What are we interested in ?

➡ Investigating excited state dynamics of species in solution to try to understand how energy moves in these strongly interacting systems

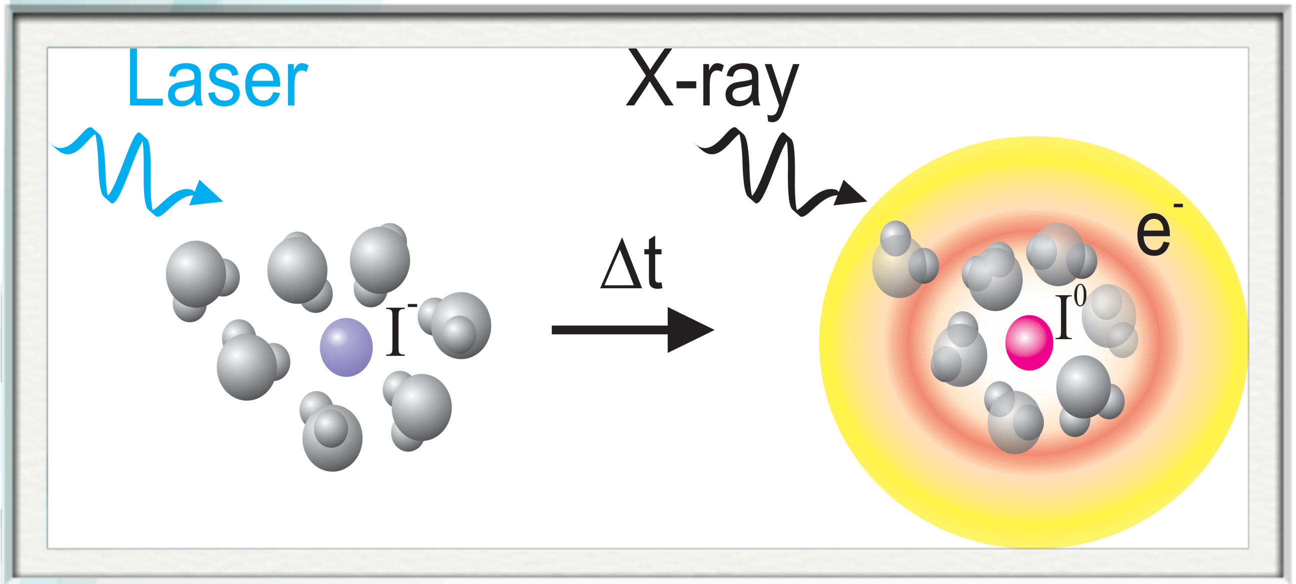
➡ How does the solvent interaction play a role in the relaxation of these systems ?

➡ How does the excitation perturb the structure and how does this structural change affect the energy transfer and relaxation ?

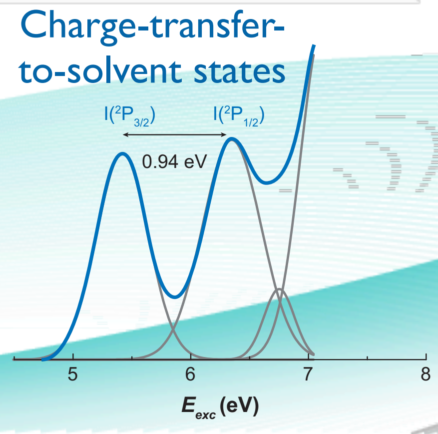
➡ Can we relate this information to functionality ?

Solvation dynamics: aqueous iodide

$$I^- + H_2O \xrightarrow[\text{UV}]{h\nu} I^0 + e^- + H_2O$$



- How does the water respond ?
- What changes if we excite the charge-transfer-to-solvent states ?
- Can we extract electronic information as well as structural ?



