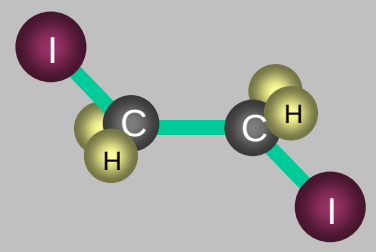
“Picosecond pump and probe experiments with CCD detectors on ID9B”

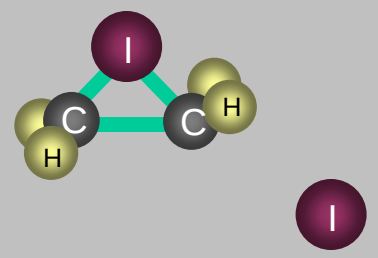
Michael Wulff, Maciej Lorenc, Qingyu Kong, Manuela Lo Russo, Marco Cammarata,
Wolfgang Reichenbach, Laurent Eybert, Laurent Claustre, Friedrich Schotte and Philip Anfinrud

The two-step dissociation of iodine from $C_2H_4I_2$ in methanol(Ihee et al.)

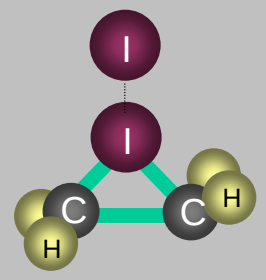
$$C_2H_4I_2 + h\nu(267nm) \rightarrow C_2H_4I_2^* \rightarrow C_2H_4I + I \rightarrow C_2H_4 + I_2$$



