Ultrafast laser-plasma sources for 50-fs hard x-ray pulse generation and laser pump x-ray probe measurements of solvated transition metal complexes

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Outline

- Ultrafast XANES of $Fe(CN)_5^{4-}$
- 50-fs laser-plasma x-ray sources
- Ultrafast x-ray sources and Cornell ERL-II experiments

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Multiple scattering paths of IPC-Bz at 0 Kelvin

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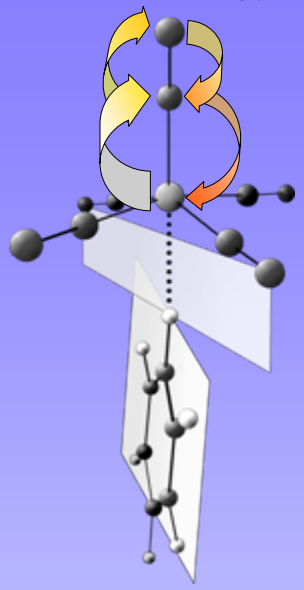

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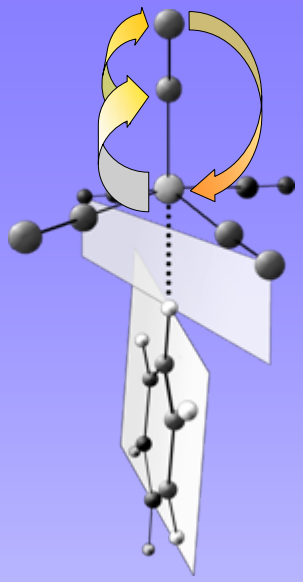

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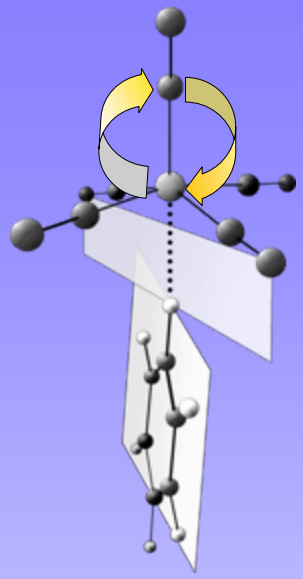

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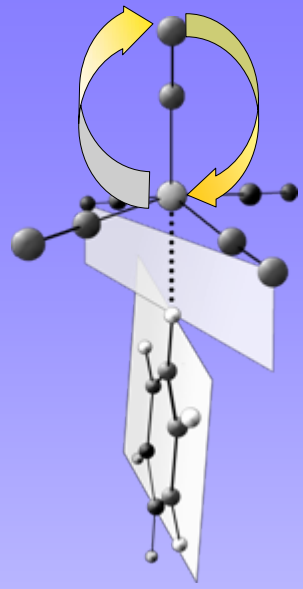

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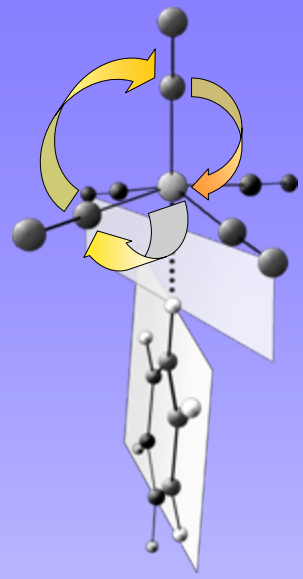

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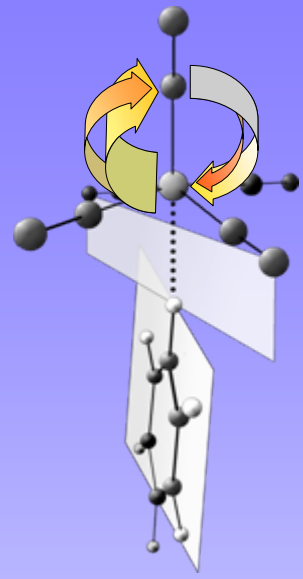

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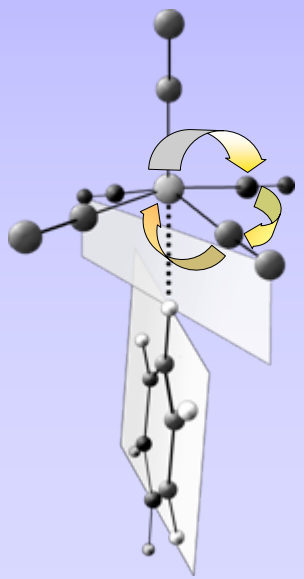

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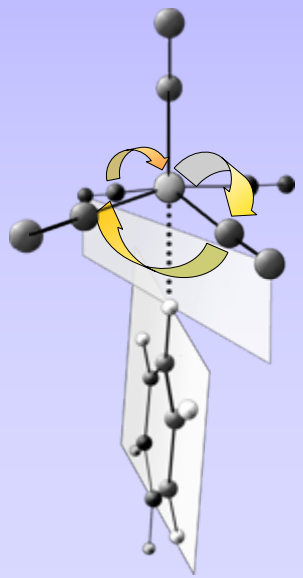

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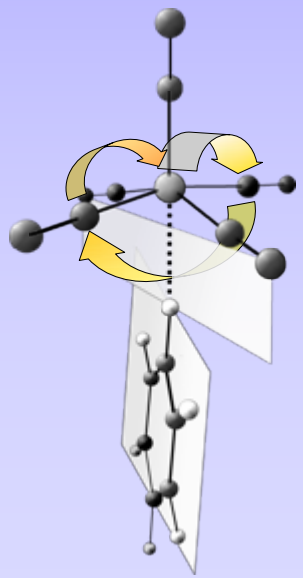

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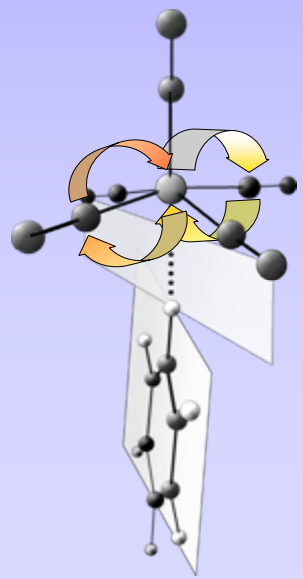

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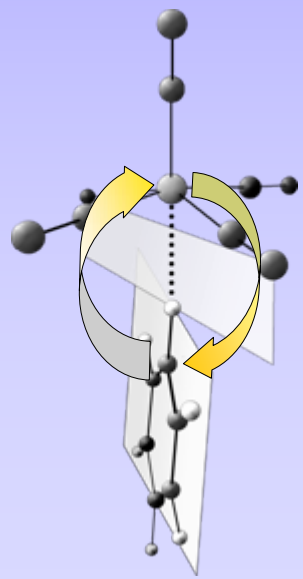

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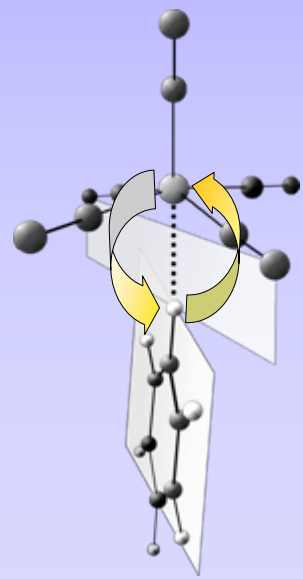
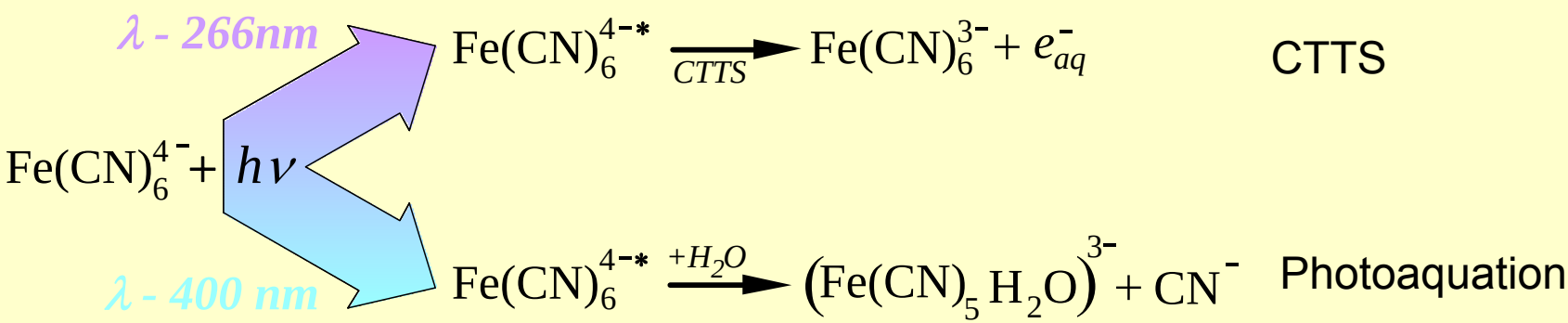
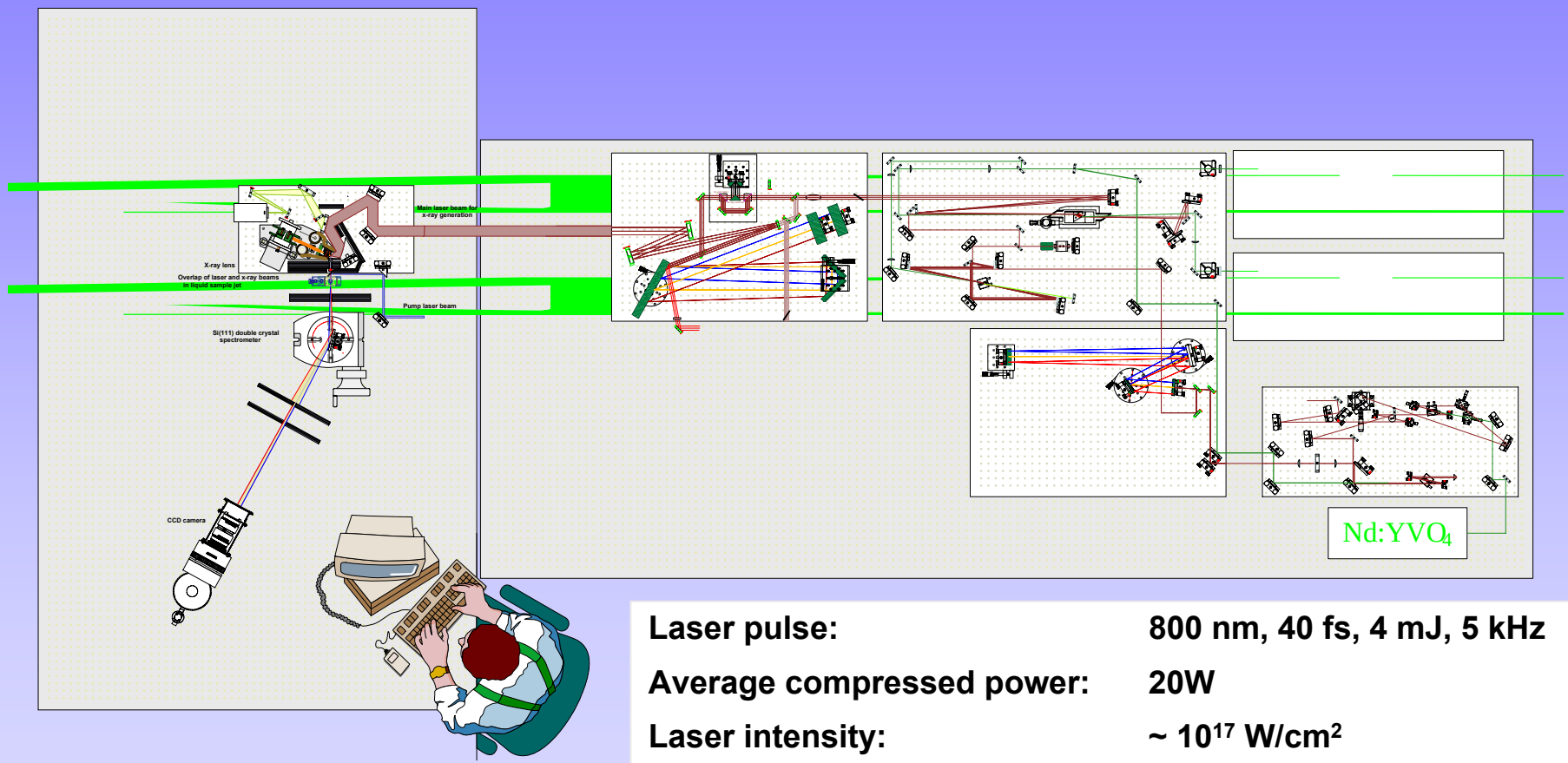