Myoglobin: unresolved issues

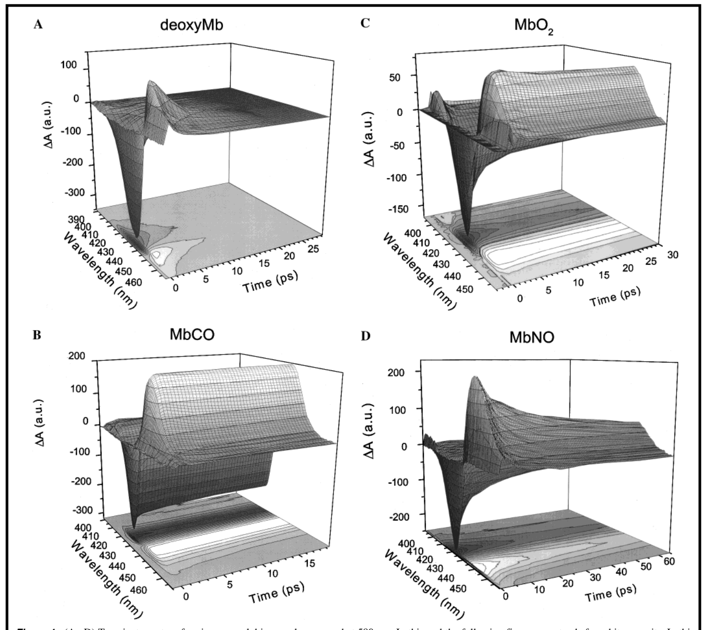
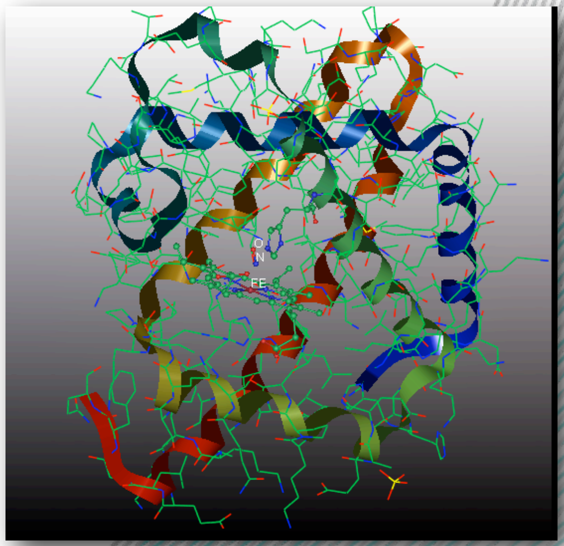
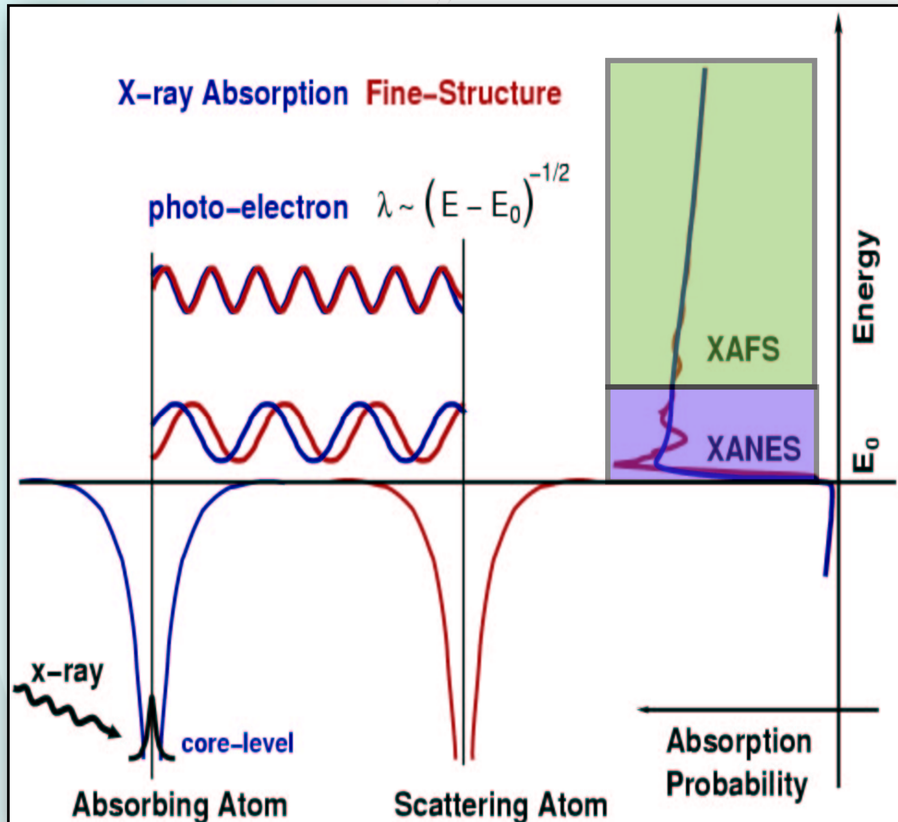