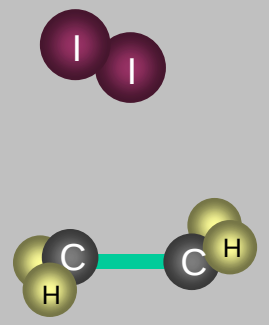
Initial state t< 0



Bridged state ~100 ps

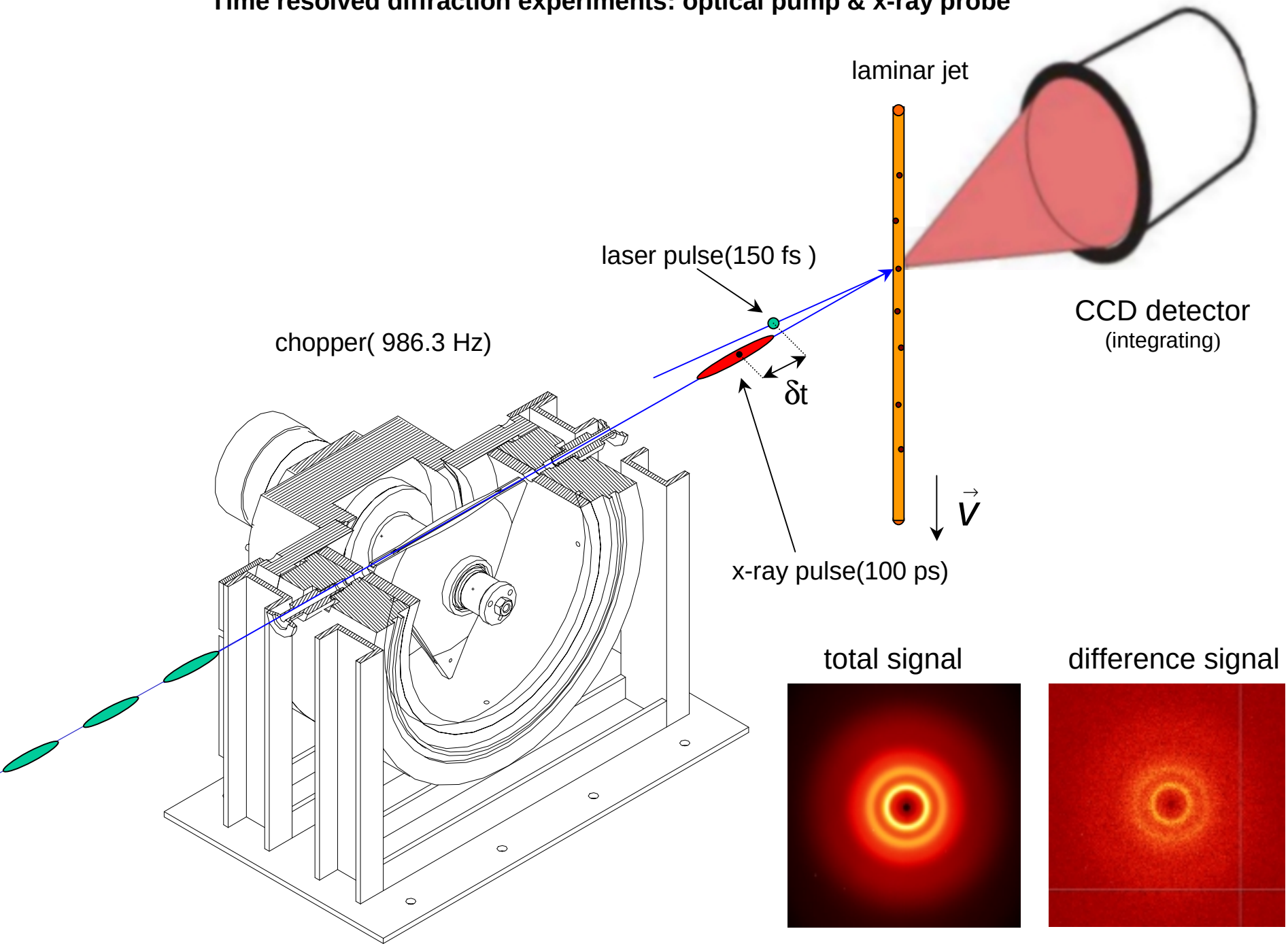


Abstraction ~10ns



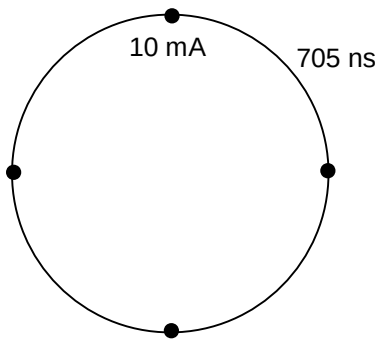
End state t > 10 ns

