Prospects on stimulated x-ray Raman scattering in the gas phase with XFEL radiation

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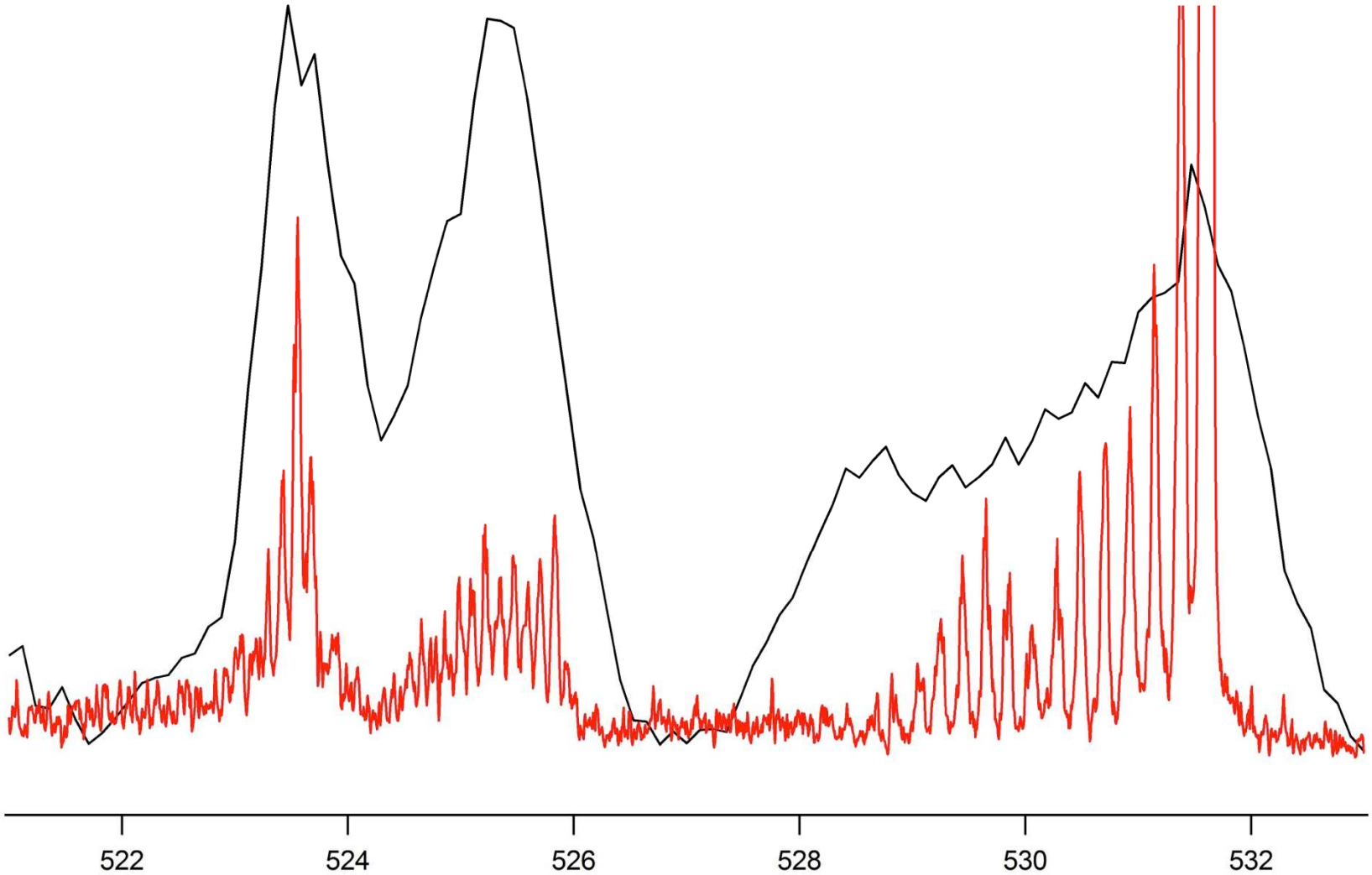
- Dilute Samples
- Exotic excitations
- Time-resolved RIXS
- Multiphoton excitation-RIXS
- **Stimulated RIXS**

Le Petit Prince

Mon dessin ne représentait pas un chapeau. Il représentait un serpent boa qui digérait un éléphant


522
524
526
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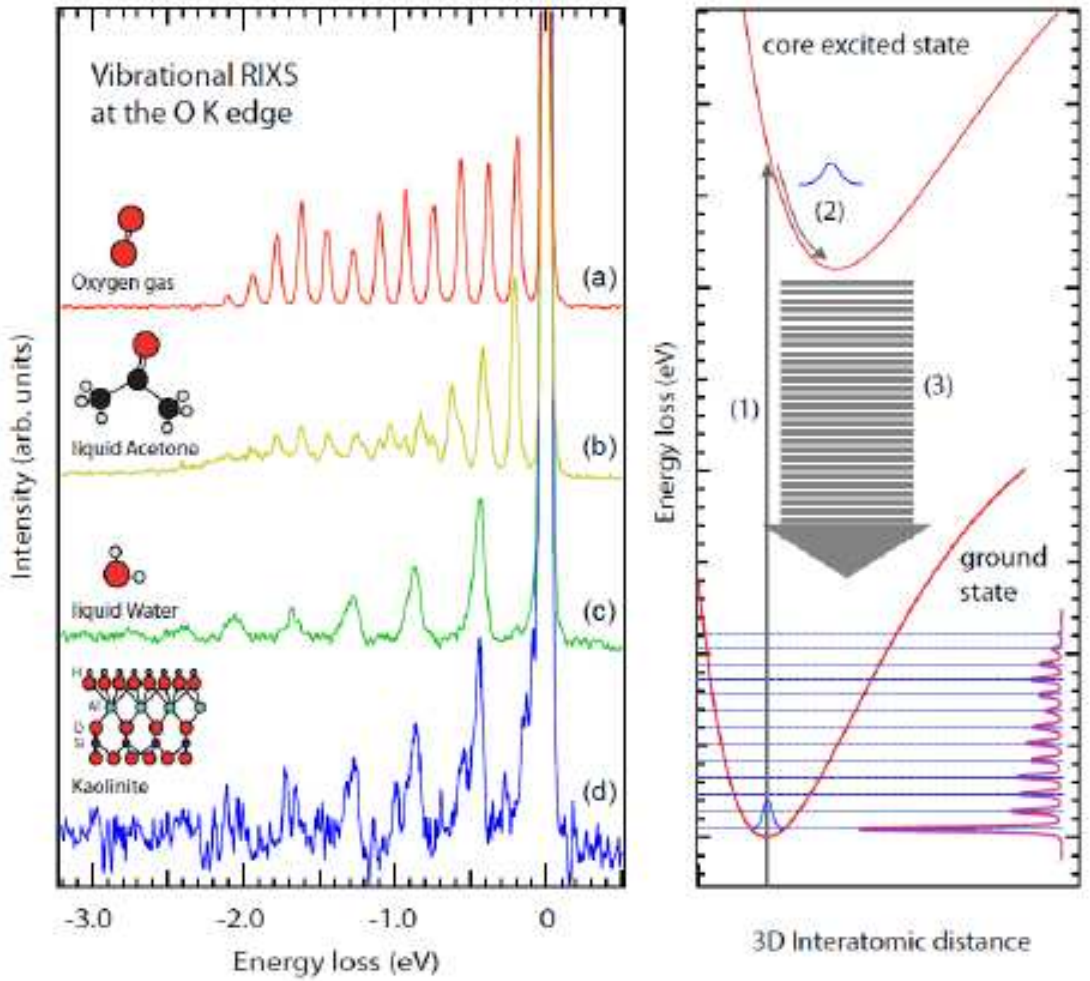
532