Excited state chemistry of iron hexacyanide in water



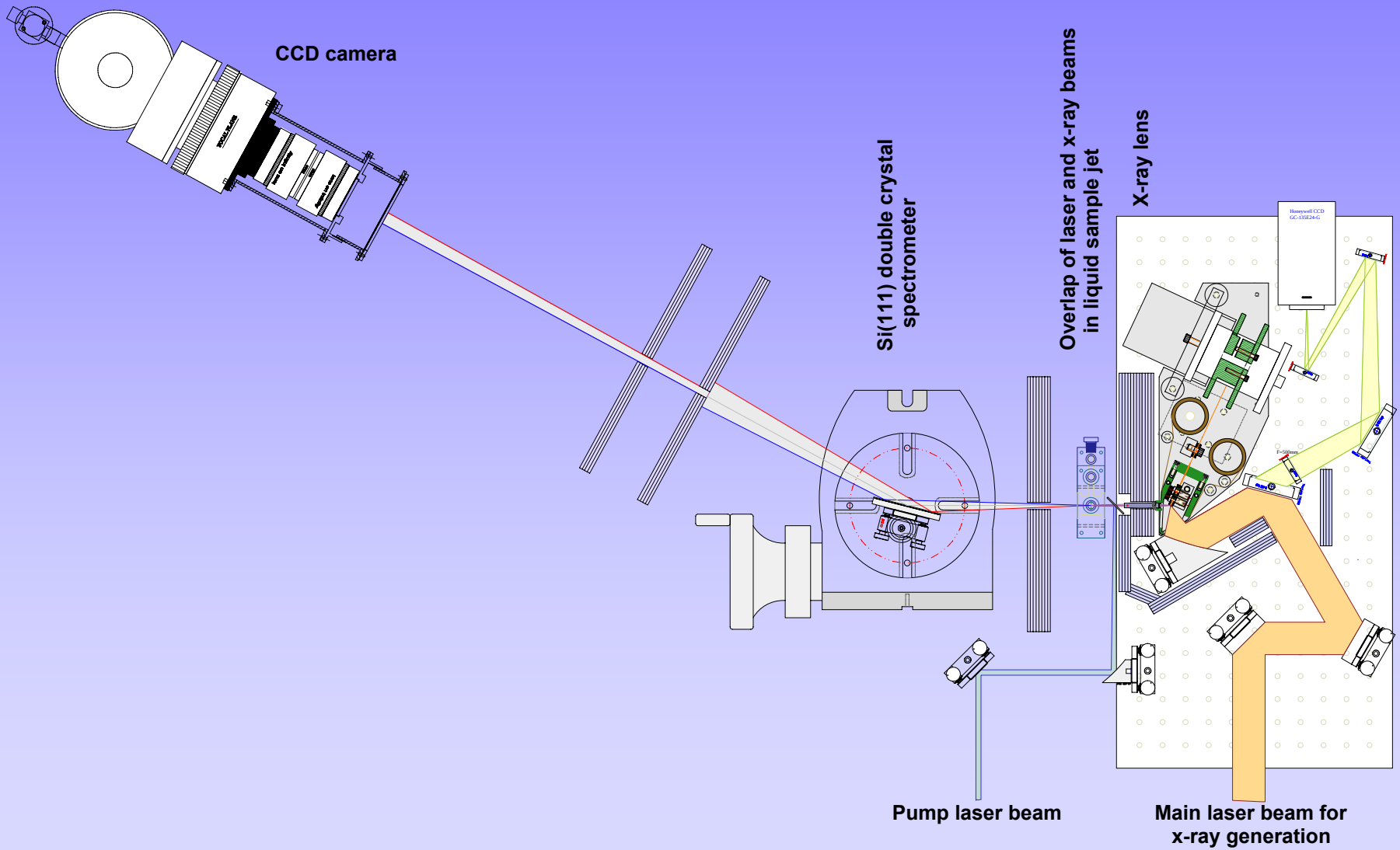
- Excitation photon wavelength < 270 nm: Charge Transfer To Solvent
- Excitation photon wavelength < 400 nm: Photoaquation

Ultrafast laser for laboratory-based plasma x-ray sources

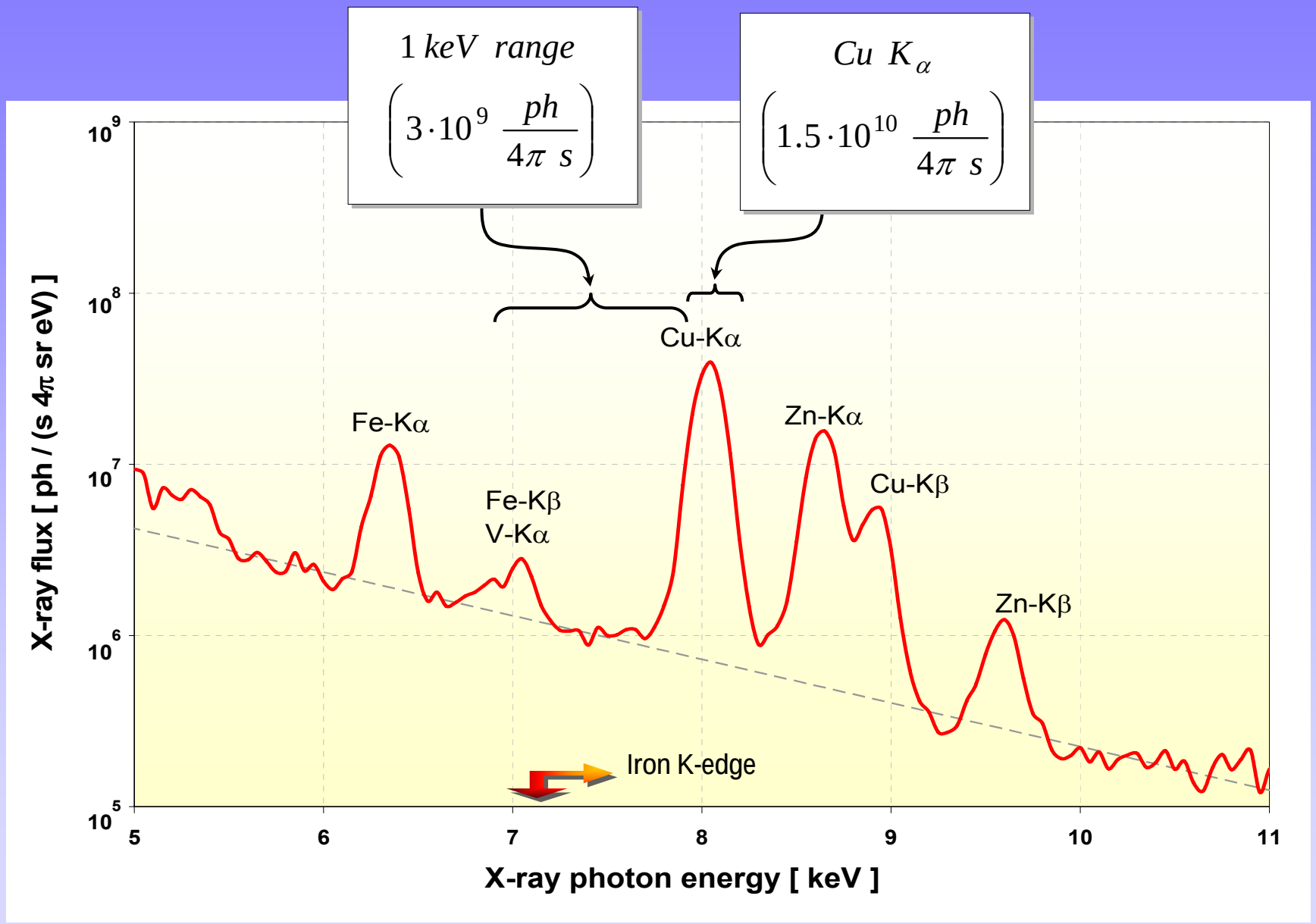


Laser pulse:	**800 nm, 40 fs, 4 mJ, 5 kHz**
Average compressed power:	**20W**
Laser intensity:	**~ 10^{17} W/cm^2**

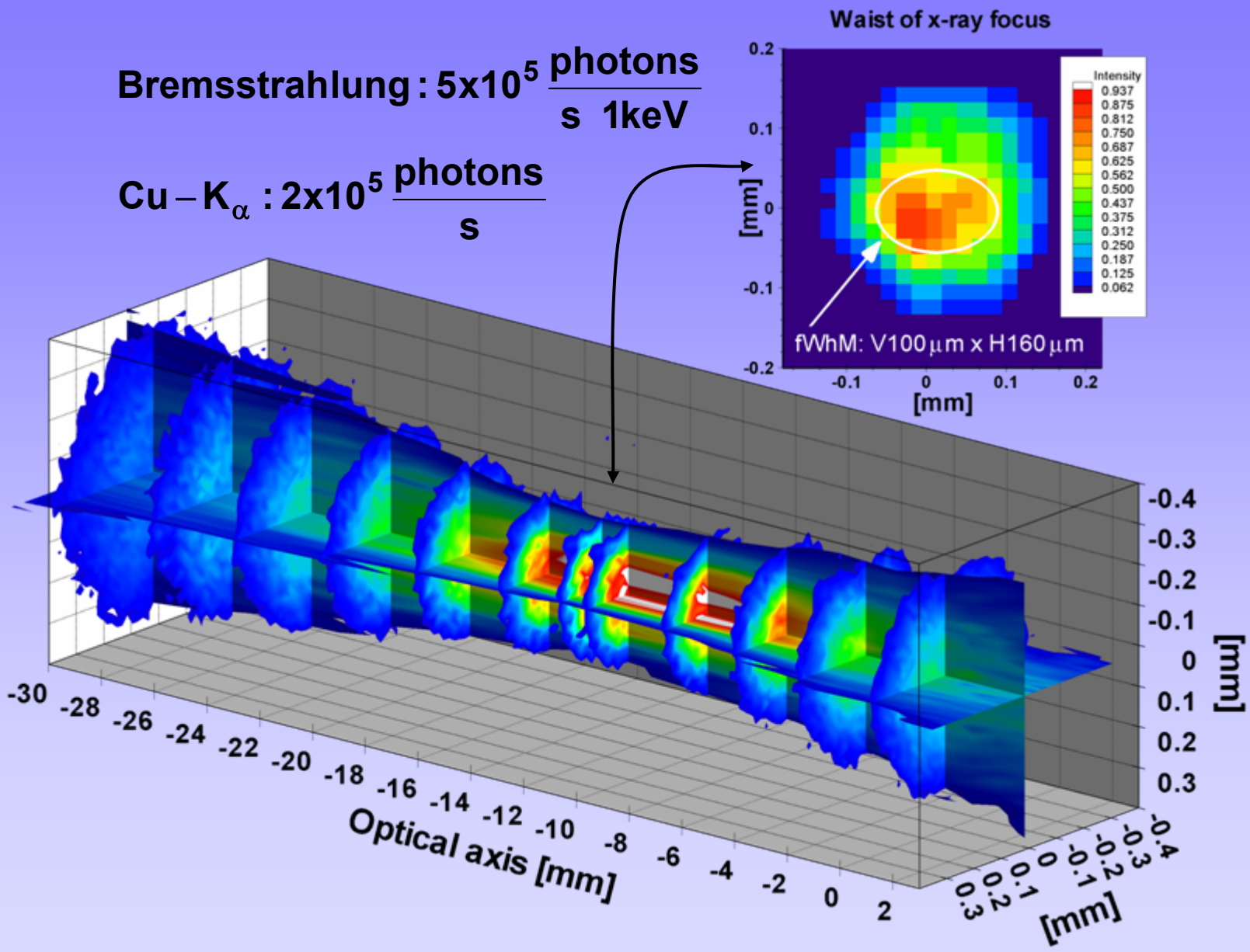
Ultrafast laser-driven XAFS spectrometer