Time resolved diffraction experiments: optical pump & x-ray probe

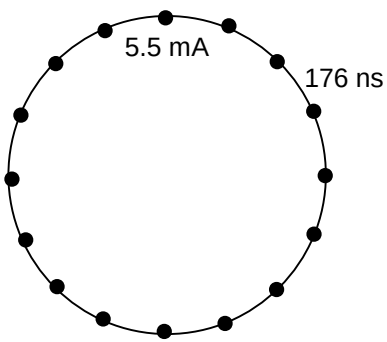


Bunch modes for timing experiments

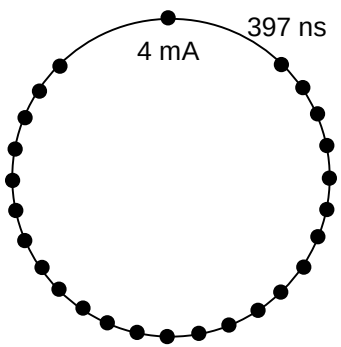
4-bunch mode



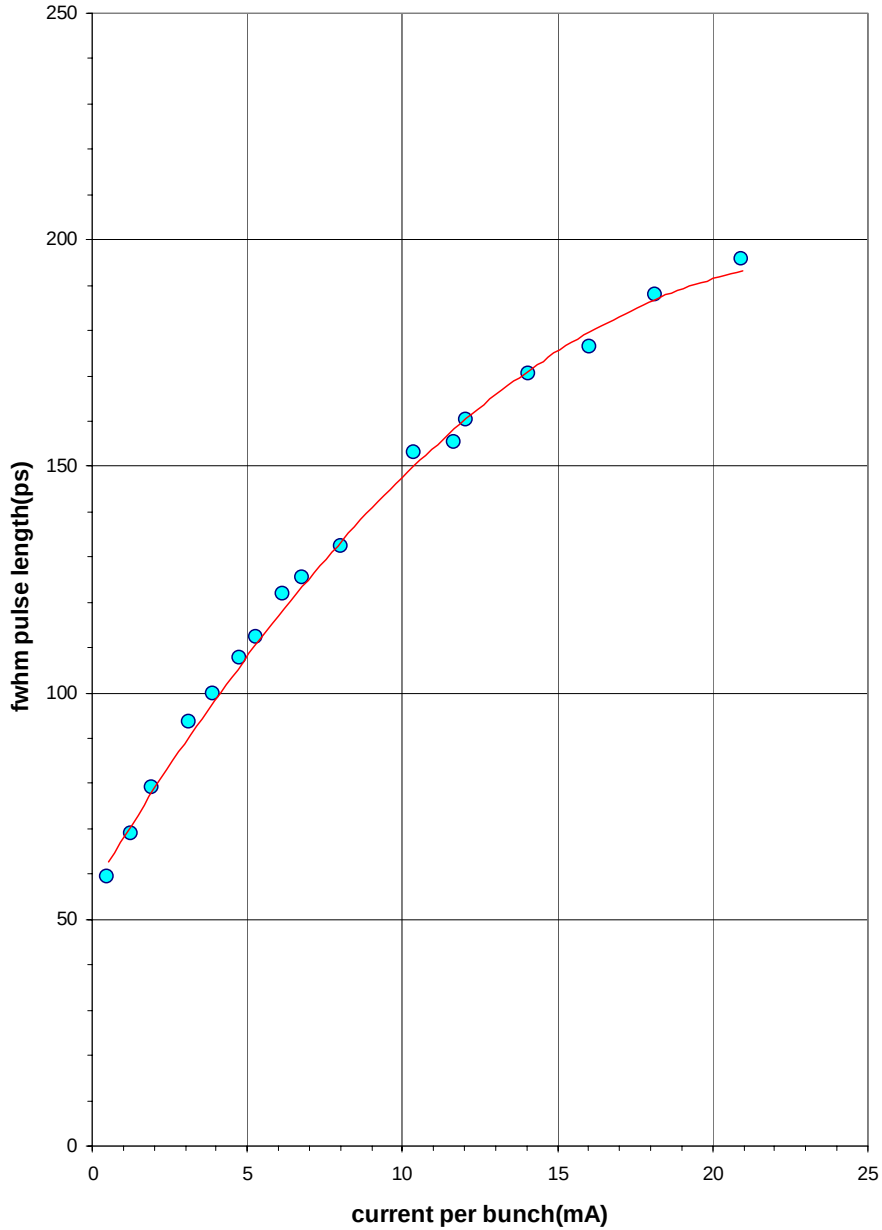
16-bunch mode



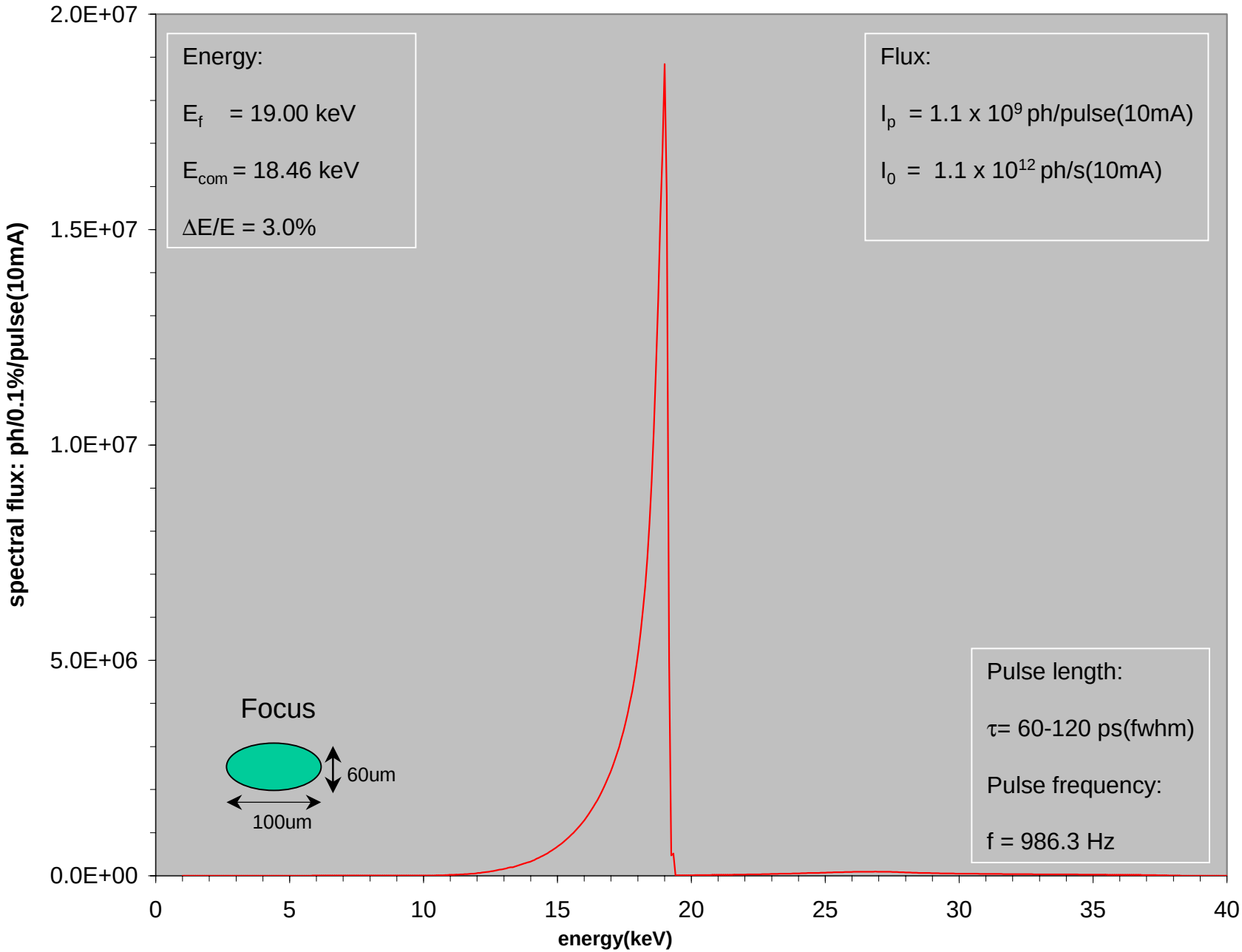
Hybrid mode: 24 x 8+1 bunches



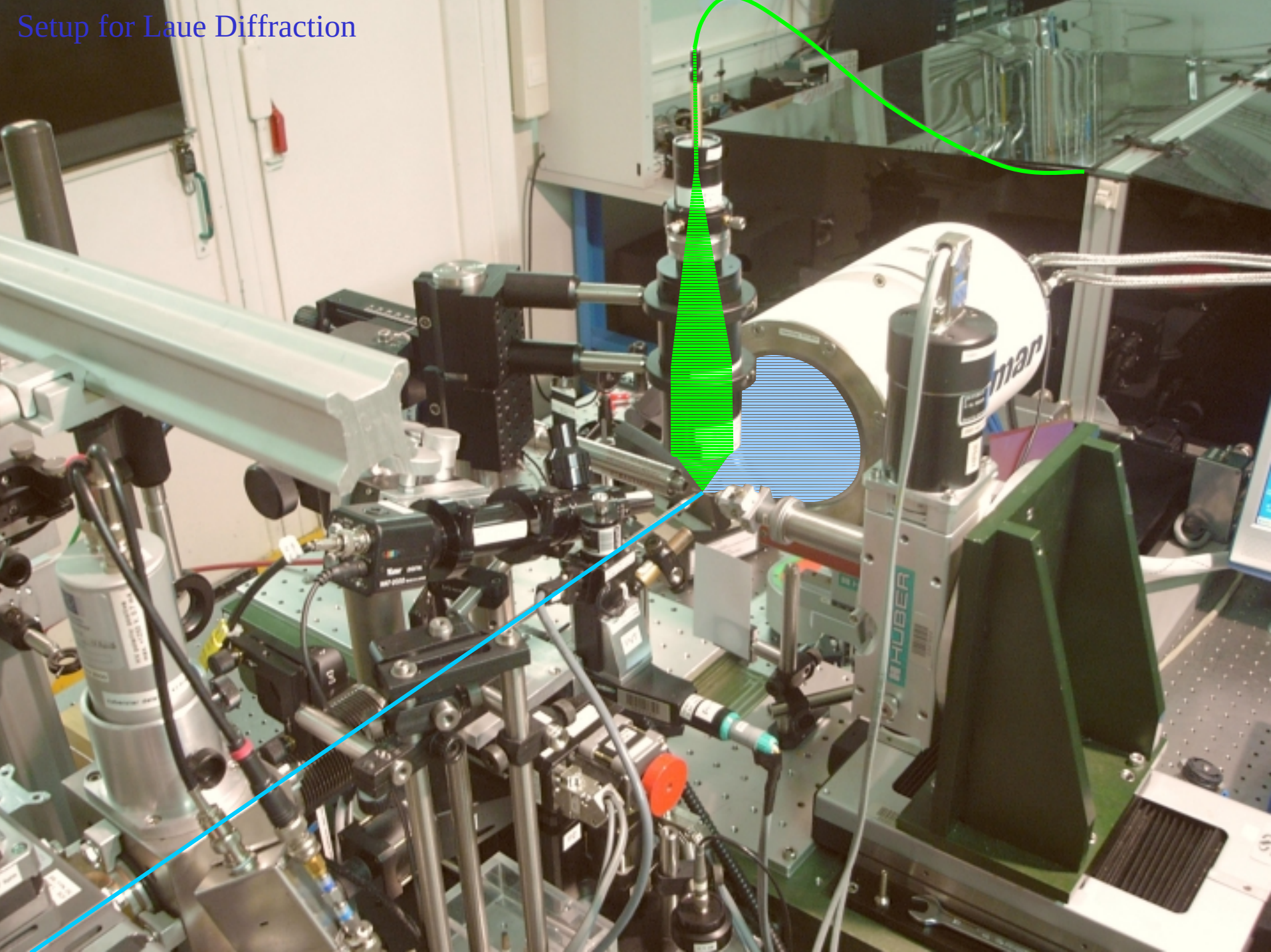
Pulse duration(fwhm)