Molecular Materials and Processes

Mapping of local potential surfaces

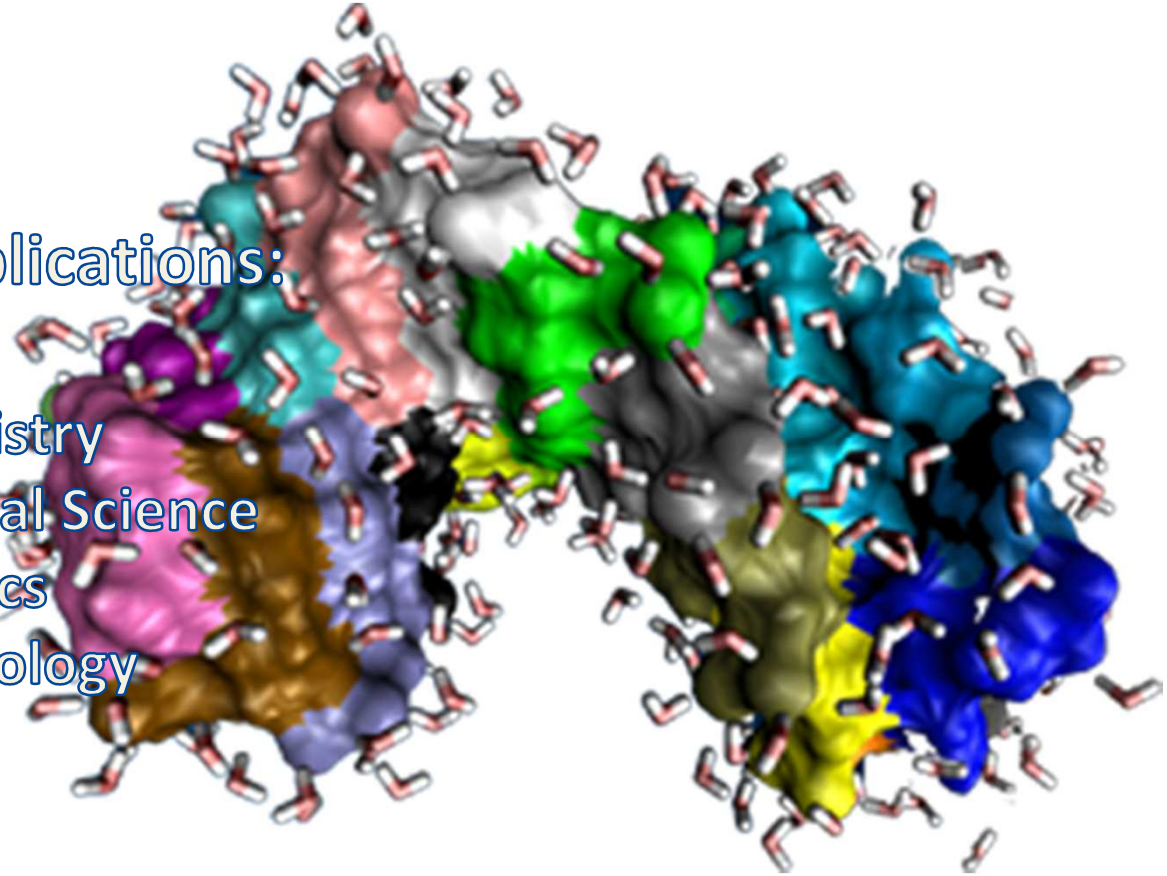


El Dorado also for linear AMO

Dilute samples require more intensity

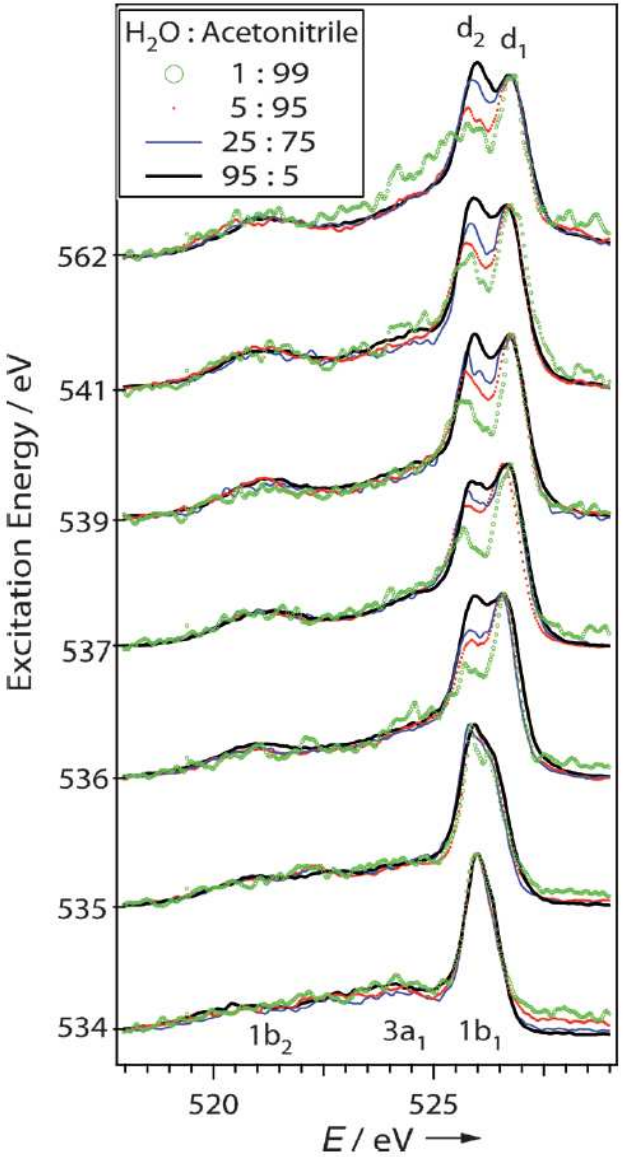
Lots of Applications:

Electrochemistry
Environmental Science
Pharmaceutics
Molecular Biology



From http://www.iiserpune.ac.in/~arnabm/

Water in acetonitrile
K. Lange et al., Angew. Chem. Int. Ed. 2011, 50, 10621 –10625



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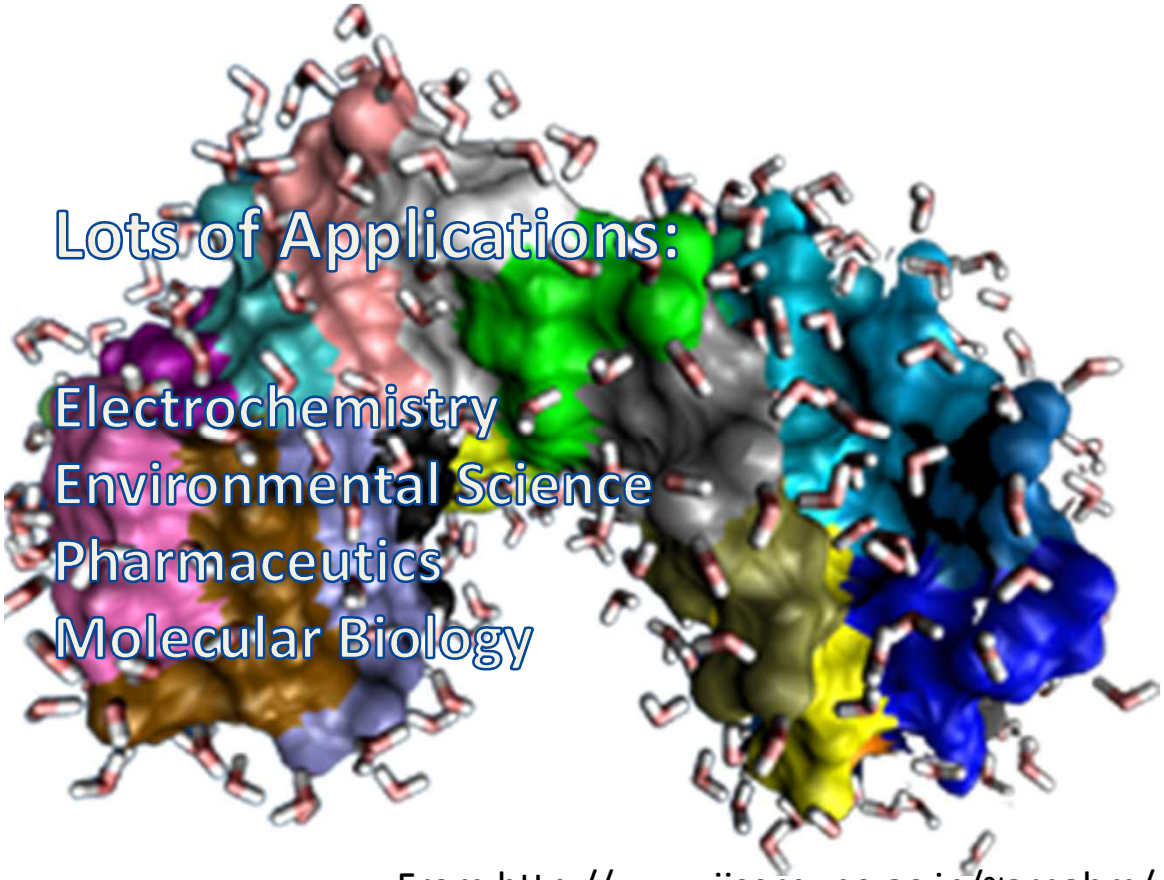

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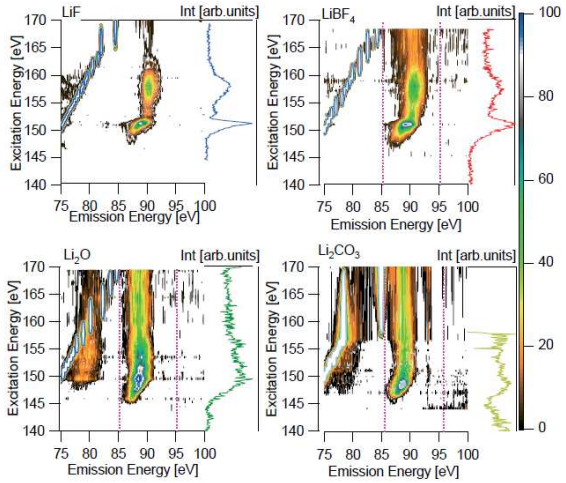
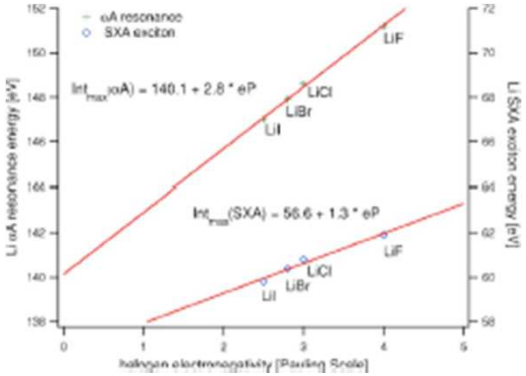
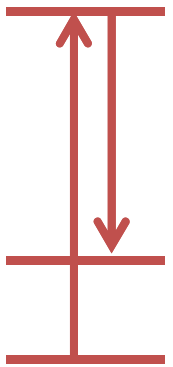
Exotic Excitations require intensity:
Chemical Sensitivity of Double Core Hole Resonances