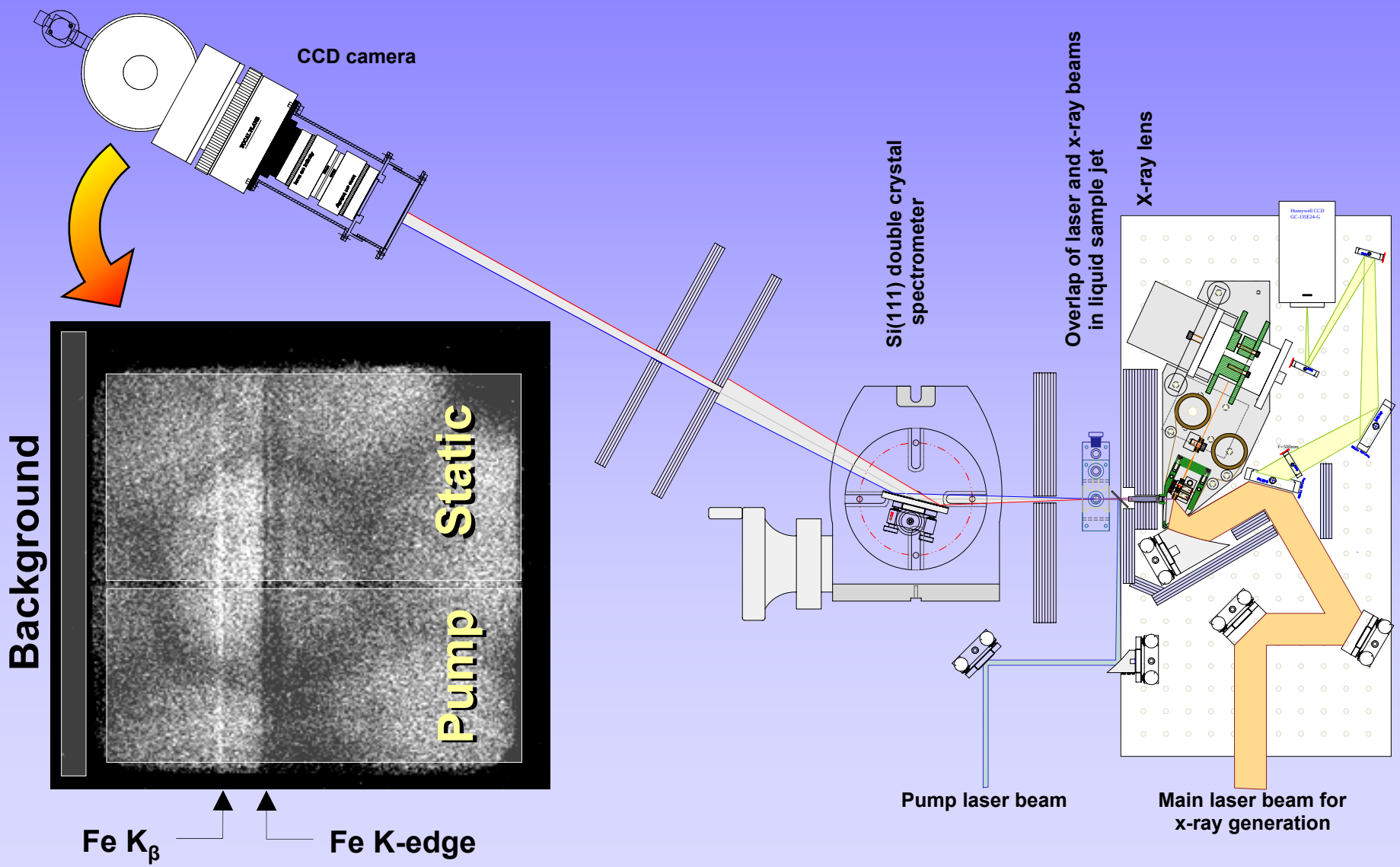
X-ray emission spectrum of 2-kHz source



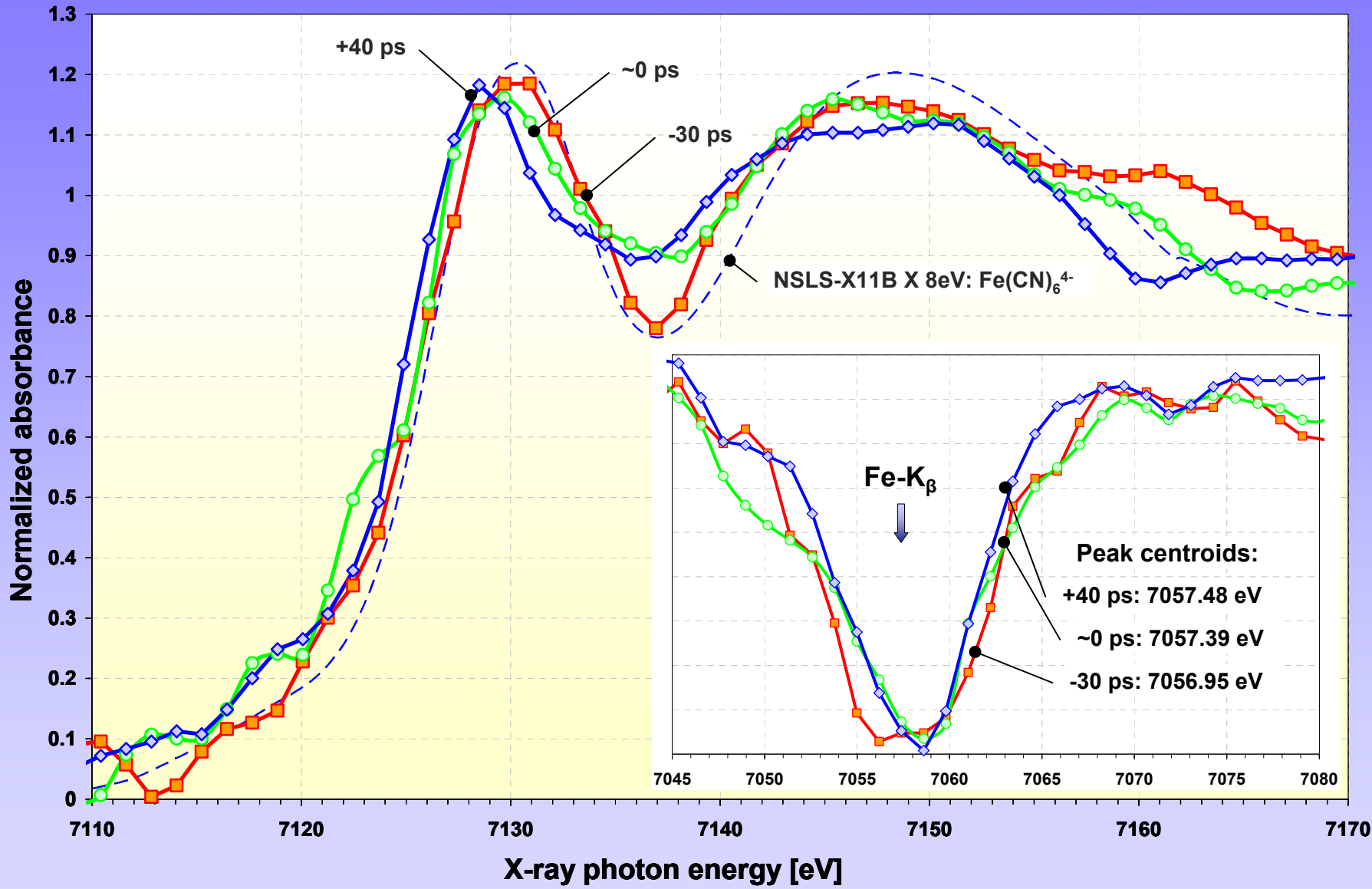
Laser plasma x-radiation imaged into sample by x-ray lens



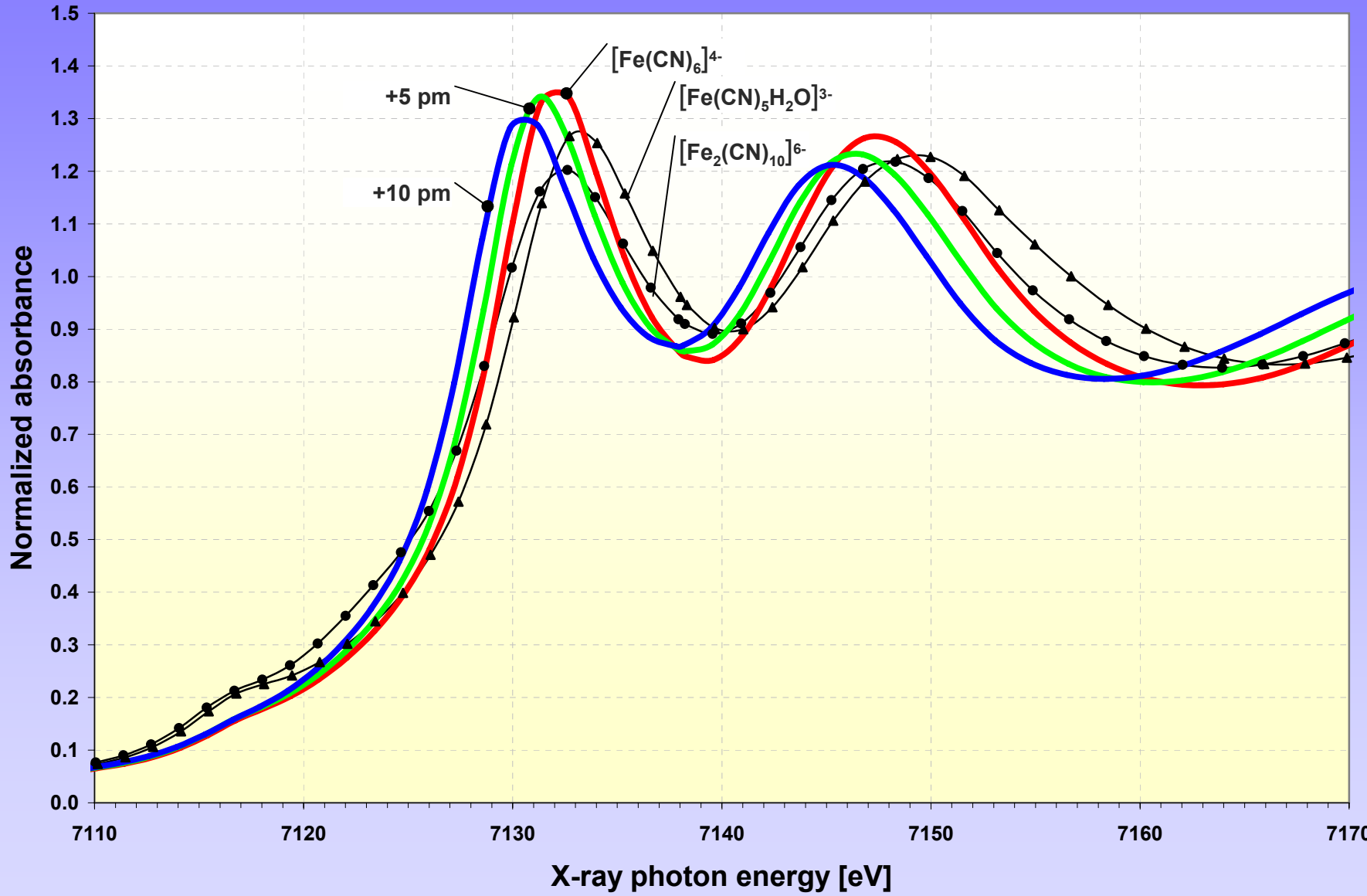
Ultrafast laser-driven XAFS spectrometer



UXANES at –30 ps, 0 ps, and +40 ps after photoexcitation



Radial Fe-ligand bond lengths increase of $Fe(CN)_6^{4-}$



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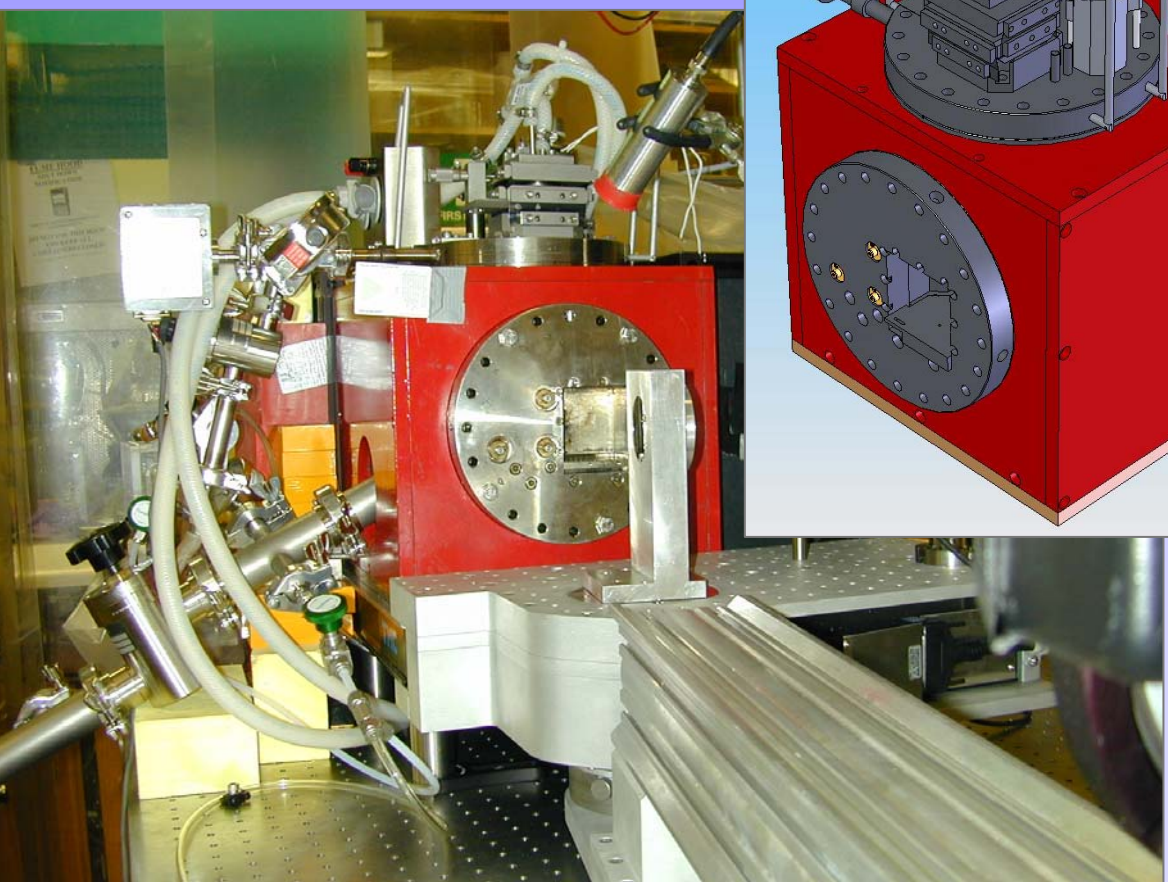
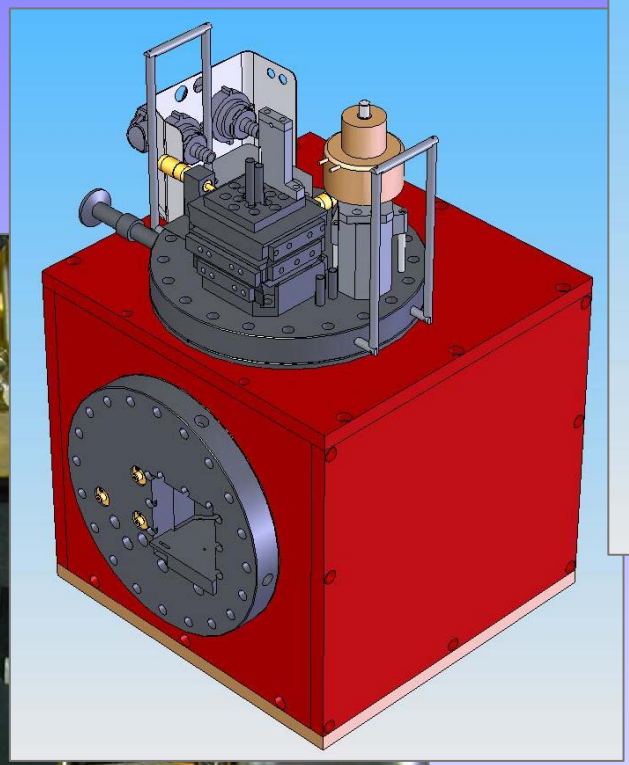
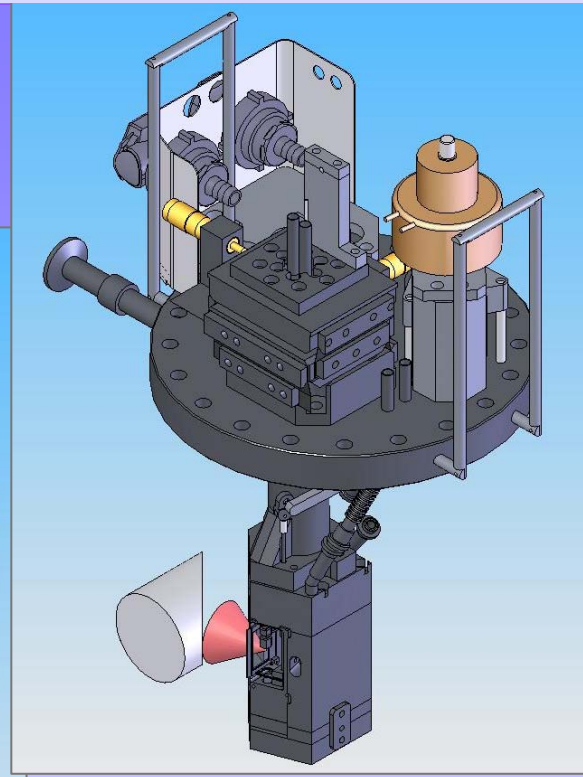
- Ultrafast x-ray sources and Cornell ERL-II experiments