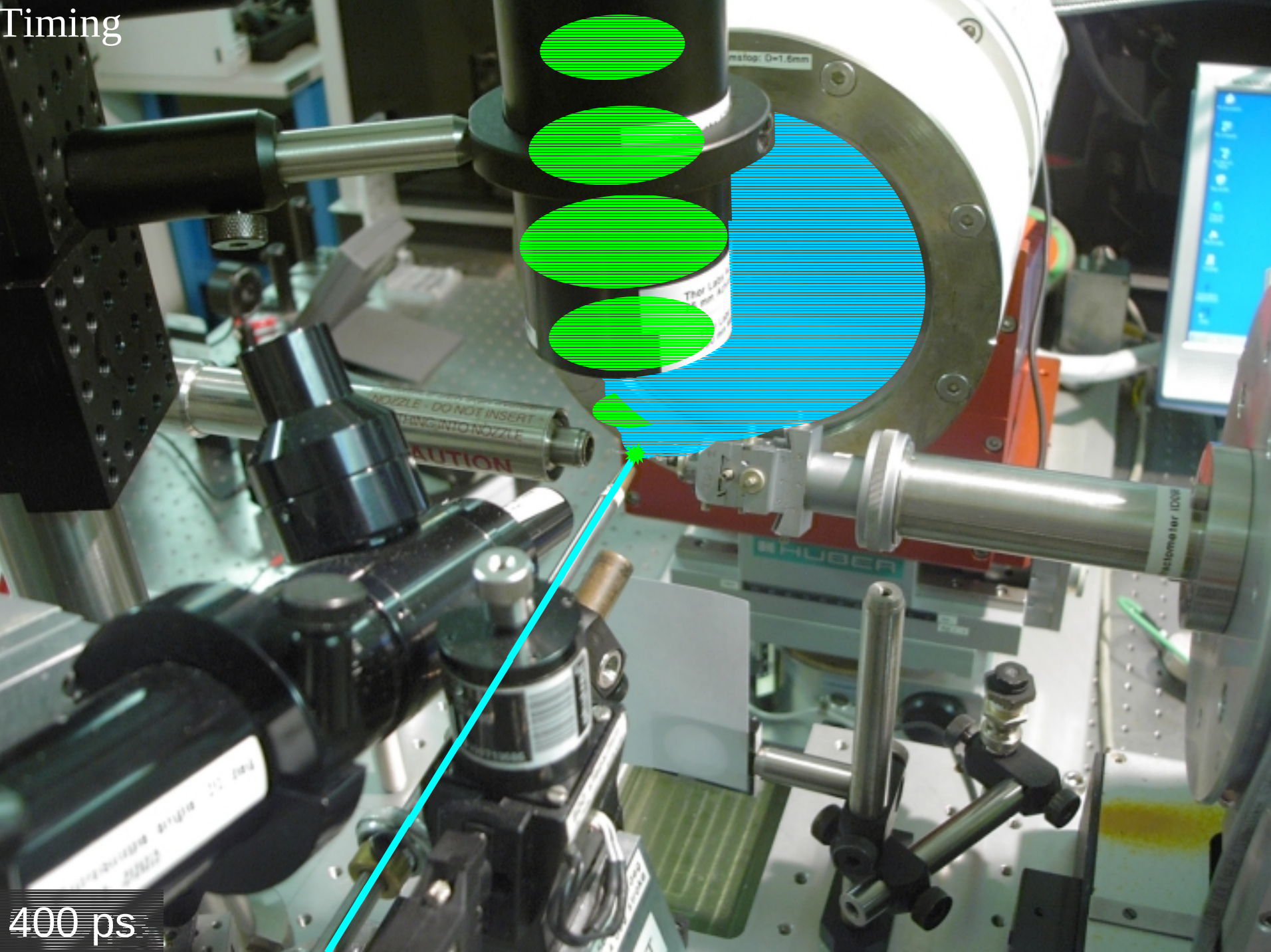
Spectrum of the mono-harmonic undulator U17