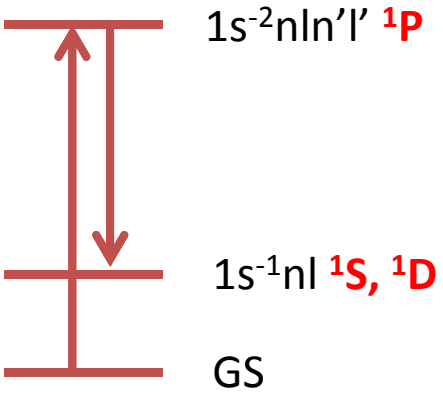


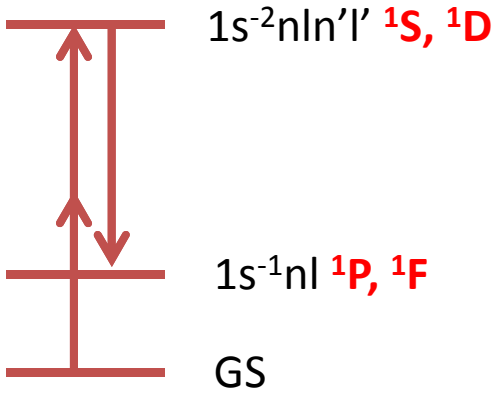
- Dipole Selection
- No Influence of Plasma Potentials

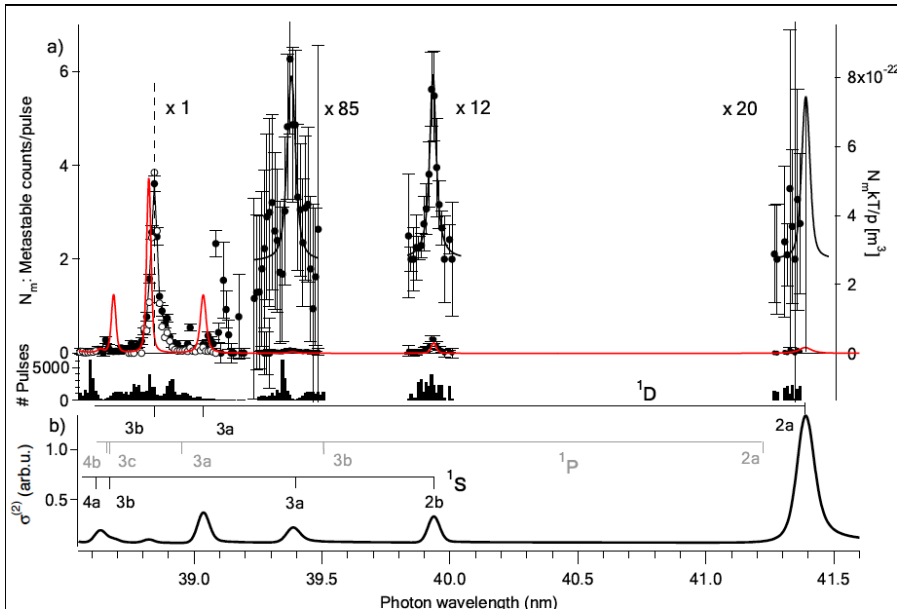
Investigation for lithium compounds in the thesis of Marcus Agåker, PRL93, 016404 (2004), PRB73, 245111 (2006), PRB74, 241105 (2006), PRB75 045112 (2007)

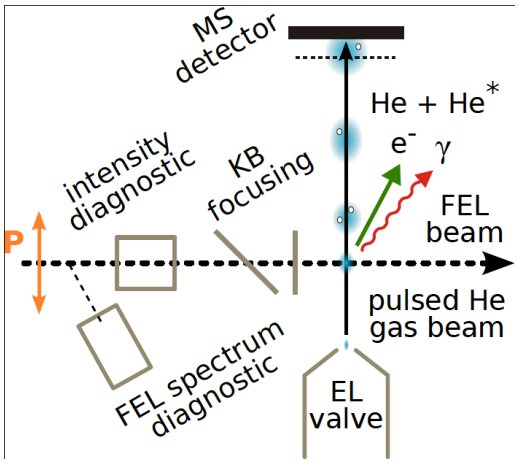
Scanning two-photon resonances

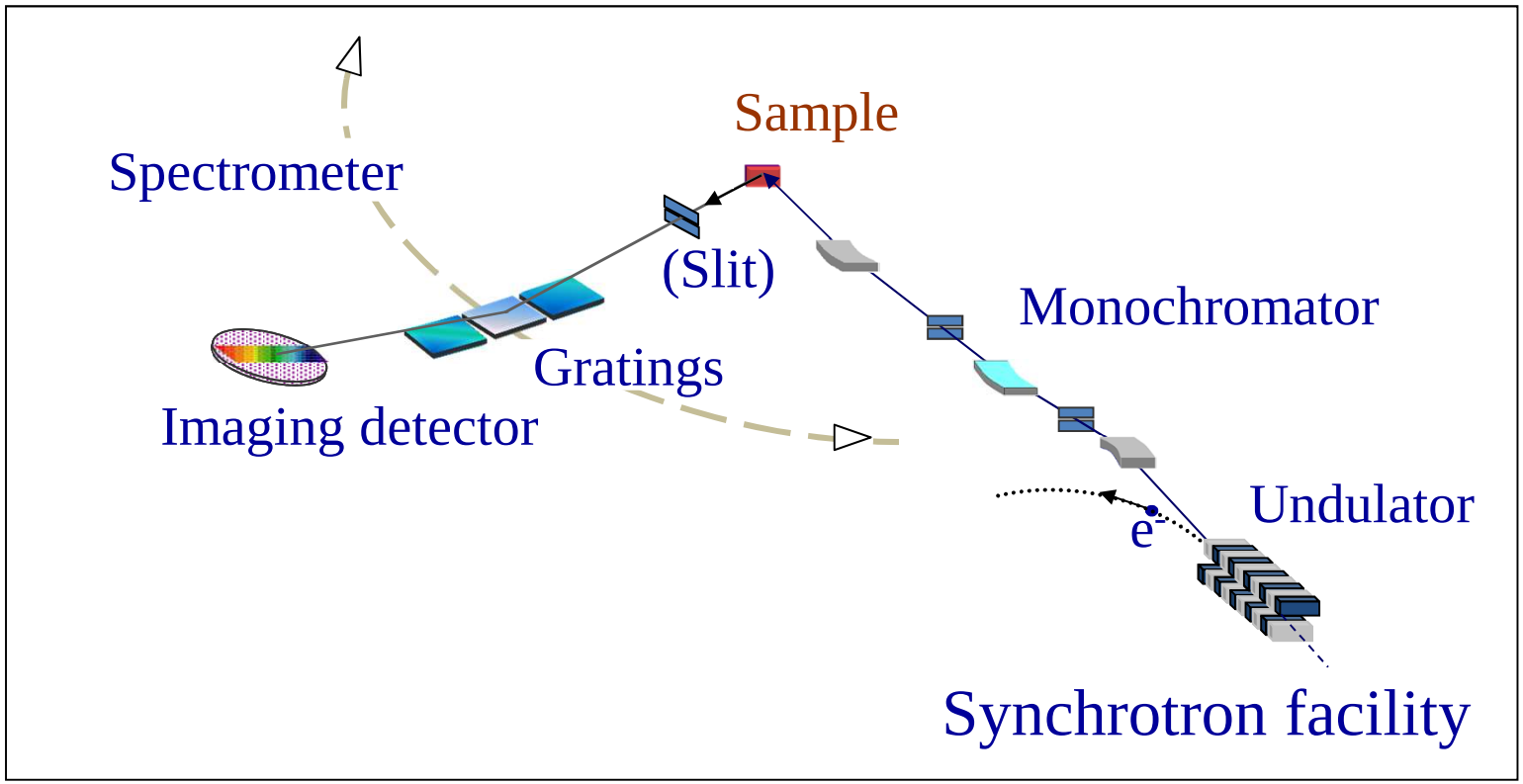
Collaboration with Matjaz Zitnik et al., JSI Ljubljana, the FERMI crew, UPMC Paris, MAX IV, Lund, European XFEL, Hamburg