Figure 1. (A−D) Transient spectra of various myoglobin samples pumped at 580 nm. In this and the following figures, au stands for arbitrary units. In this figure, the ordinate values are proportional to the lock-in amplifier read-out in microvolts, while in the following figures, the ordinate has been renormalized to accommodate the fitting procedure.



- NO rebinds very quickly
- Geminate recombination occurs on two timescales
- There's an indication of a 6-coordinate domed structure
- MbNO & MbO_2 have similar binding geometries but very different affinities
- The geminate recombination has an excitation wavelength-dependence

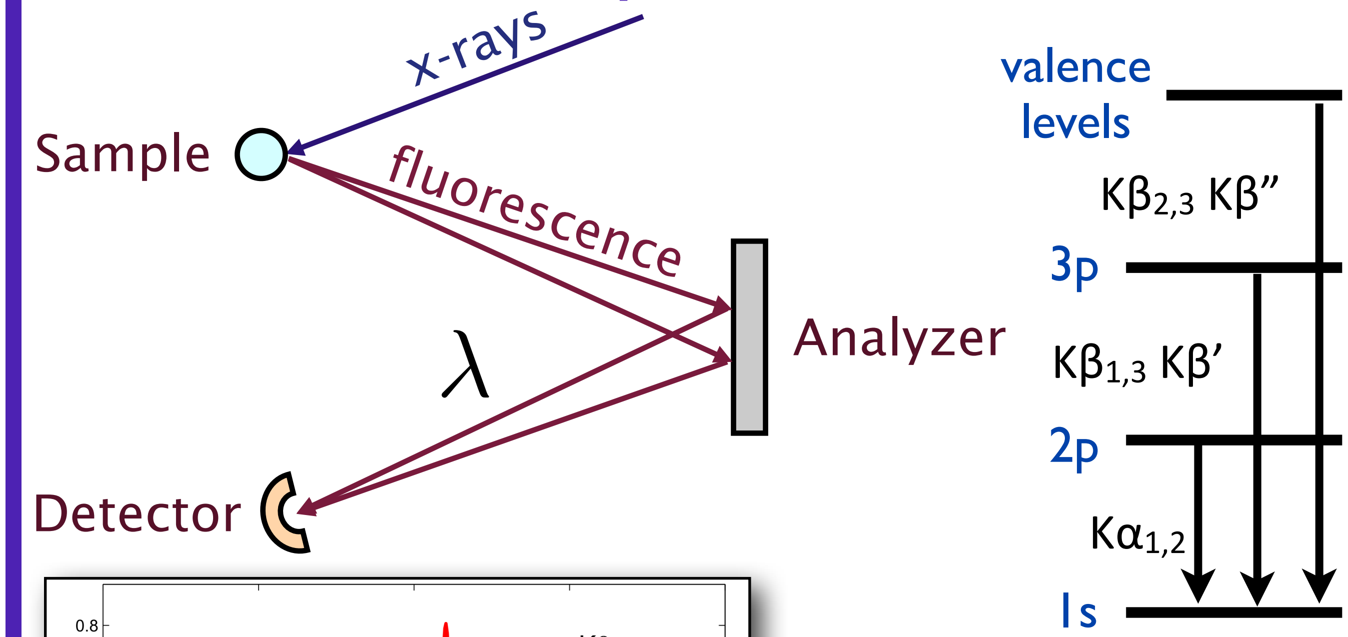
X-ray absorption

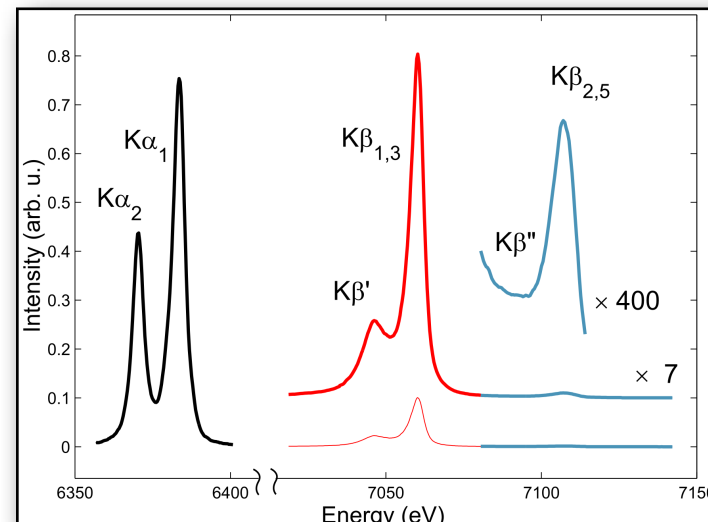


EXAFS: distances to neighbouring atoms

XANES: oxidation state, geometry, coordination environment