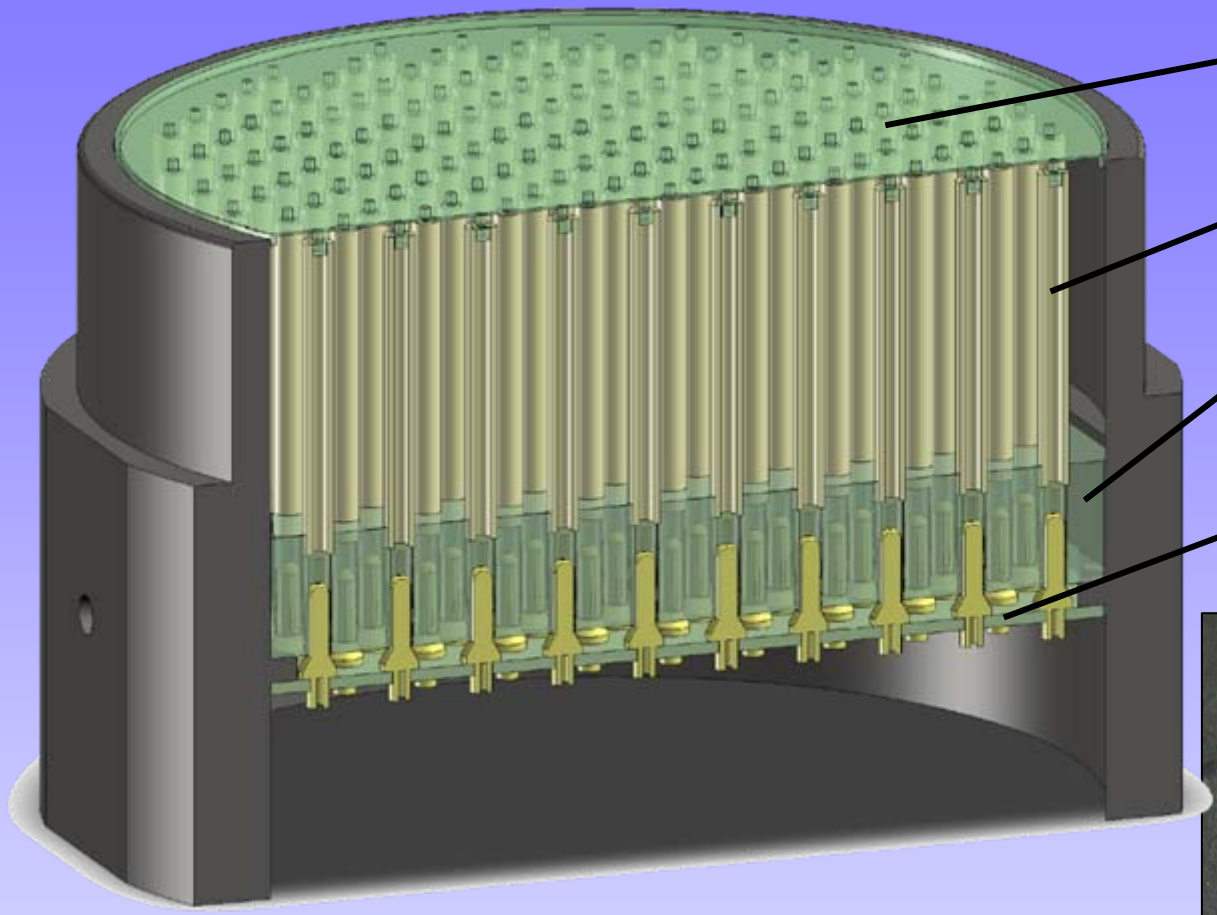
New liquid metal x-ray source

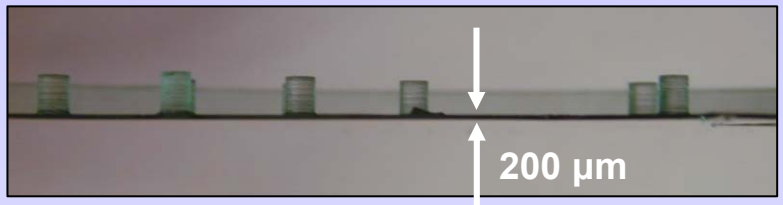




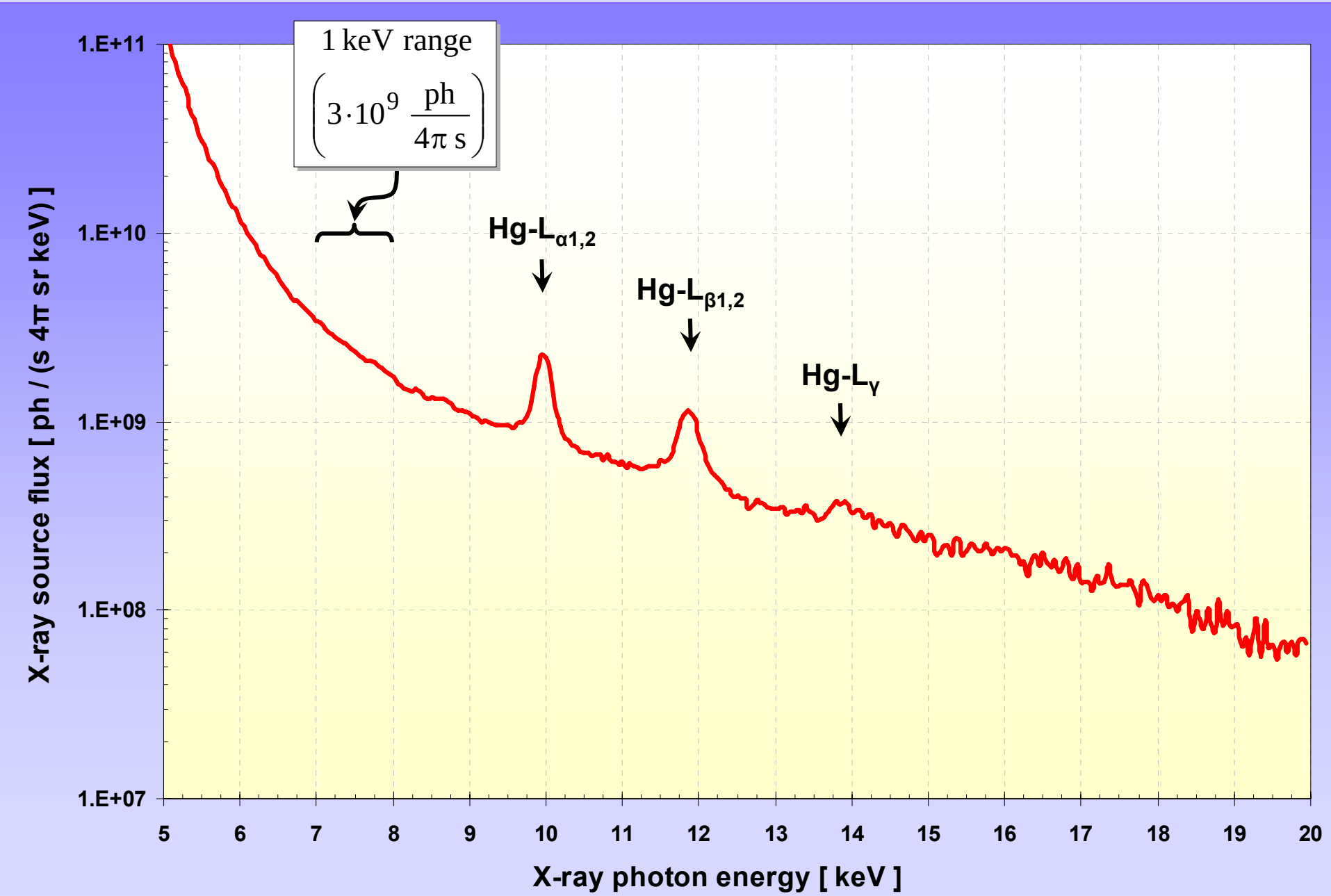


4” diameter deformable mirror for focus optimization

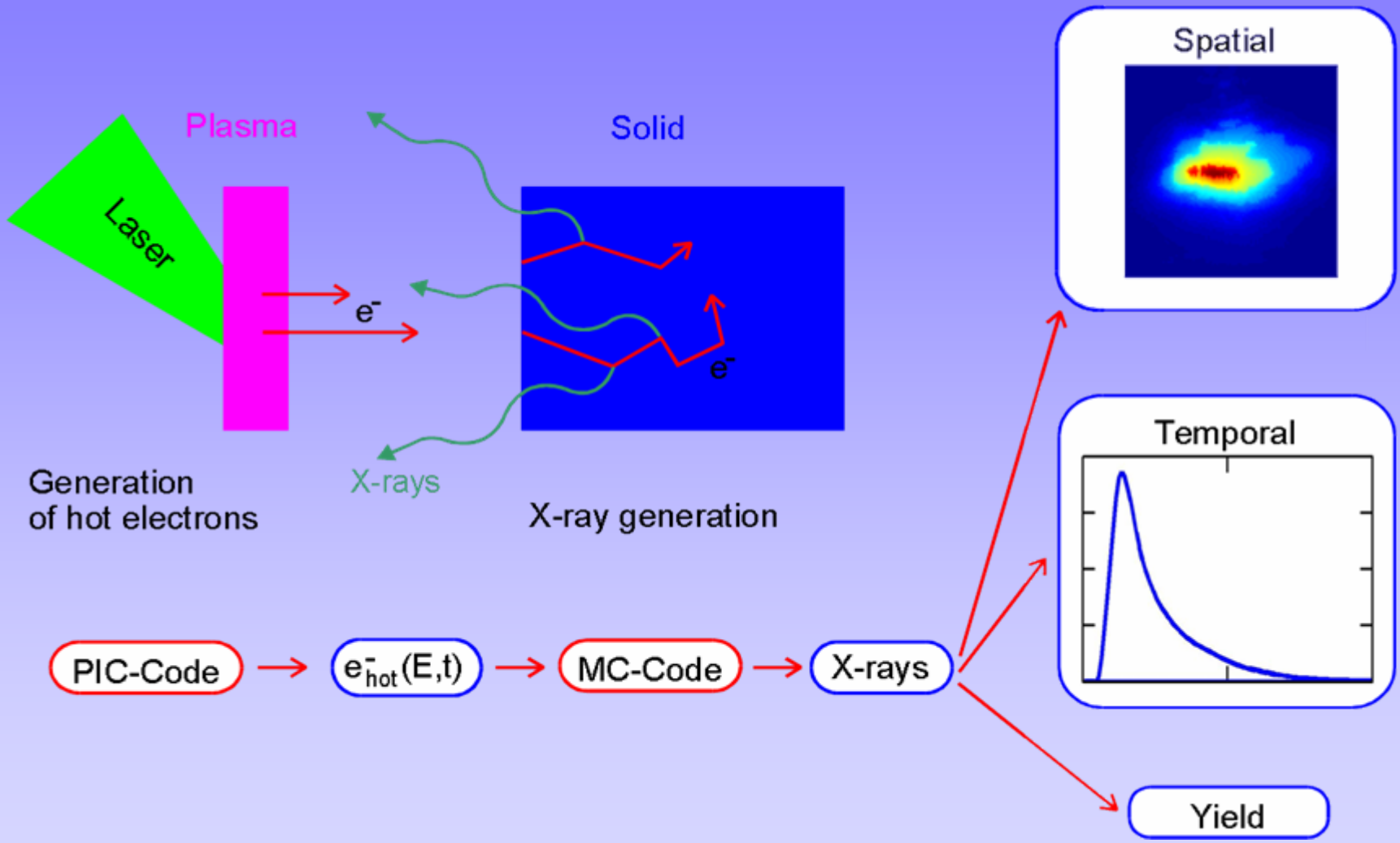




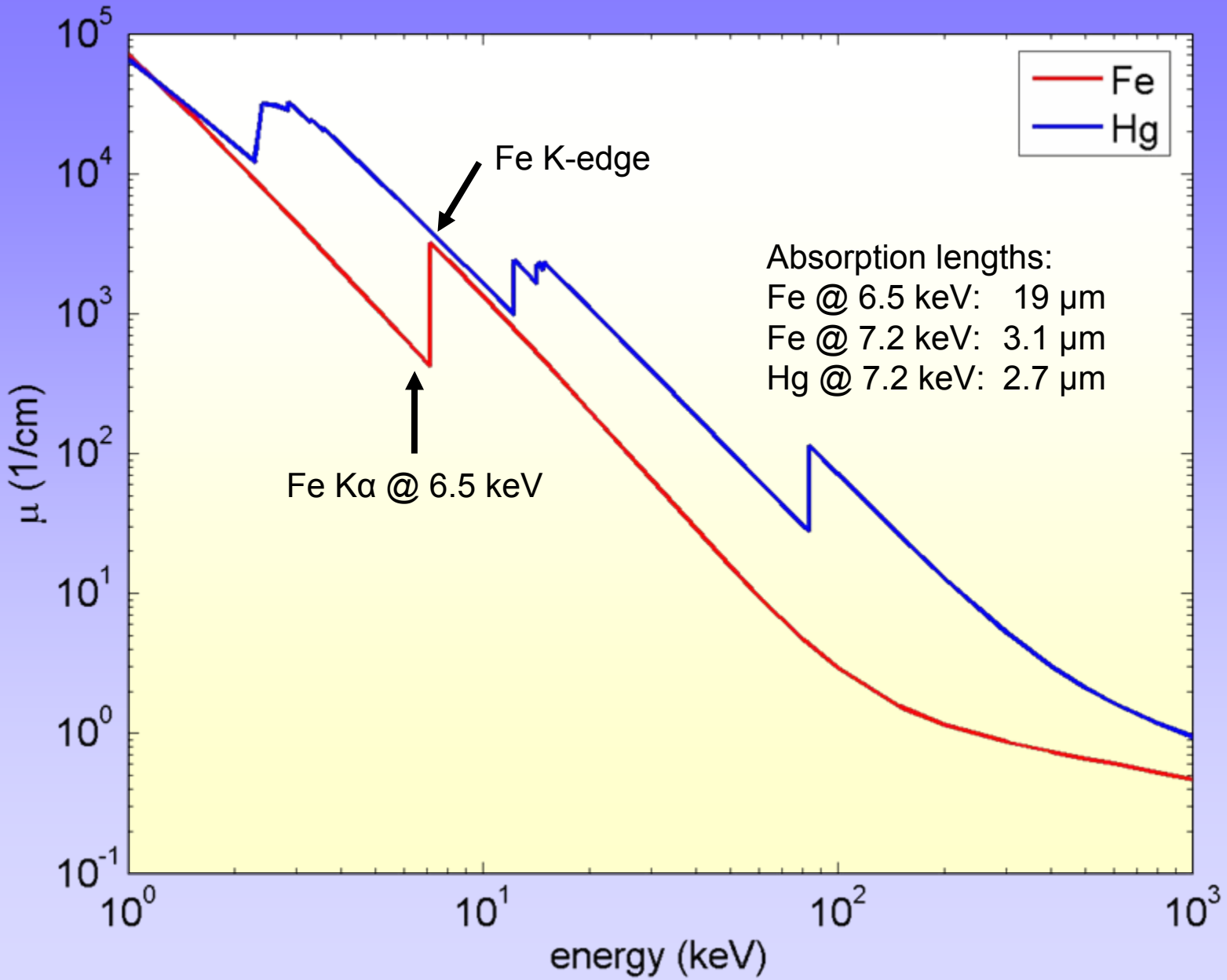