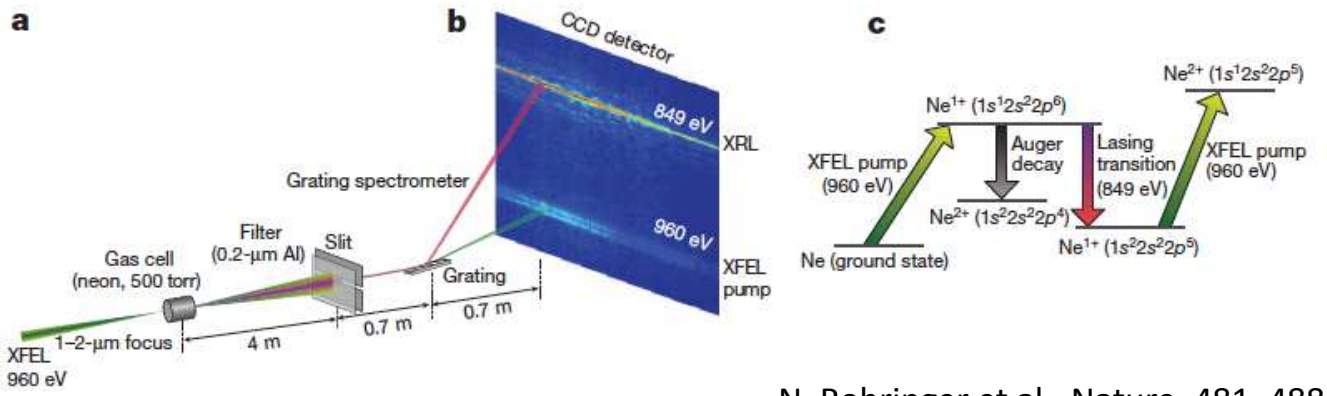








Classically: low fluorescence yield, almost isotropic emission….

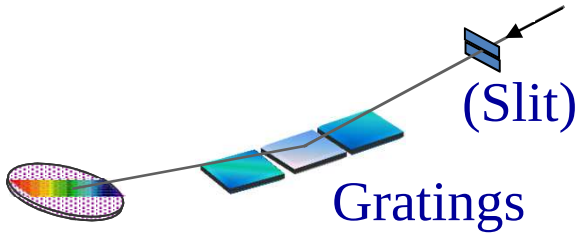


N. Rohringer et al., Nature, 481, 488 (2012)

Stimulated RIXS “promises” a brilliant source of secondary emission loaded with physical information:

End of compromises between count rate and resolution!

Photon-in-photon-out: spectroscopic technique maintaining high resolution and selectivity in high fields