Setup for Laue Diffraction

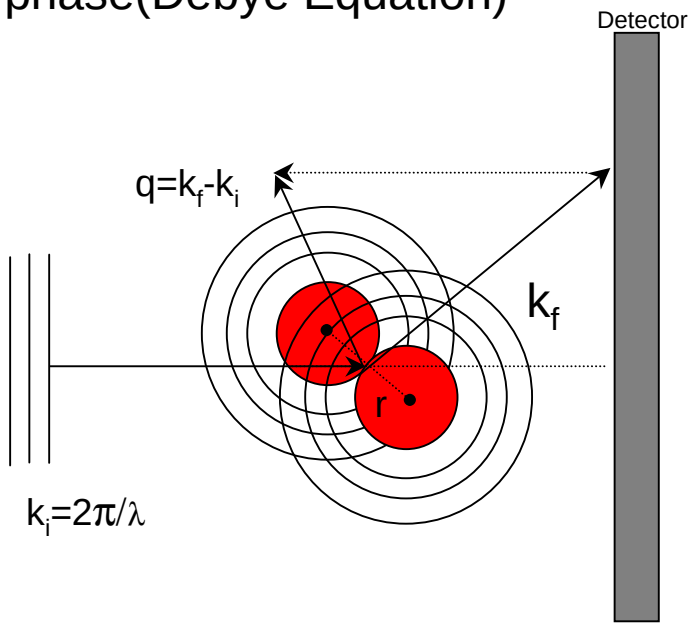


Timing

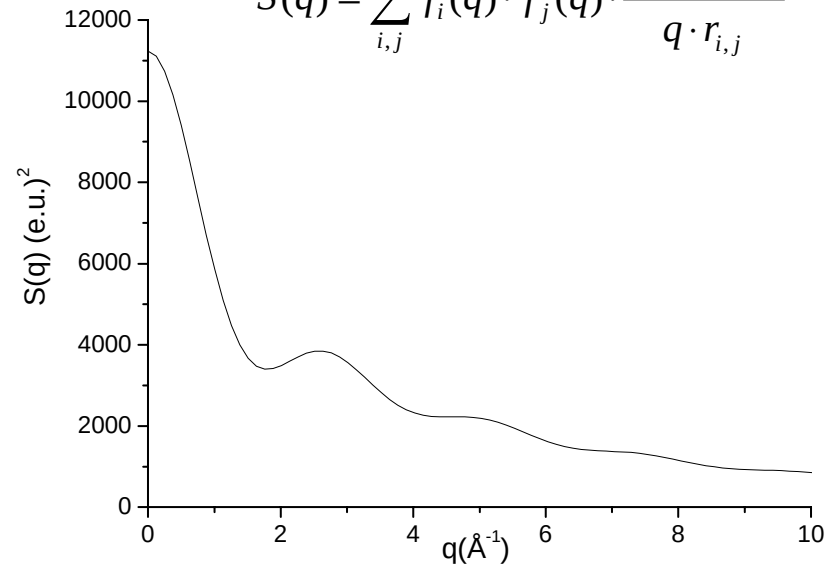


Diffracted Intensity S(q, t)

Gas-phase(Debye Equation)

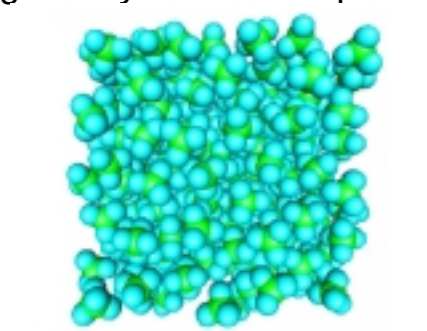


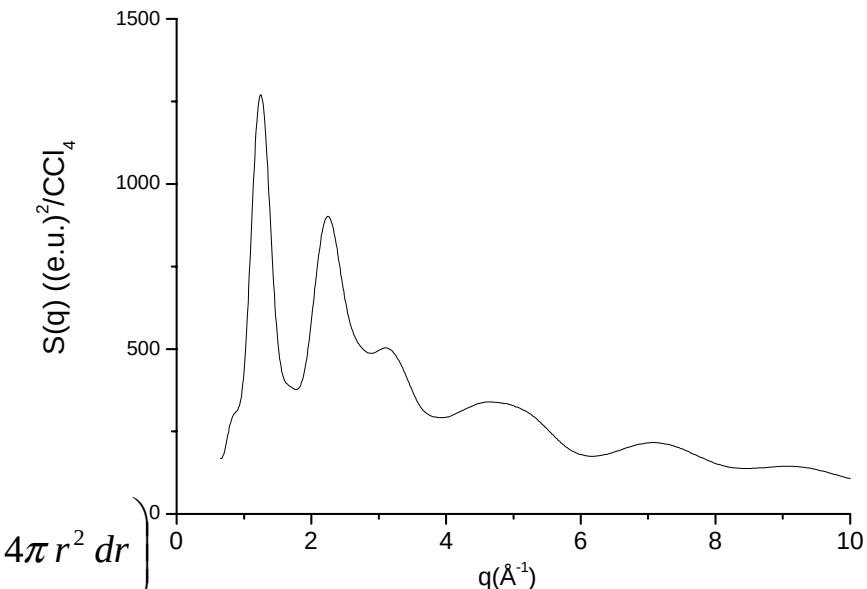
$$S(q) = \sum_{i,j} f_i(q) \cdot f_j(q) \cdot \frac{\sin(q \cdot r_{i,j})}{q \cdot r_{i,j}}$$