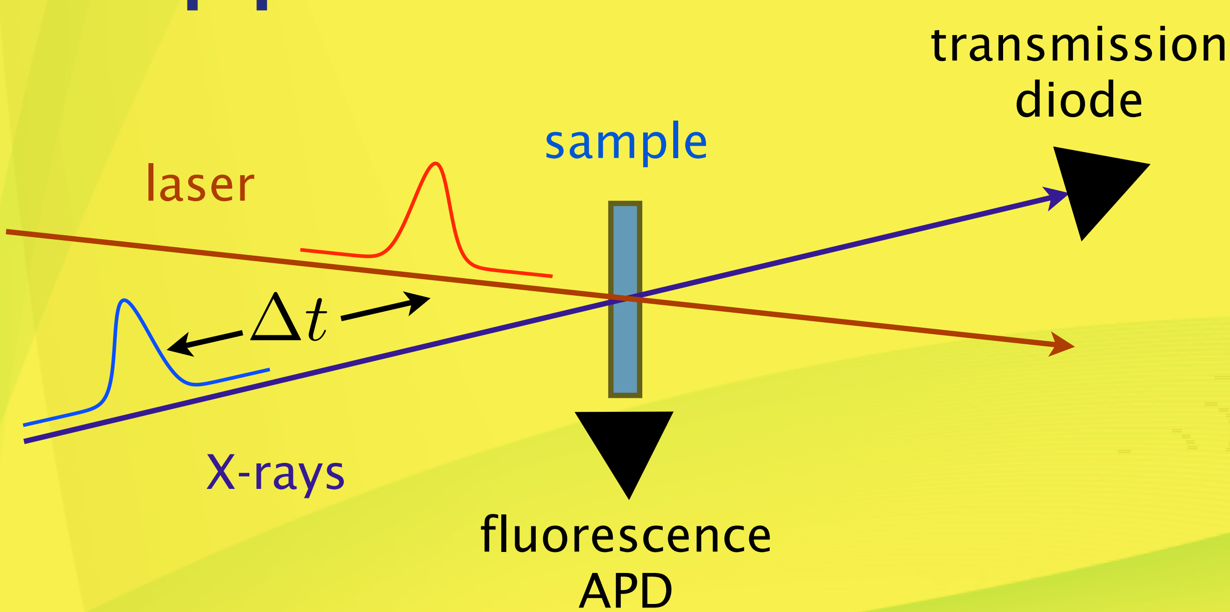
X-ray emission





XES provides complementary electronic information to XAS

Pump-probe measurements



SuperXAS
- SuperBend from 4.5 to 35 keV
- Si(111) monochromator
- X-ray emission spectrometers
- 10^{11}-10^{12} photons/second

PHOENIX beamline
- in-vacuum undulator (0.8-8 keV)
- Si (111), KTP, Be, InSb mono crystals
- micro-focus capability (< 1 μm^2)
- 10^{11}-10^{12} photons/second

microXAS beamline
- in-vacuum undulator (4-20 keV)
- Si (111), Ge(111) & Si(311) mono crystals
- micro-focus capability (< 1 μm^2)
- 10^{12} photons/second

The FEMTO slicing source at microXAS
- 4 to 20 keV
- bandwidth 1%, 0.03%, 0.015%
- 140 ± 30 fs x-ray pulse duration
- timing stability of < 30 fs RMS over days
- 10^5 photons/second @ 1% BW