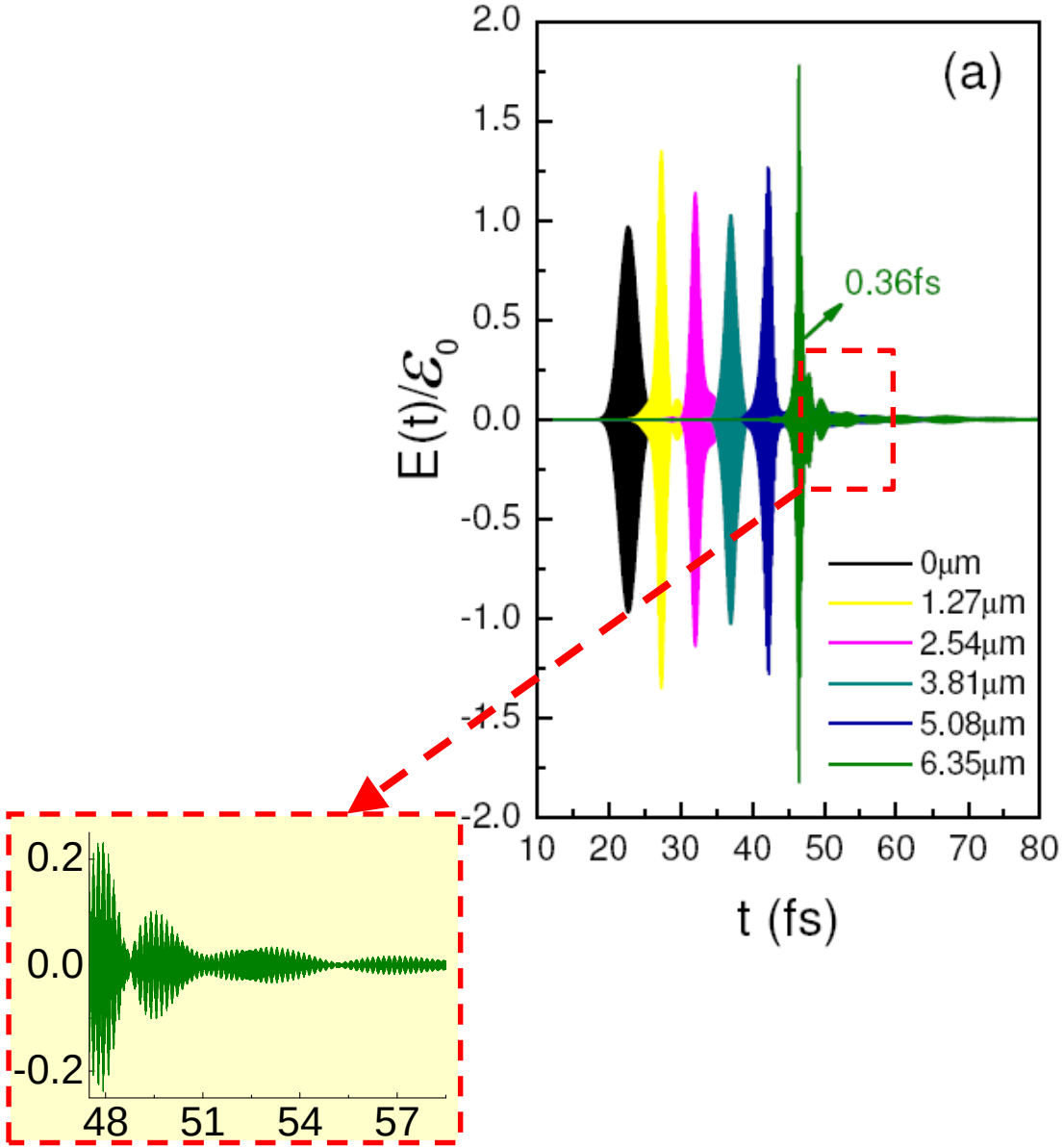


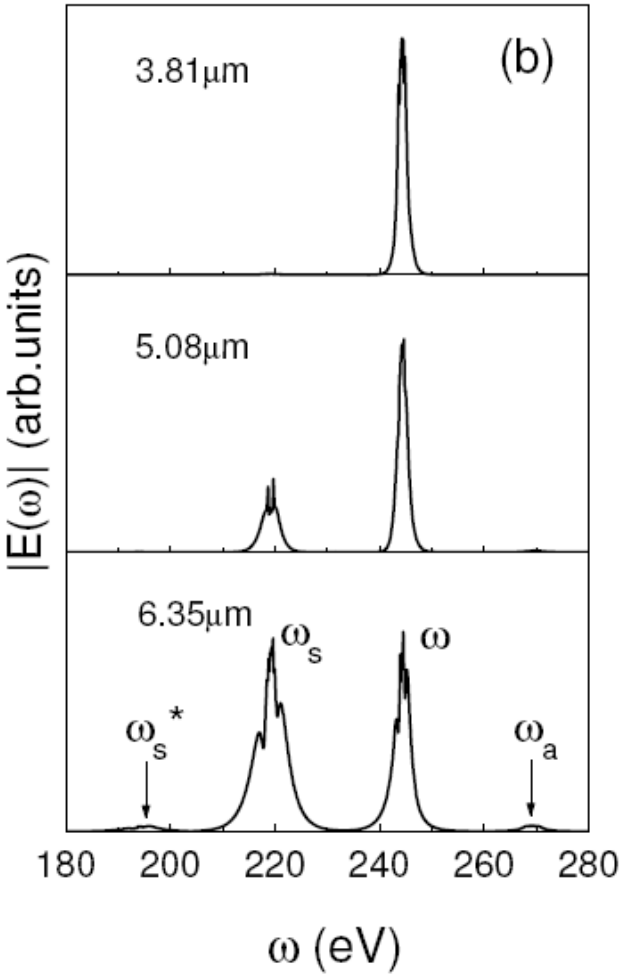
Conventional spectrometer in the forward direction--- no compromises:

- small slit
- high groove density
- high orders
- large radius

Alternative beamline-type schemes, where eventually the slope errors of the optics set the limit.

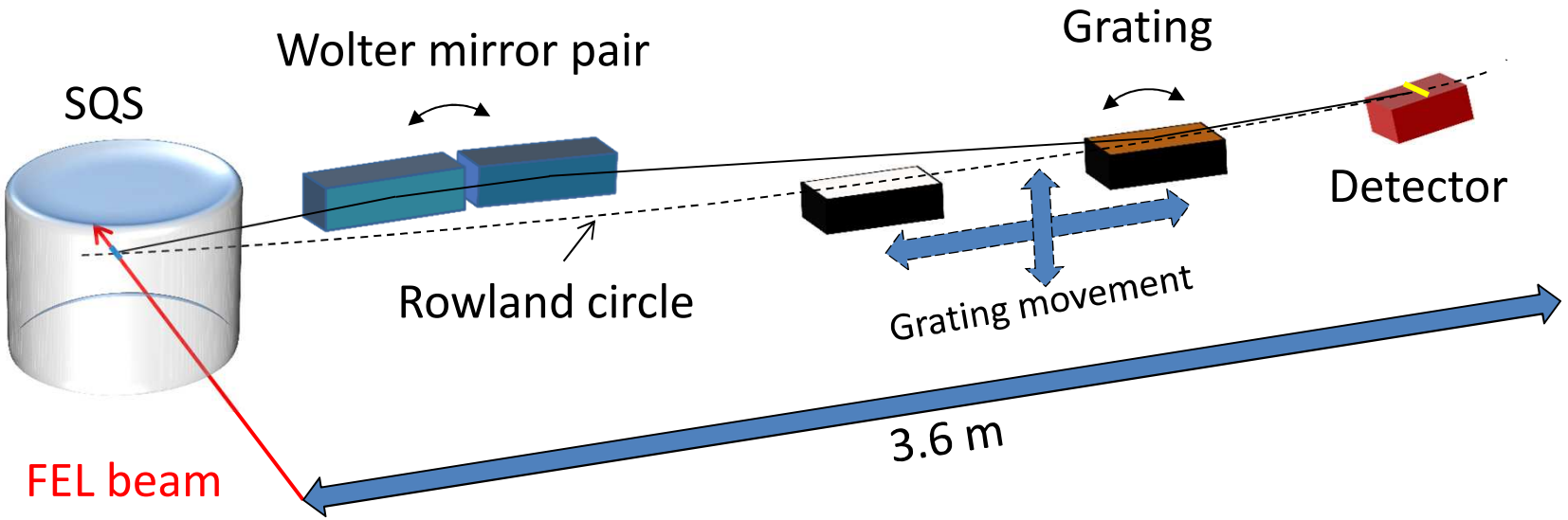
Pulse compression, Burnham-Chiao modulation and four wave mixing



