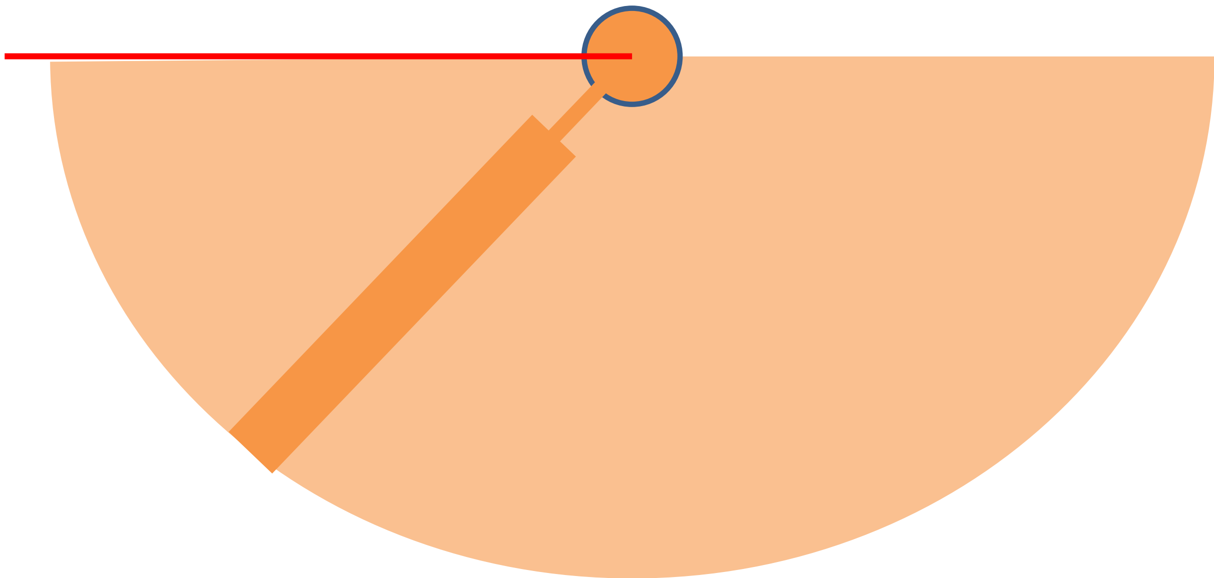
Energy resolution: 40 000 @ 1keV

Time optimized

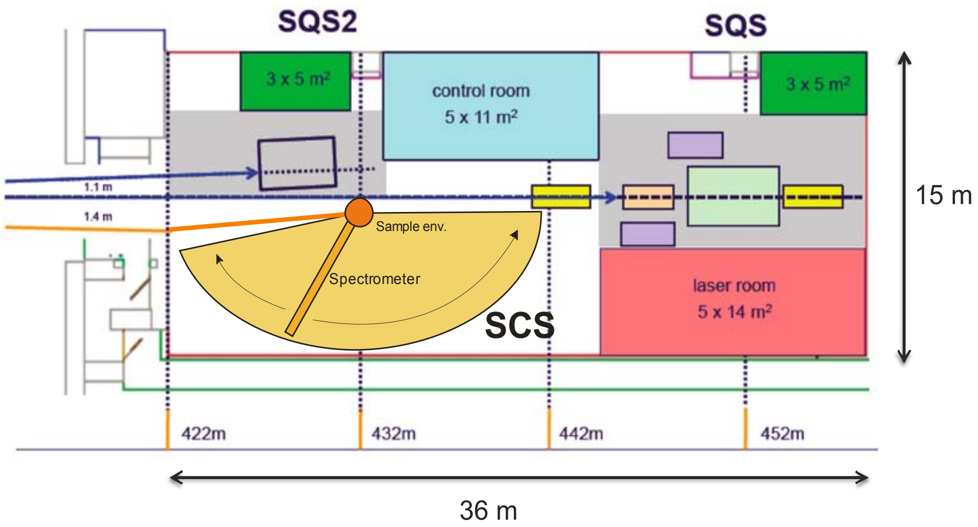
Flexible sample environment

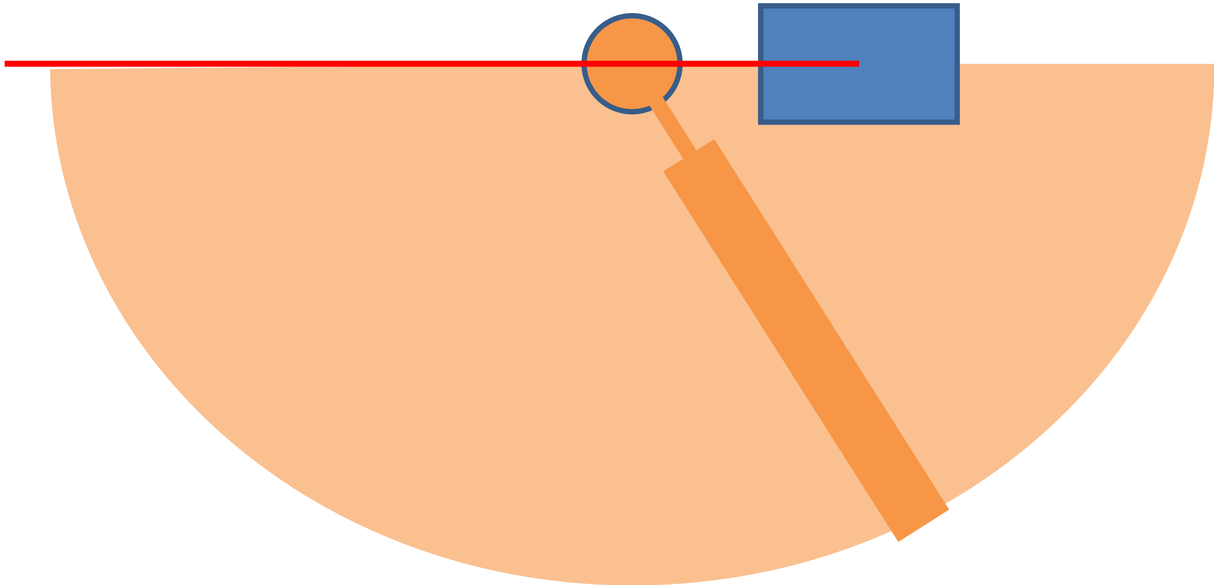




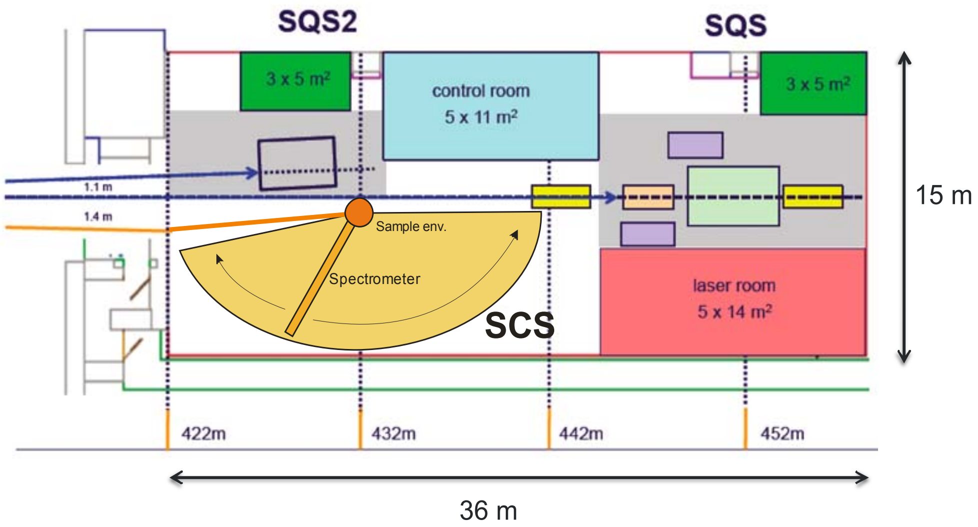


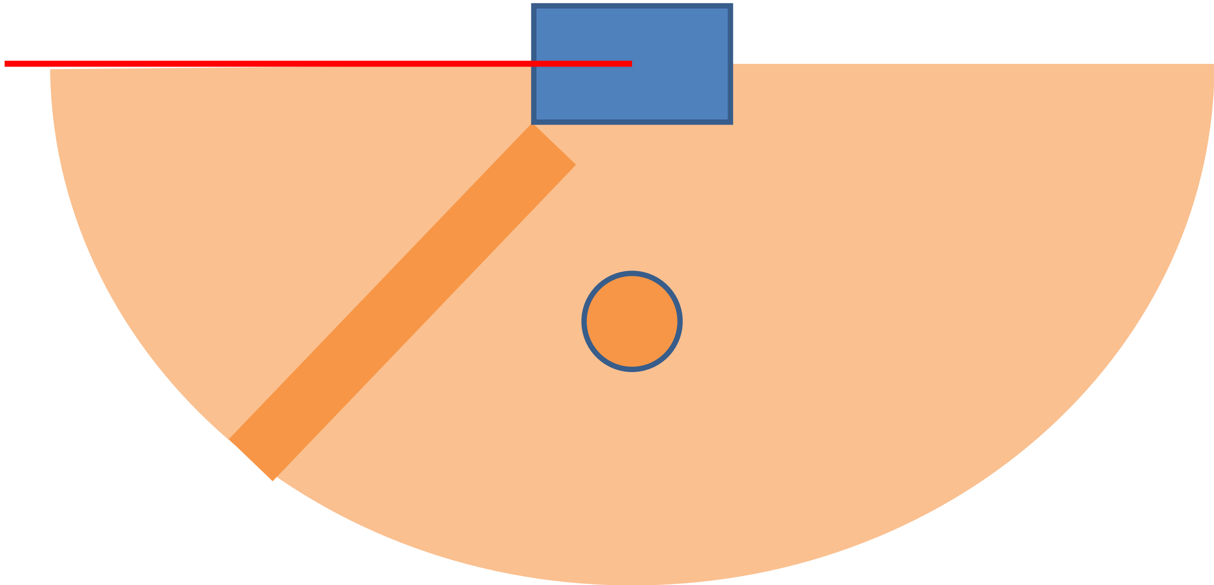
Option 1: Shoot-through geometry for other experiments





Option 2: Replace experiment in interaction point





Integration into the XFEL SCS infrastructure

Techniques for SASE 3

- Elastic scattering/diffraction, NEXAFS/XANES
- Photoemission
- Coherent small-angle scattering/Imaging
- Coherent wide-angle scattering

Flexibility

- Shoot through
- Remove sample chamber and rotate detector arm out of the way

Technique	Integrated	Separate
Diffraction	●	
Photoemission		●
Coherent SAXS	●	●
Coherent WAXS		●
NEXAFS/XANES	●	

Funding

Budget estimates	
Spectrometer	1,5 Mio
Beamline extension	0.8 Mio
Personal	2 Postdocs, 1 Engineer
Total	**2.9 Mio**

Positive Reccomendation from XFEL.EU SAC.
SCS Scientist, 1.2 Mio.

Detectors ?

We need to figure out how to bring in the ressources:

1) Call in Germany
2) Sweden/in kind?
3) Italy Giacomo Ghiringelli (PostDoc)
4) Broaden membership base

Full hRIXS consortium Proposal due March 25. 2012
Get design more final.

Summary

Strong scientific case: go far beyond present frontiers in one of the most powerful spectroscopies

- fundamental low-energy coupling mechanism in materials
- full analysis of momentum and polarization dependence
- chemical dynamics
- transient phases
- Non-linear and stimulated processes

Highly qualified user consortium with

- proven track record in complex large scale instrumentation projects
- strong research interests
- well embedded in the user community

Thank you for your attention!