Liquid-phase(Zernike-Prinz Equation)

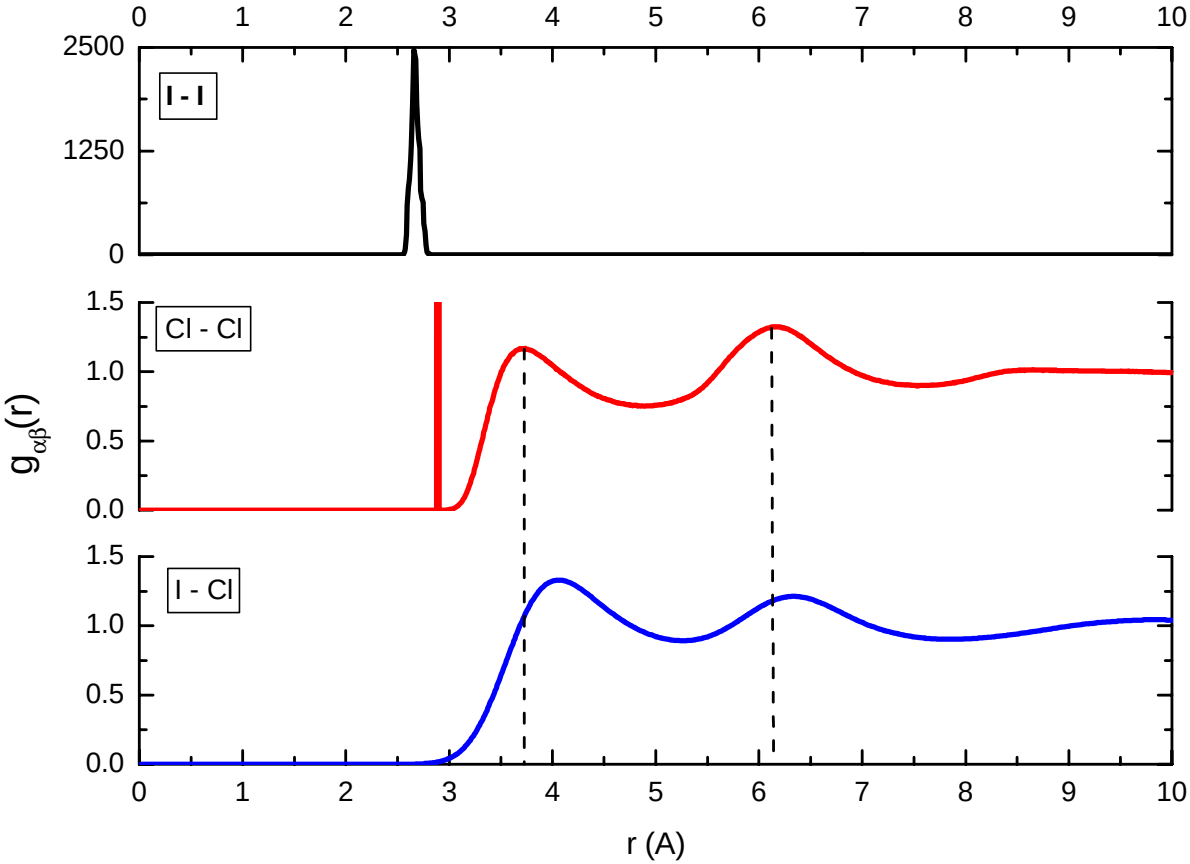
Positions given by atom-atom pair distributions $g_{\alpha\beta}(r)$