X-ray emission spectrum of Hg-source



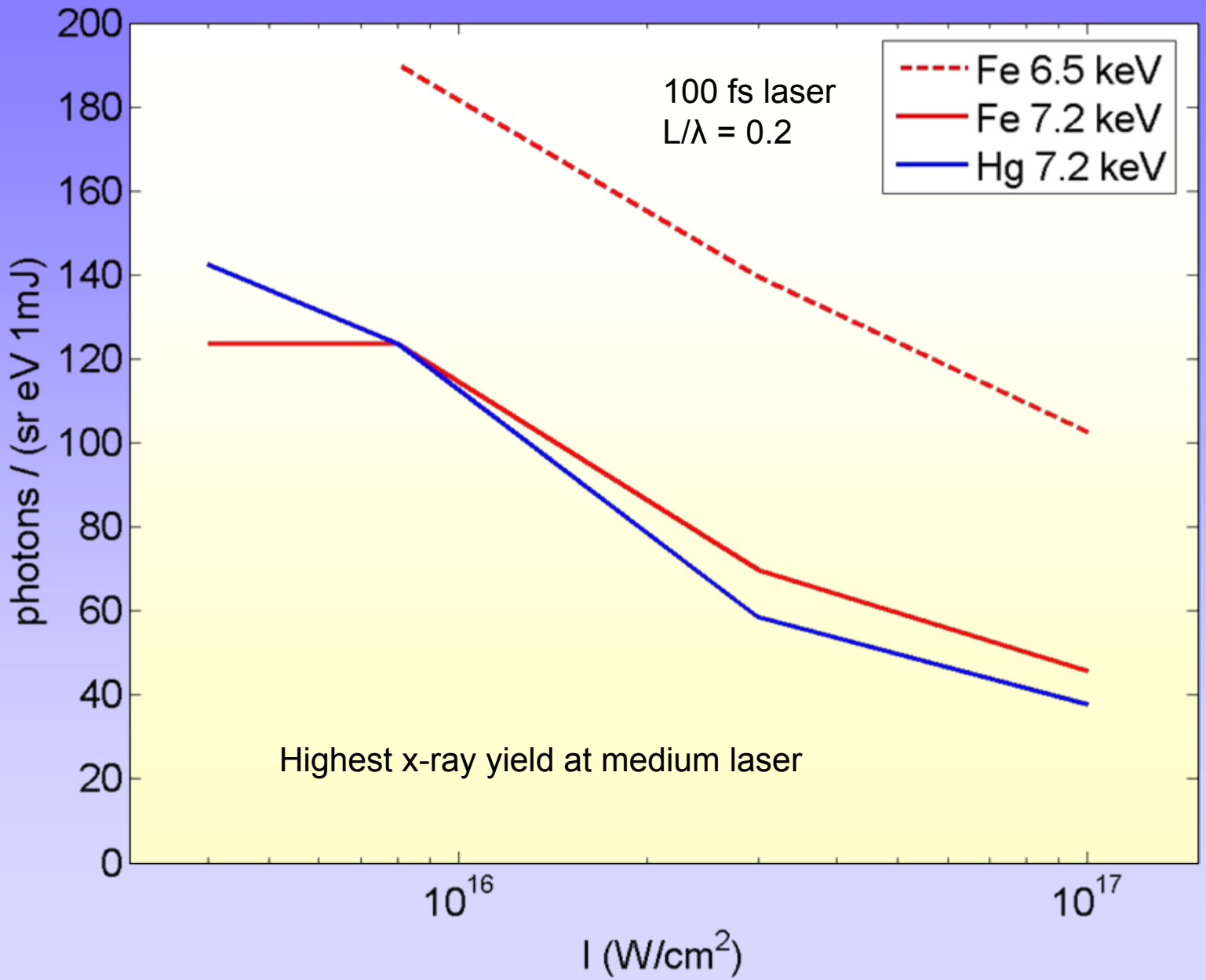
Numerical modeling of laser-produced x-ray emission



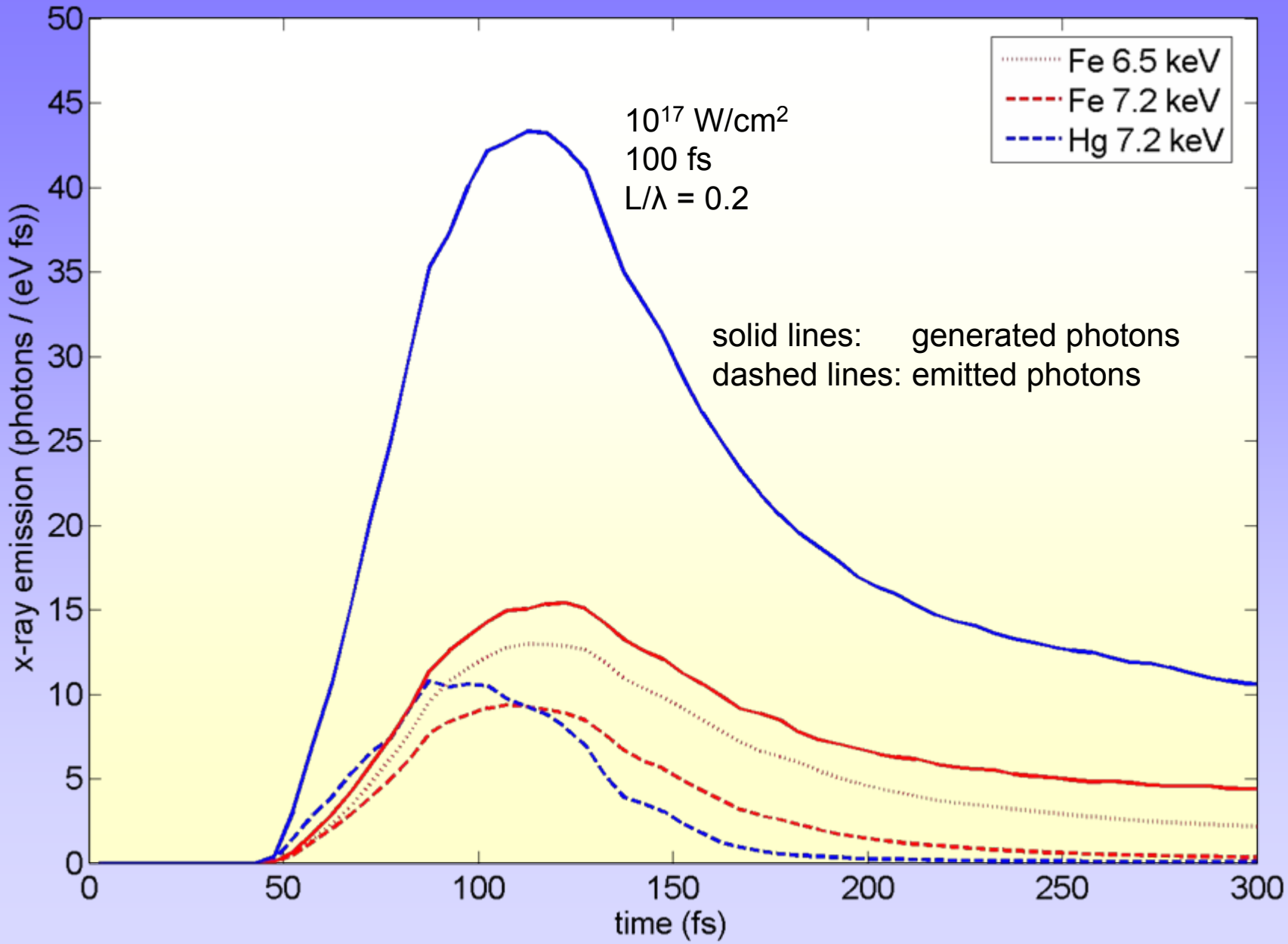
X-ray absorption in Iron and Mercury



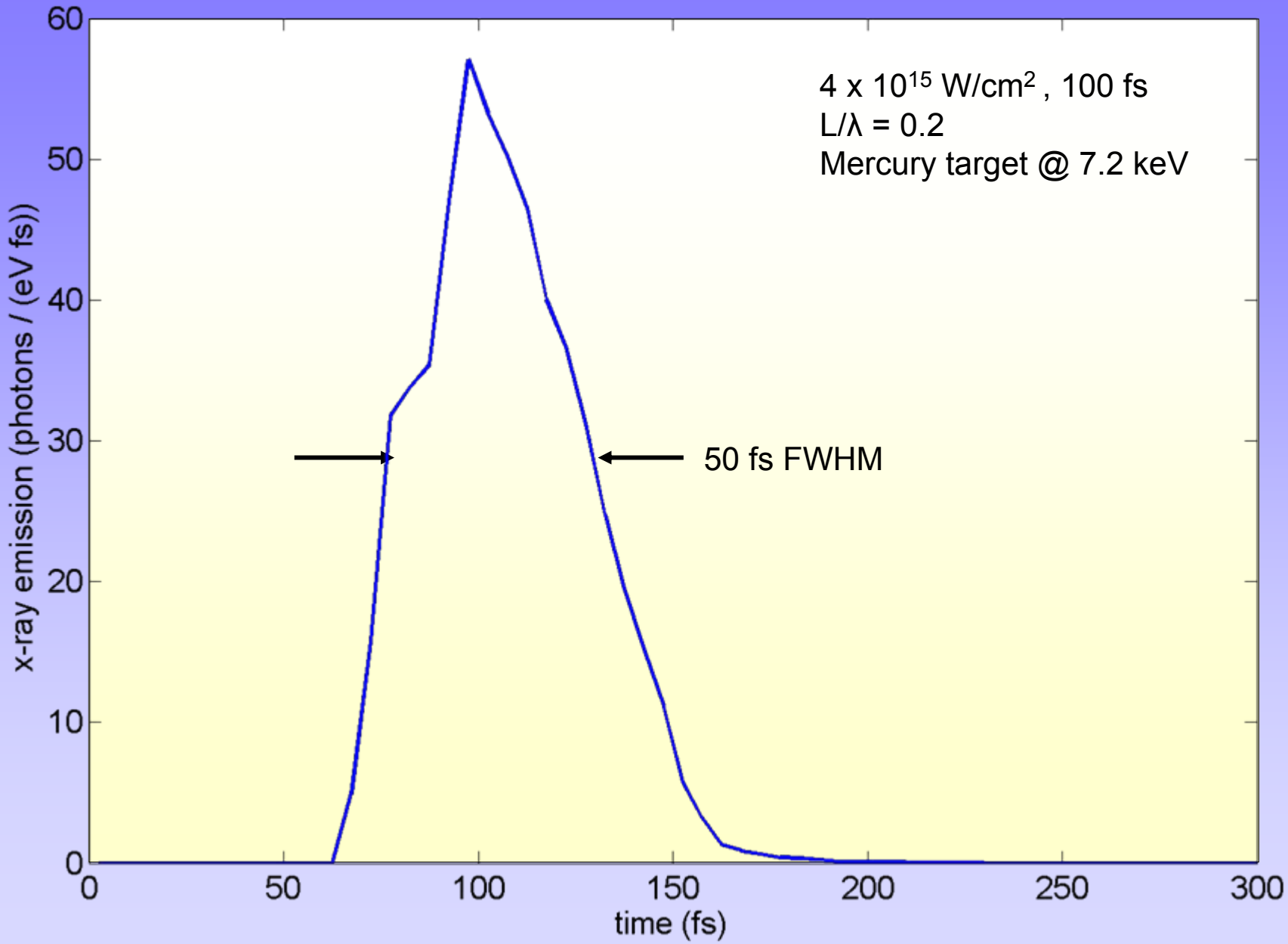
Similar x-ray yields for Fe and Hg @ 7.2 keV