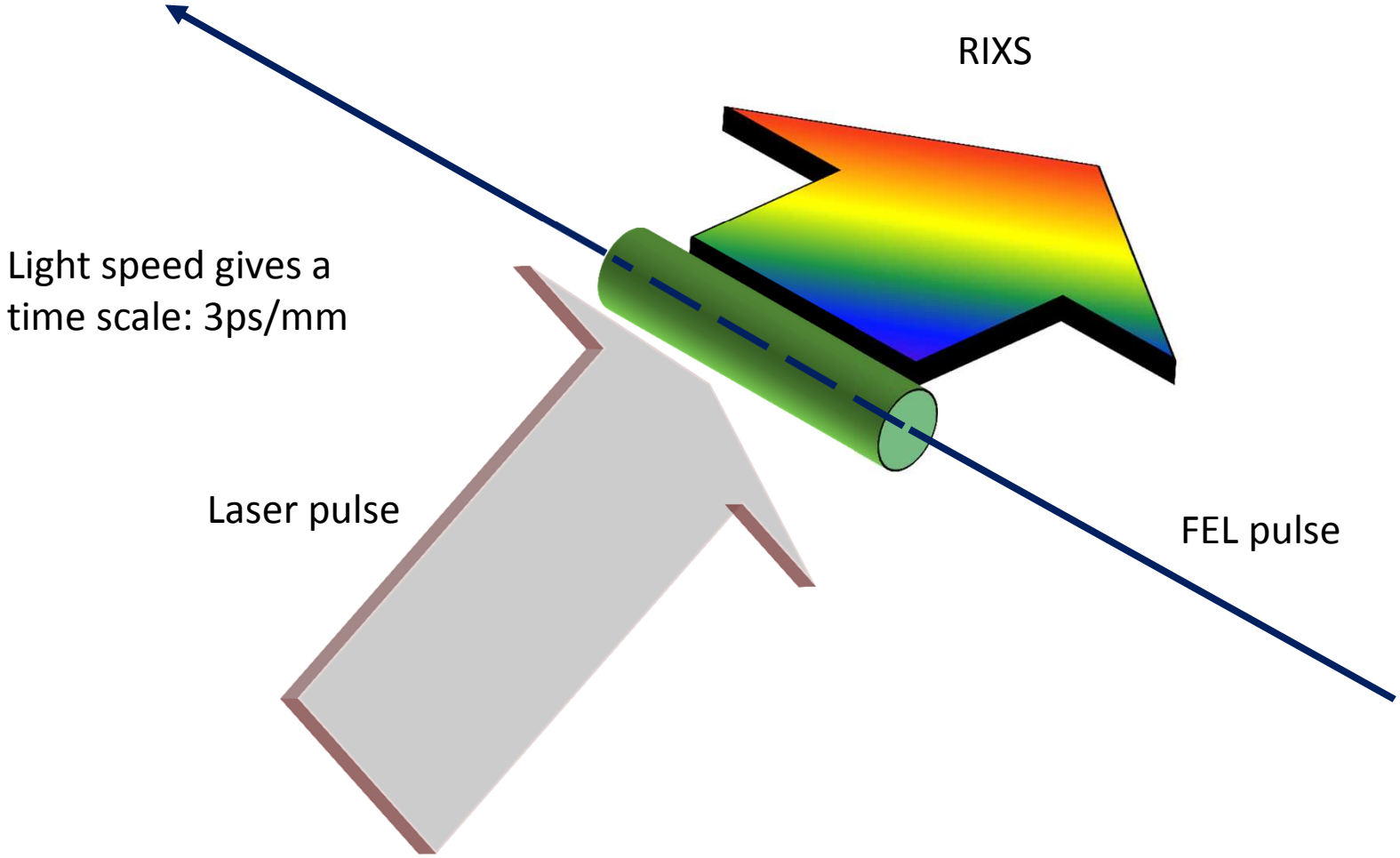


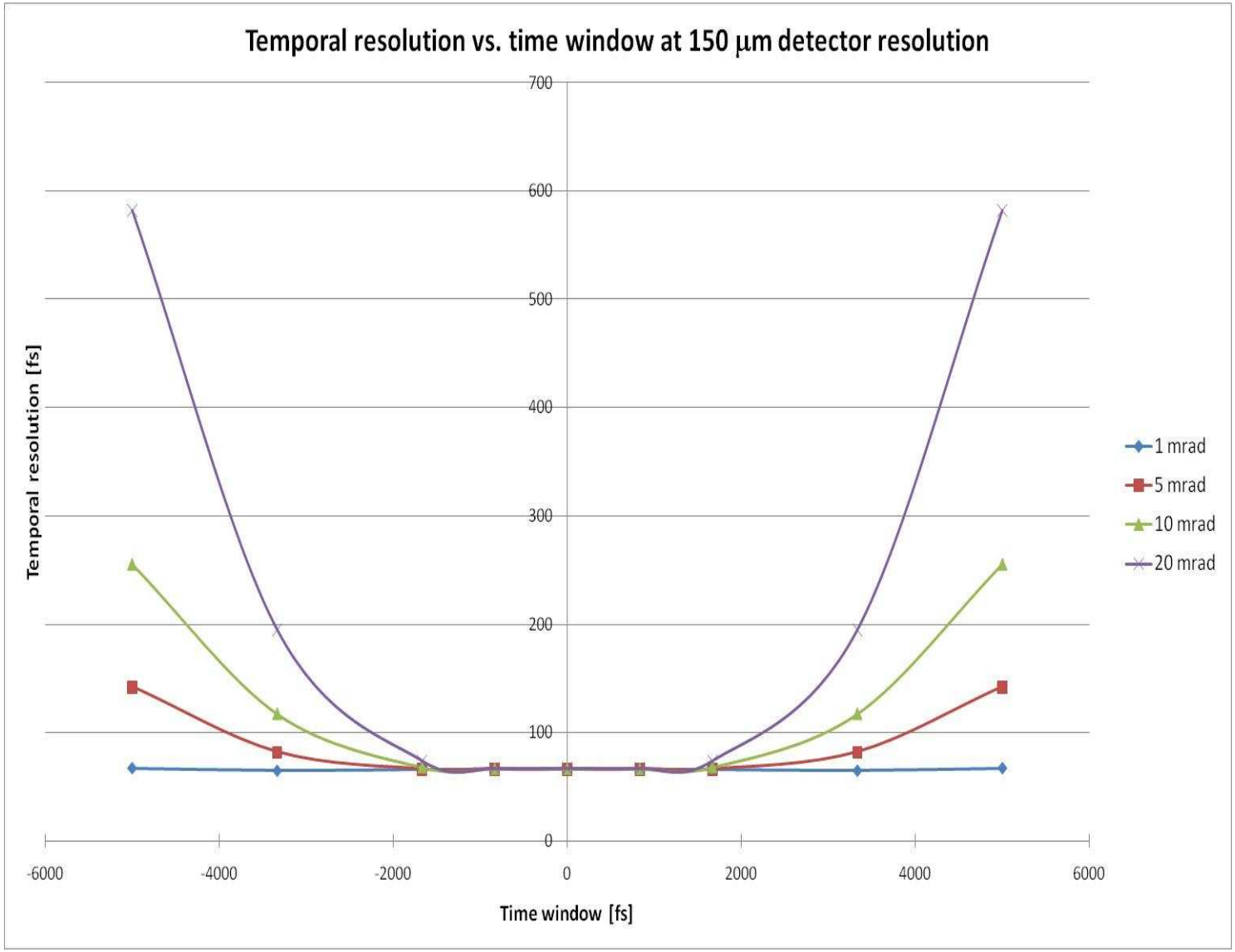
Phys Rev A 81 (2010) 013812
Phys Rev A 81 (2010) 043412
EuroPhys.Lett. 87(2009)64002
J. Phys.B:At.Mol.Opt.Phys. 42(2009)201001
Ph D Thesis of Y.-P. Sun 2011