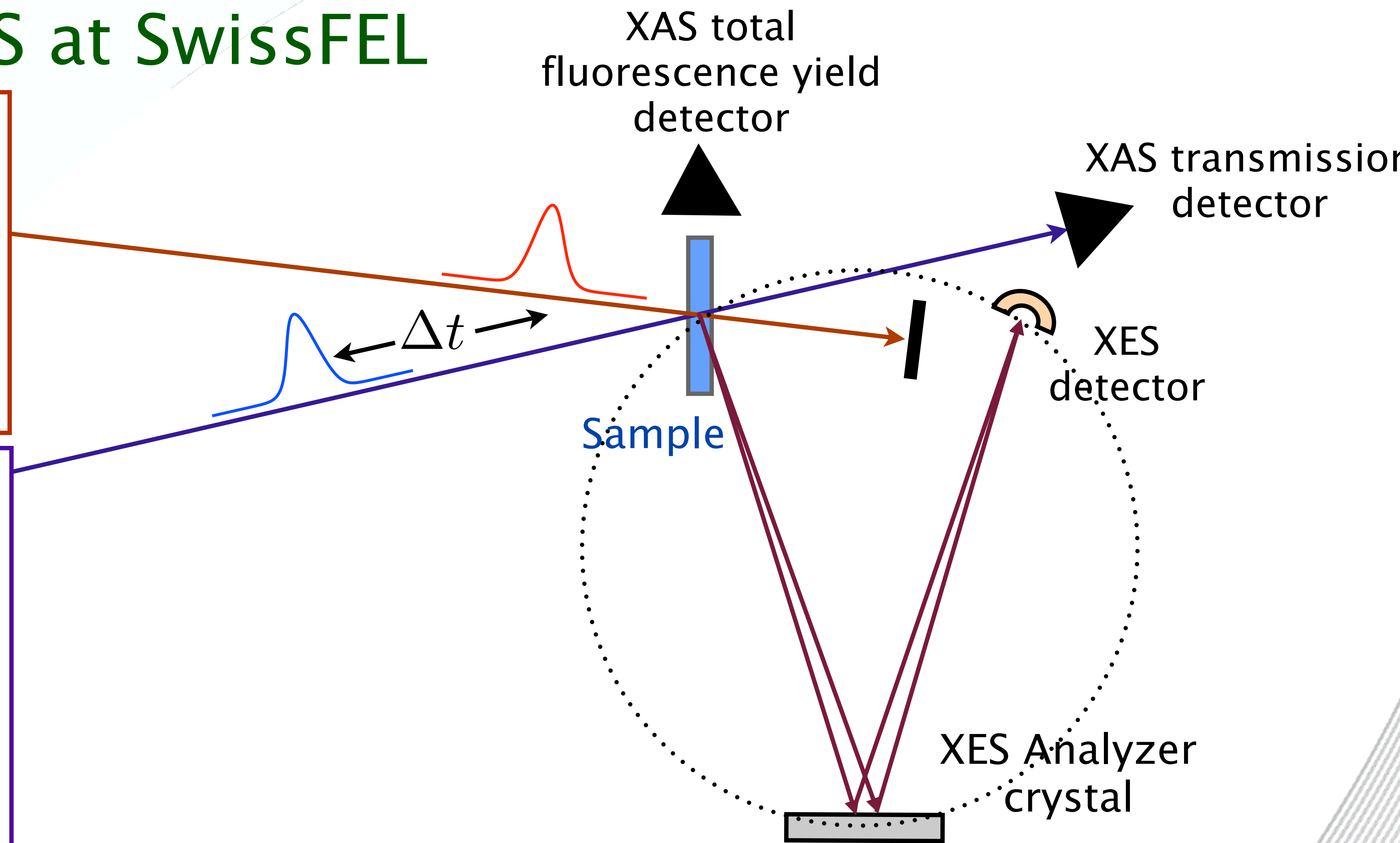
Pump-probe XAS/XES at SwissFEL

Laser pump

- tuneable from IR to UV
- femtosecond pulses
- possibility to stretch (>1 ps)
- rep rate matched to SwissFEL
- controlled delay (0-1 ns)

X-ray probe

- monochromatic (0.015%)
- scannable within undulator bandwidth
- ability to remove mono
- jitter diagnostic
- focussed spot (<100 μm)
- I_{zero} detector



Parameter		Unit	Requirement	Motivation / Remarks
Beam parameters				
Energy		keV	2-12 keV	determines which elements can be investigated
	stability		0.01%	ideally less then bandwidth of undulator
Bandwidth		%	0.01% or better	after monochromator
	stability		<±5%	if the bandwidth of the mono is unstable that's not good
Beam position	stability		<±10% beam FWHM	this is coupled to the pump laser available
Beam size		μm	20 μm to 500 μm	Gaussian profile ideally
Photons per pulse		#ph	the more the better	also requires possiblity to attenuate
	stability		1% peak-to-peak	or an excellent Izero monitor is required
Pulse length		fs	5 fs to 10 ps	ability to stretch x-ray pulse lowers peak power
	stability		<10%	
Pulse arrival time	stability	fs	< pulse duration	otherwise short pulse duration is worse then useless
Beam parameter changes during experiment				
Energy	range / step	eV	0.2 eV	more appropriate would be in BW: 0.005%
	rate	eV / min	60 eV/min	for spectroscopy this is a core requirement
Beam size (microfocus only)	range / step	μm	---	not generally required
	rate	μm / min	---	not generally required
Pulse length	range / step		---	not generally required
	rate		---	not generally required
Beam geometry				
Beam slope	max. tolerable	μrad	---	Thin samples make this not an issue
Working distance	min. required	mm	>300 mm	Some space is required for laser optics

No matter what comes out we're going to be able to use it

Wish List

- ➡ Jitter < x-ray pulse duration
- ➡ Sample chamber for anaerobic conditions
- ➡ Ability to easily scan x-ray energy
- ➡ multiple crystal von Hamos-type single-shot XES spectrometer
- ➡ chemical preparation facilities (fume hood, glove box, fridge/freezer sample storage, analytical balance...)
- ➡ pixel detectors with a **HIGH** energy threshold for fluorescence detection with the ability to take fast differences
- ➡ chemical diagnostics on-site (UV-Vis spectrometer)
- ➡ microscope for alignment
- ➡ single-shot dispersive XAS (ESRF ID24)