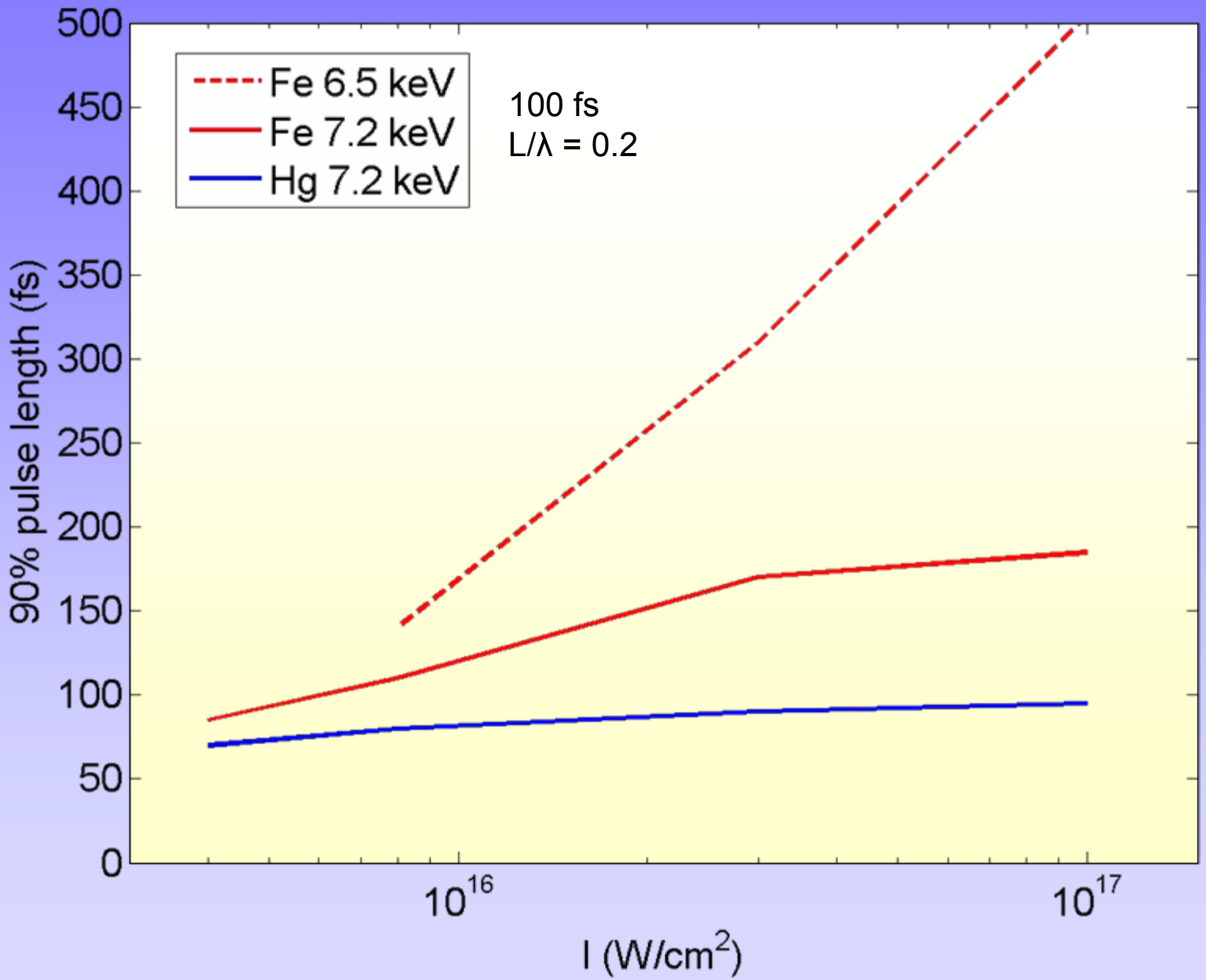
Temporal x-ray pulse shape from Iron and Mercury



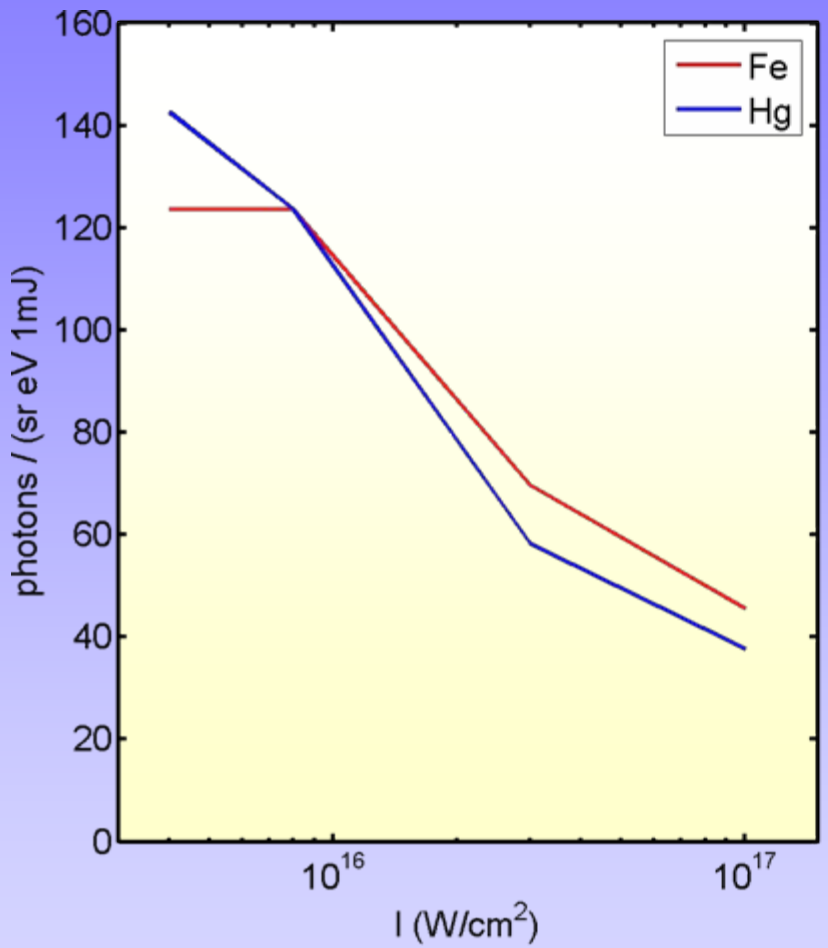
50 fs x-ray pulses are possible

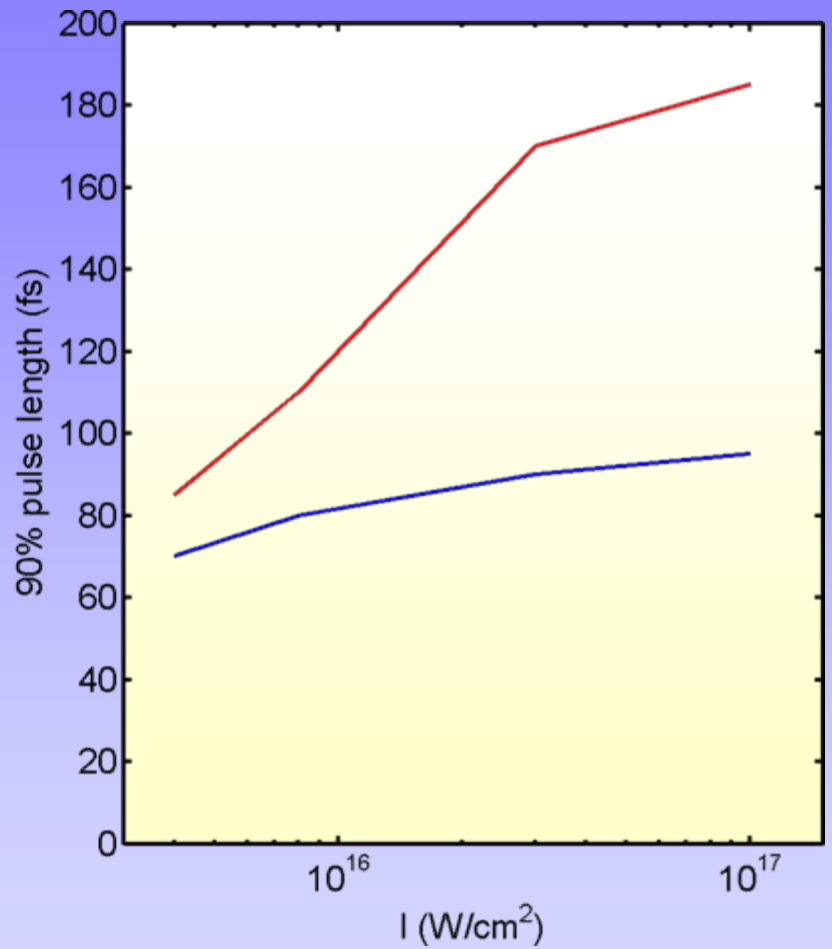


The continuum pulse can be shorter than the laser pulse



X-ray yield and pulsewidth for Fe and Hg target

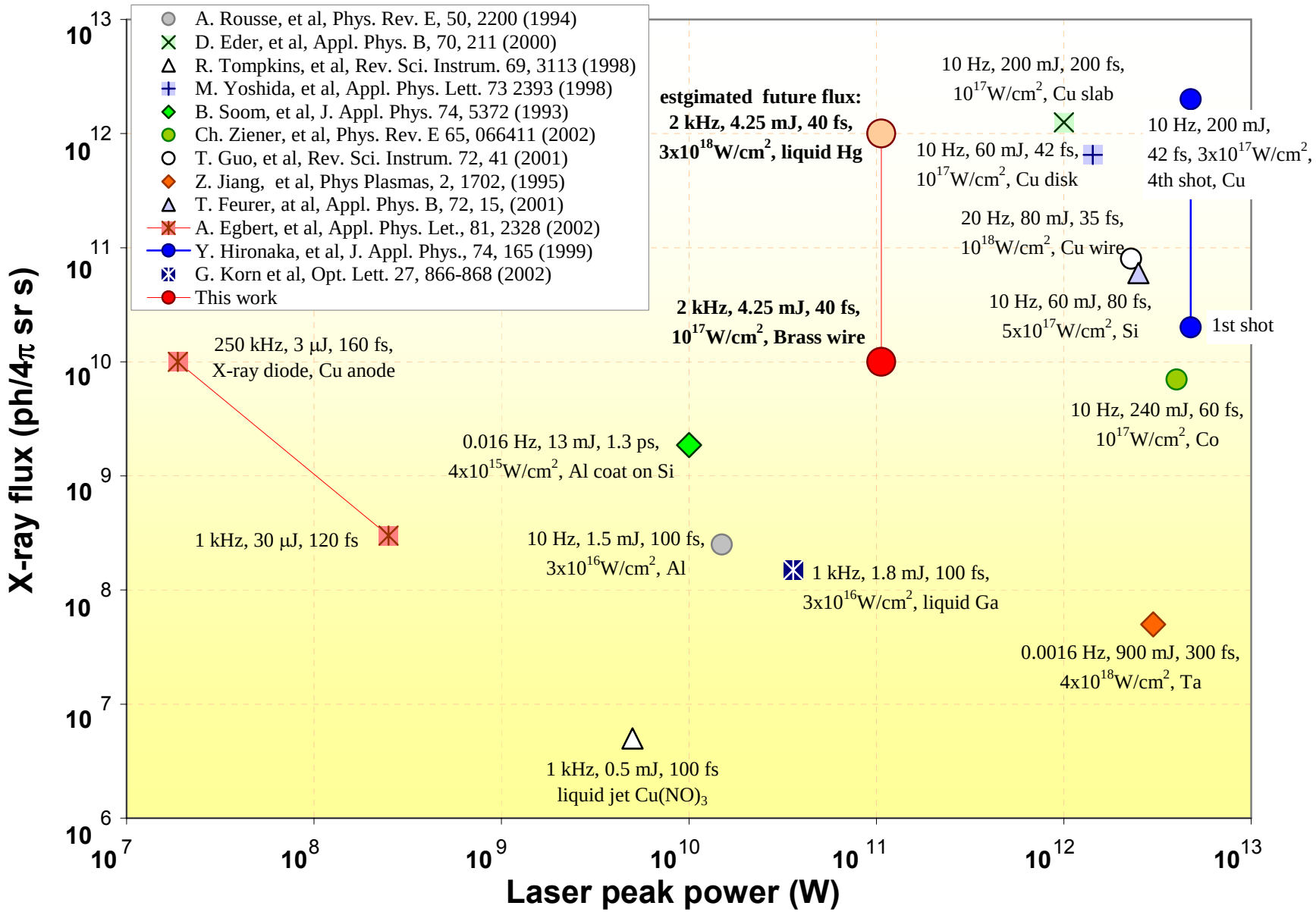




Outline

- Ultrafast XANES of $Fe(CN)_5^{4-}$
- 50-fs laser-plasma x-ray sources
- **Ultrafast x-ray sources and Cornell ERL-II experiments**