Outline of 1-D Imaging RIXS Spectrometer



Pump-probe RIXS

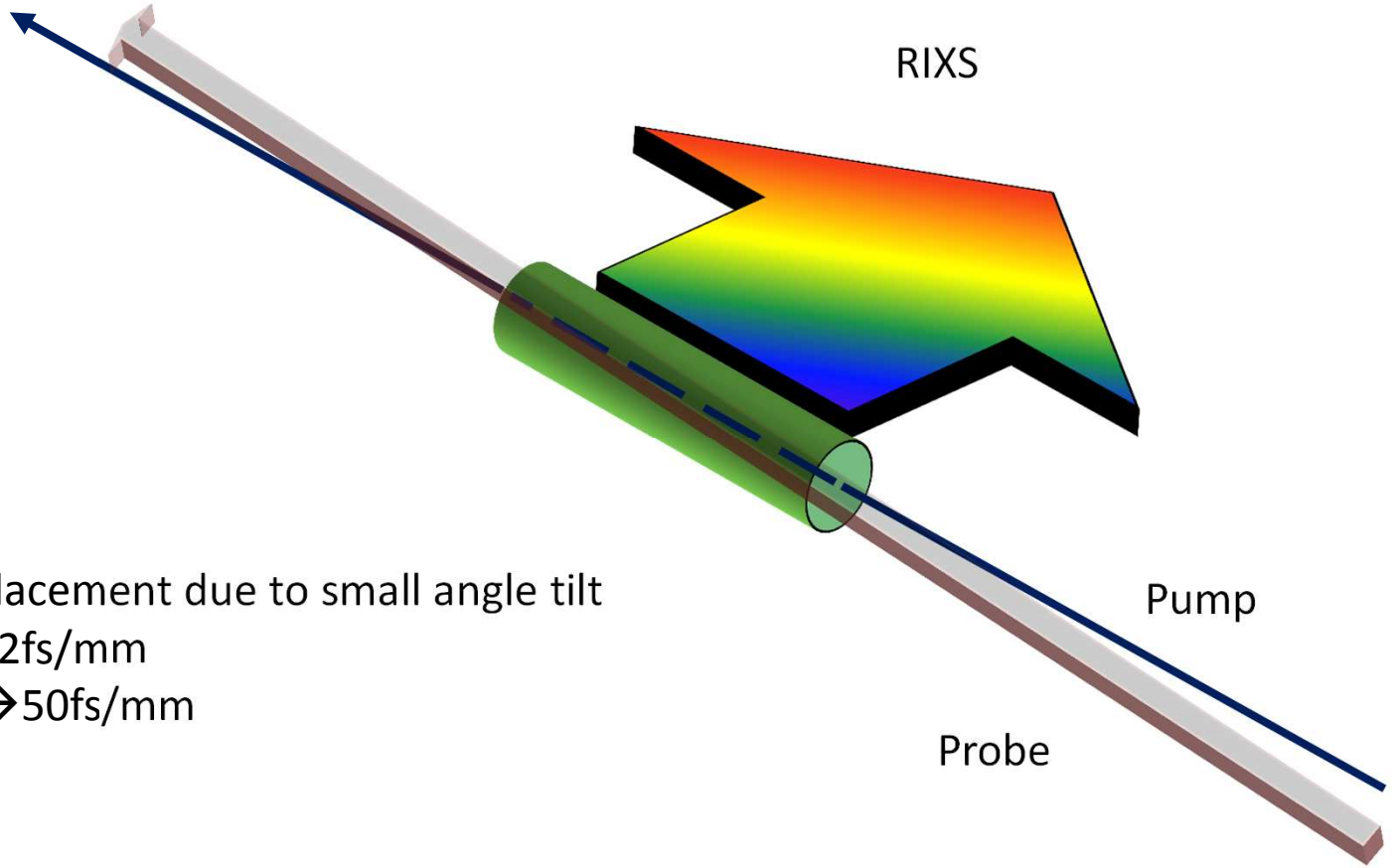



Temporal resolution vs. time window at 150 μm detector resolution
Temporal resolution [fs]
700
600
500
400
300
200
100
0
-6000
-4000
-2000
0
2000
4000
6000
Time window [fs]
1 mrad
5 mrad
10 mrad
20 mrad

For the split and delay

- Dynamics within the core-hole lifetime
  - ✧ Stimulation-pulse delayed and energetically displaced up to around 30 eV.
  - ✧ Delay scan up to around 10 fs.
- Ultrafast energy dissipation in large (bio-relevant) molecules, following core ionization.
  - ✧ Delay scan up to around 150 fs.
- For core-hole induced chemistry, delays on the picosecond scale is interesting. On long timescales primarily “conventional” laser pump is relevant

Pump-probe experiments with RIXS



Displacement due to small angle tilt
2^{o}→2fs/mm
10^{o}→50fs/mm

Parameters Wish List

	Day 0	Nice to have
Experimental techniques	Time-resolved and stiumulated RIXS	
Source properties		
Energy range	From N K and up to (say) Al K	Down to C K
Pulse duration	Shorter than typical core hole lifetimes (a few fs) For non-linear processes down to around 1 fs.	
bandwidth	As small bandwidth as possible, without compromising time resolution.	Heisenberg limit would be nice
Device properties		
Maximum Temporal delay	In the range of the core-hole decay times; 0-10 fs. For energy dissipation in large biorelevant molecules following core ionization up to 150 fs. Longer delays are not relevant for X-ray pump/X-ray probe measurements.	
Pulse intensity ratio	Controllable but difficult to predict: A guess is from 50/50 to 1/99	
2 Colors	A difference within a range of 0-30 eV would be sufficient for stimulated RIXS. This is relevant only if the the time delay between the pulses is also controlled.	
Symmetric delay around t=0	Not important	
Spatial separation behind sample	Room to measure spectra in the forward direction.	
Add your suggestions		

Thank you!