Comparison of laboratory-based hard x-ray sources



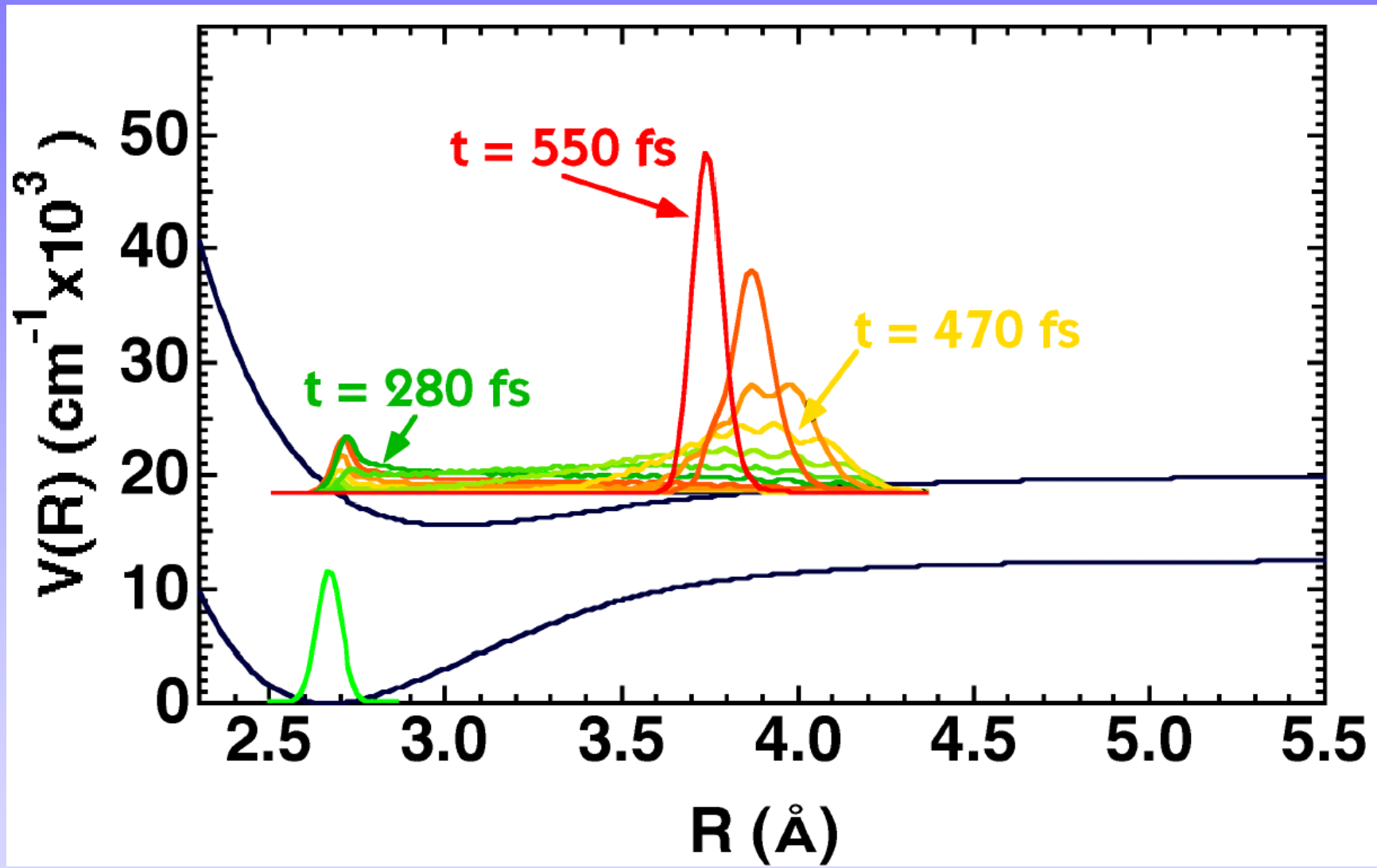
Comparison of x-ray sources

X-ray absorption spectrometer with …	Repetition rate [kHz]	Pulse length [ps]		Average x-ray flux [ph / s]	Peak x-ray flux [ph / s ps]
12 kW rotating anode tube	c.w.			6×10^{9}	6×10^{-3}
APS, sector 7ID, @ 1kHz pump laser rep. rate, special-timing-mode	1	80	At sample	unfocused: 2×10^{9} KB-focused: 5×10^{7}	unfocused: 2×10^{7} KB-focused: 5×10^{5}
APS, sector 11ID-D, @ 1kHz pump laser rep. rate, special-timing-mode	1	80	At sample	10^{8}	10^{6}
Tabletop ultrafast x-ray sources (8 - 15 W)	4	0.1	At sample	Old Source: 10^{6} New Source: 10^{8} + crystal optics: 10^{10}	Old Source: 10^{7} New Source: 10^{9} + crystal optics: 10^{11}
			At detector	Old Source: 100 New Source: 10^{4} + crystal optics: 10^{6}	Old Source: 1000 New Source: 10^{5} + crystal optics: 10^{7}

Prospective ERL parameters

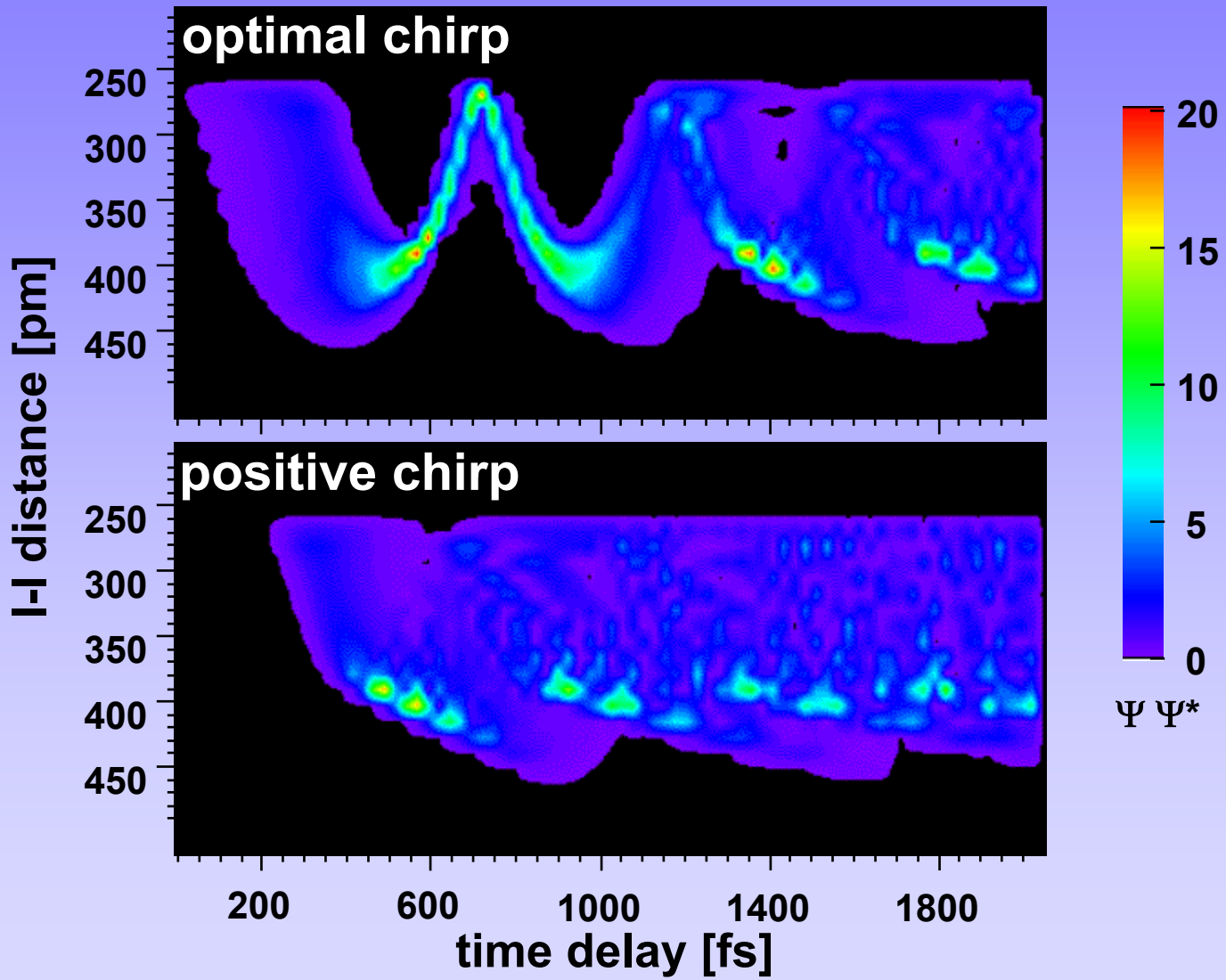
	Cornell ERL-II			Cornell ERL-IIa		
Mode of operation	Hi-flux	Hi-coh	Ultrafast	Hi-flux	Hi-coh	Ultrafast
Machine energy E (GeV)	5.3	5.3	5.3	5.3	5.3	5.3
Charge per bunch q (nC)	0.077	0.008	1	0.154	0.0192	1
Repetition rate f (MHz)	1300	1300	1	1300	1300	10
Machine current I (mA)	100	10	1	200	25	10
Horizontal emittance ε_x (nm-rad)	0.1	0.015	0.1	0.1	0.008	0.1
Vertical emittance ε_y (nm-rad)	0.1	0.015	0.1	0.1	0.008	0.1
Rms bunch length σ_t (ps)	3.0	3.0	0.1	2.0	2.0	0.02
Energy spread σ_E/E	0.0004	0.0004	0.0027	0.0002	0.0002	0.0027
Undulator length L (m)	25	25	4.1	25	25	4.1
Number of periods N_u	1470	1470	240	1470	1470	240
Deflection parameter K	1.34	1.34	1.34	1.34	1.34	1.34
Fundamental energy E_1 (keV)	8.27	8.27	8.27	8.27	8.27	8.27
Average flux F_n (p/s/0.1%)	5.11E+15	5.31E+14	7.65E+12	1.76E+16	2.20E+15	7.65E+13
Average brilliance B_n (std units)	9.08E+21	1.86E+22	1.10E+19	3.13E+22	1.40E+23	1.10E+20
Peak flux F_p (p/s/0.1%)	4.91E+17	5.10E+16	2.86E+19	2.54E+18	3.16E+17	1.43E+20
Peak brilliance B_p (std units)	8.72E+23	1.78E+24	4.10E+25	4.51E+24	2.02E+25	2.05E+26
Photons per pulse n_p (p/0.1%)	3.93E+06	4.09E+05	7.65E+06	1.36E+07	1.69E+06	7.65E+06
Coherent flux F_c (p/s/0.1%)	5.10E+13	1.04E+14	6.15E+10	1.76E+14	8.20E+14	6.15E+11
Peak coherent flux F_{cp} (p/s/0.1%)	4.90E+15	1.00E+16	2.30E+17	2.53E+16	1.13E+17	1.15E+18
Coherent flux fraction p_c (%)	1.0	19.6	0.8	1.0	37.3	0.8
Photons per coherent volume δ_D	2	5	115	13	57	576
Total undulator output power P_0 (W)	31,679	3,291	51.7	63,358	7,899	517
On-axis power density @20m dP/dA (W/mm^2)	2,655	276	4.3	5,311	662	43
Peak coh. electric field @ exit E_c (V/m)	1.42E+06	4.56E+05	5.71E+04	2.00E+06	7.07E+05	1.81E+05

The I_2-reflectron

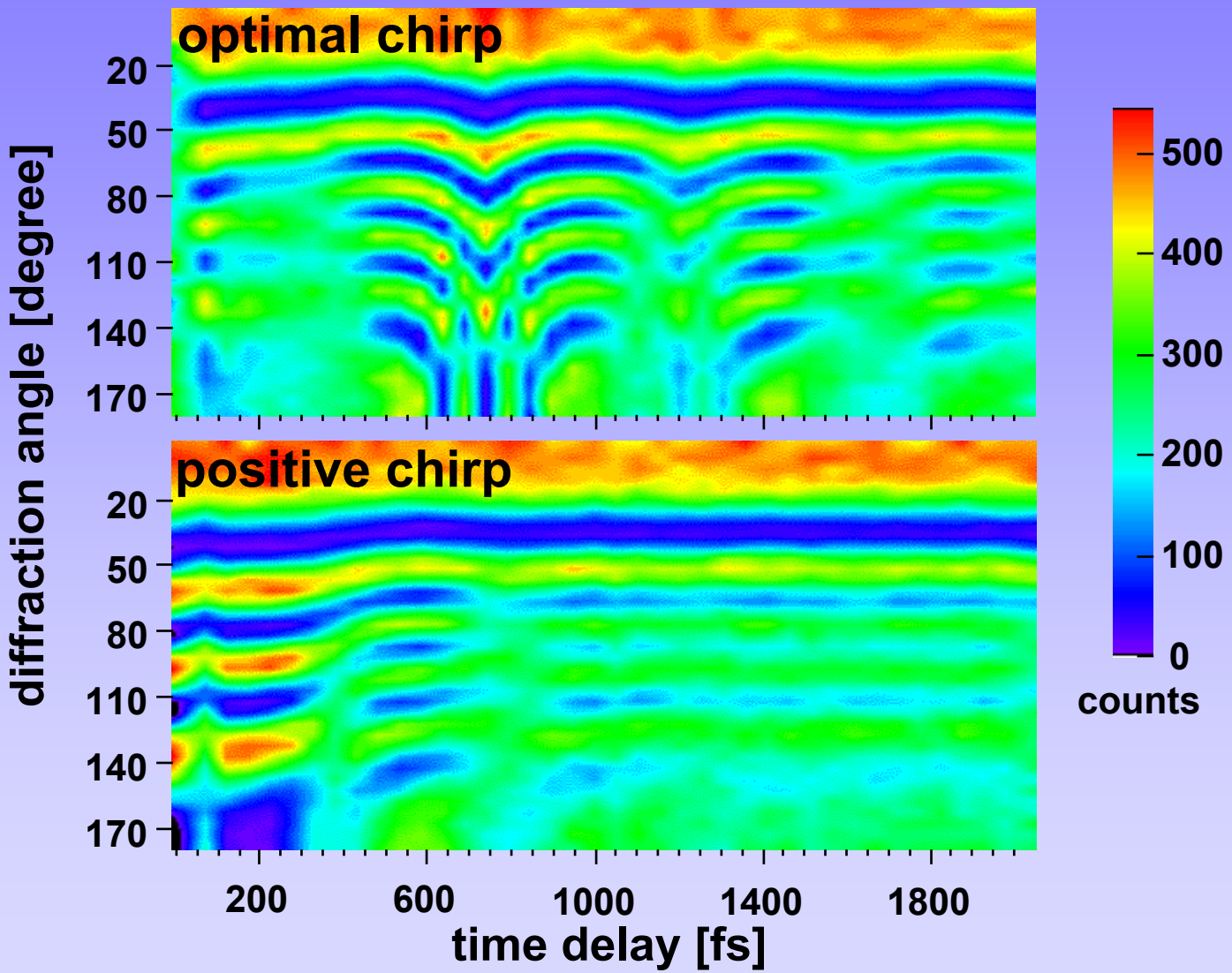


B. Kohler, J.L. Krause, F. Ráksi, C. Rose-Petruck, R.M. Whitnell, K.R. Wilson, V.V. Yakovlev, Y. Yan, S. Mukamel, *J. Phys. Chem.* ***97***, 12602 (1993)

I-I distance probability distributions



Simulated diffraction pattern for Cu-K_α x-ray pulses



Funding

National Science Foundation, Chemistry

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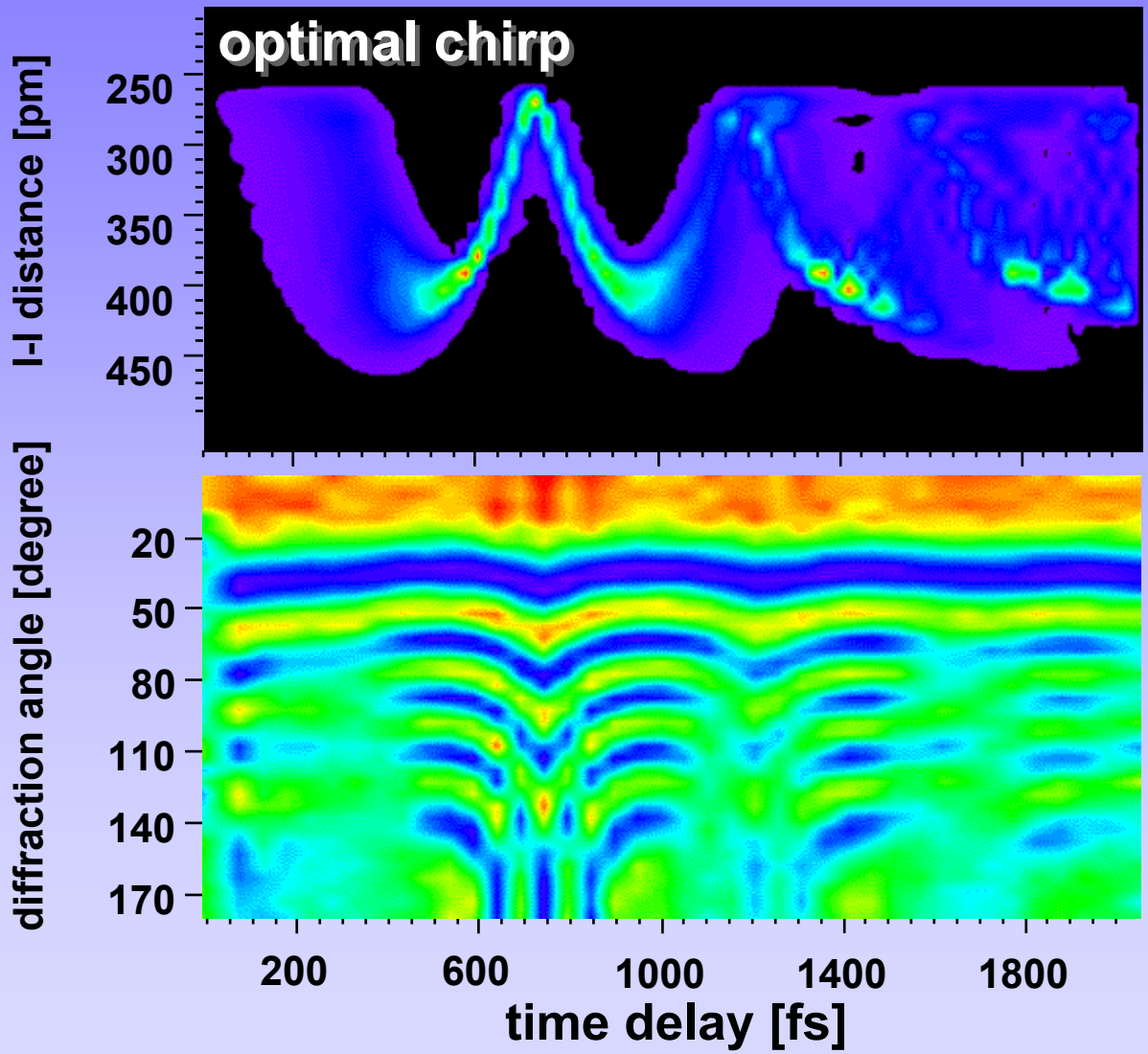
Army Medical Res. and Material Command

Argonne National Laboratory

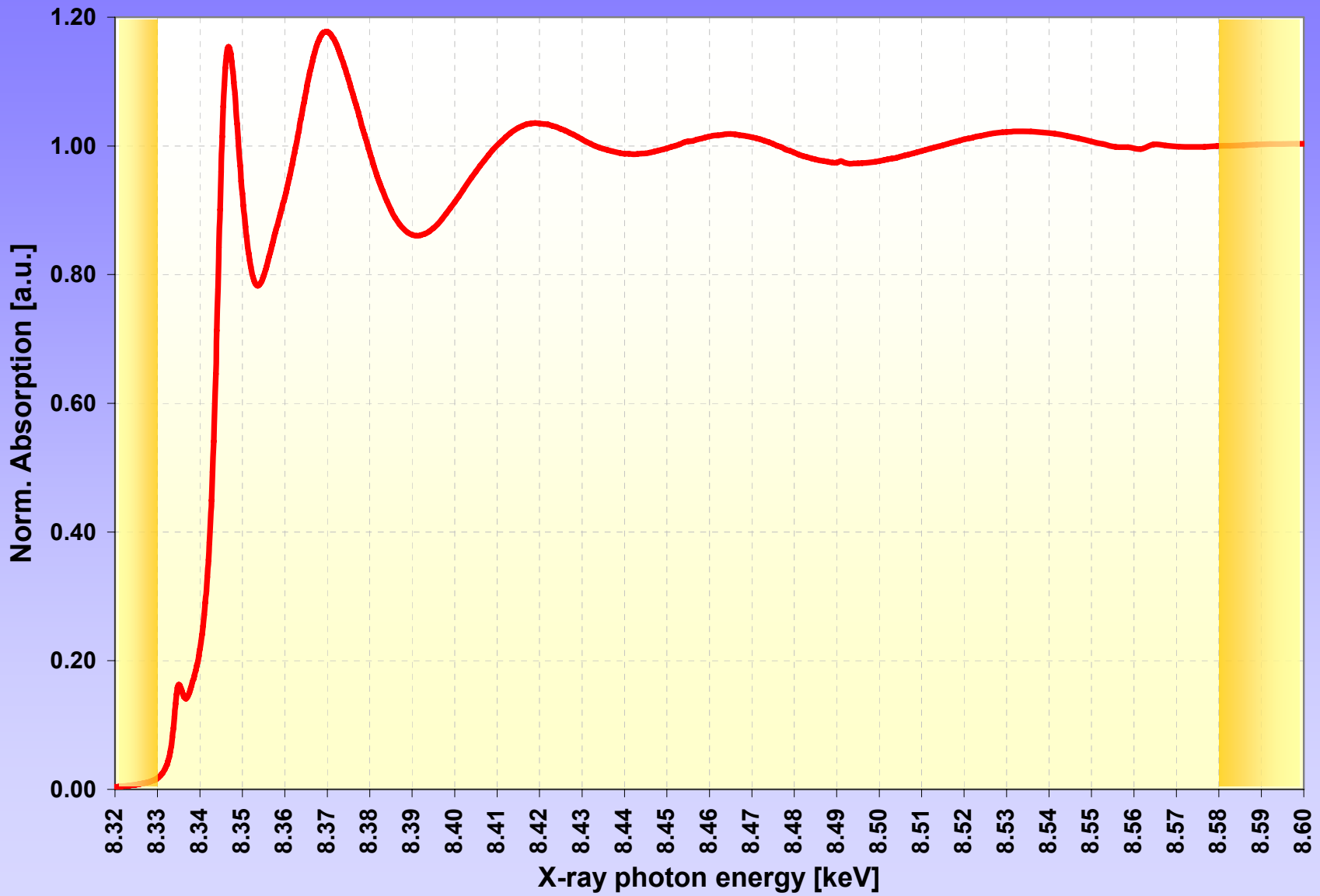
Andrew W. Mellon Foundation

Research Corporation

Nuclear motions and ultrafast x-ray diffraction off I_2



Calculated XAFS spectrum of $NiCO_4$



S/N calculation for EXAFS of solute at “ultrafast” Cornell ERL-II

Liquid beam parameters	
Beam thickness [um]	500
Pumped beam width (time axis) [mm]	0.01
Pumped beam height (wavelength axis) [mm]	0.01
Pumped beam height restriction factor (for measuring a restricted wavelength range with increased pump laser fluence) [%]	100.00%
Beam area [mm²]	0.0001
Solute number in "Solutes" Worksheet	4
Solute concentration [mol / L]	0.030
Solute name	Ni(CO)4
Molar absorption coefficient [L / (mol cm)]	500
Pump wavelength [nm]	267
Pump pulse energy [mJ]	1.E-04
Pump repetition rate [kHz]	1000
Average laser power [W]	100
Pump pulse fluence [mJ / cm²]	100.0
Pump pulselength [fs]	100
Pump pulse intensity [W / cm²]	1.0E+12

Results for sample excitation	
# of pump photons / pulse	1.34E+11
Transmitted pulse energy [mJ]	1.78E-05
Absorbed pulse energy [mJ]	8.22E-05
Transmittance	17.78%
Absorbance	82.22%
# of absorbed pump photons	1.11E+11
Pumped beam volume [mm^3]	0.00005
Concentration of exited molecules [mol / L]	3.67E-03
Concentration of unexcited molecules [mol / L]	2.63E-02
Excitation fraction [%]	12.23%

Solvent parameters	
X-ray transmission by solvent [%]	45.8%

Solute parameters	
Edge type	Ni-K
Post-edge x-ray mass absorption coefficient [cm² / g]	328.6
Pre-edge x-ray mass absorption coefficient [cm² / g]	44.24
Molar mass of x-ray absorbing solute atom [g / mol]	58.693
EXAFS modulation depth [%]	5.00%
Accuracy of measurement for excited solutes [%]	5.00%
X-ray fluorescence efficiency [%]	30.00%

X-ray spectrometer parameters	
Detection efficiency for transmission detection [%]	50.00%
Detection efficiency for fluorescence detection [%]	10.00%
# of x-ray photons on sample [1 / eV s]	7.65E+11
Desired spectral resolution (i.e. wave step) [eV]	1
Numer of desired time-steps	100
Spectral range over which the measurements are done [eV]	1000

Results for transmission detection	
X-ray transmission by solvent [%]	45.83%
X-ray absorbance by excited molecules [%]	0.35%
X-ray absorbance by unexcited molecules [%]	2.50%
Total x-ray absorbance due to solute molecules [%]	2.85%
Total x-ray transmittance due to solute molecules [%]	97.15%
Transmission modulation due to exited molecules	0.35%
Total x-ray transmittance due to solute and solvent [%]	44.52%
Required accuracy of measurement	8.72E-06
Min. S/N of x-ray measurement	114663.90
Min. required # of x-ray ph. on sample / wave-time step	5.91E+10
Min. required # of x-ray ph. on sample / (all times full spectral range)	5.91E+15
Total data acquisition time (all times 1000eV) [s]	16846.479
Total data acquisition time (all times 1000eV) [min]	280.77
Number of wave-channels measured simultaneously for full spectral range	1
Number of time-channels measured simultaneously	1
Total data acquisition time [h : m : s]	4:40:46

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