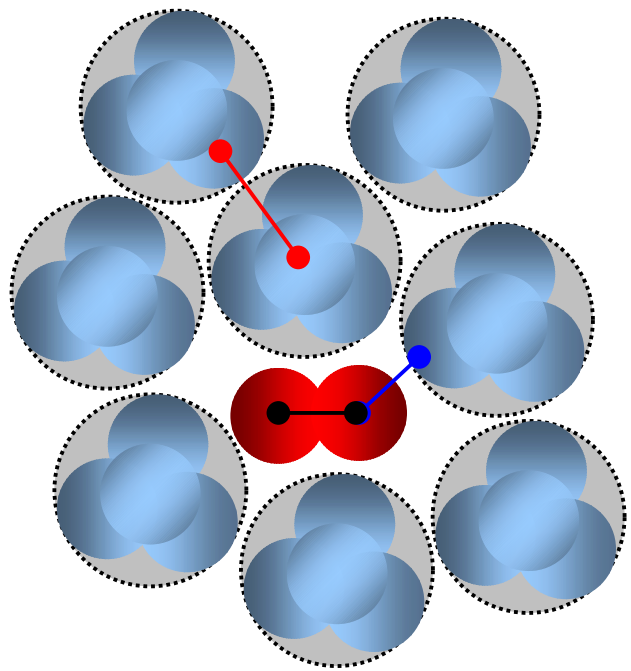




$$S(q,t) = \sum_{\alpha\beta} f_\alpha(q)\, f_\beta(q) \left(N_\alpha \delta_{\alpha\beta} + \frac{N_\alpha N_\beta}{V} \int_0^\infty g_{\alpha\beta}(r,t) \frac{\sin(q\,r)}{q\,r} 4\pi r^2\, dr \right)$$

The solvation of I_2 in CCl_4(1:500)





We would like to measure the change in:

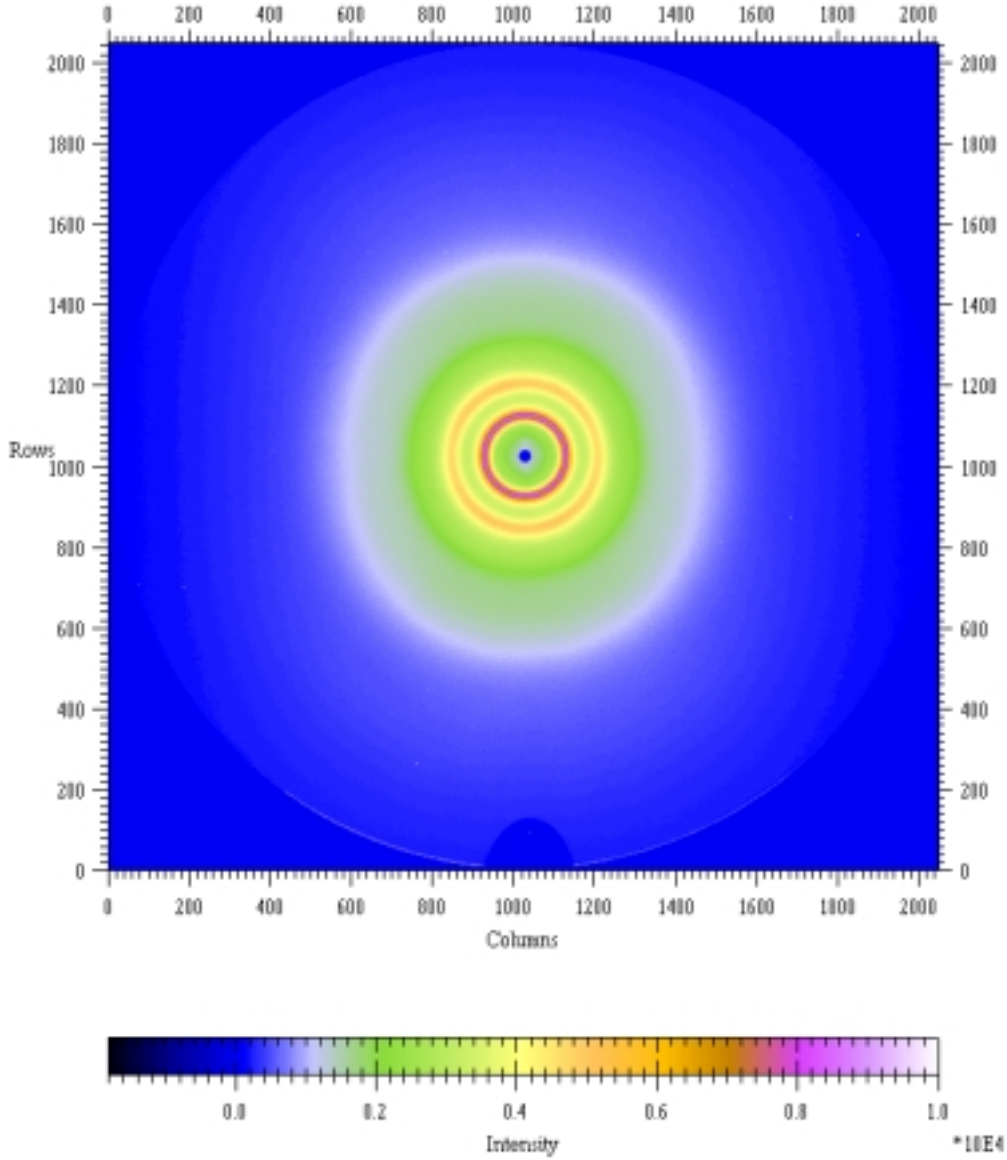
1) The solute structure(I_2)

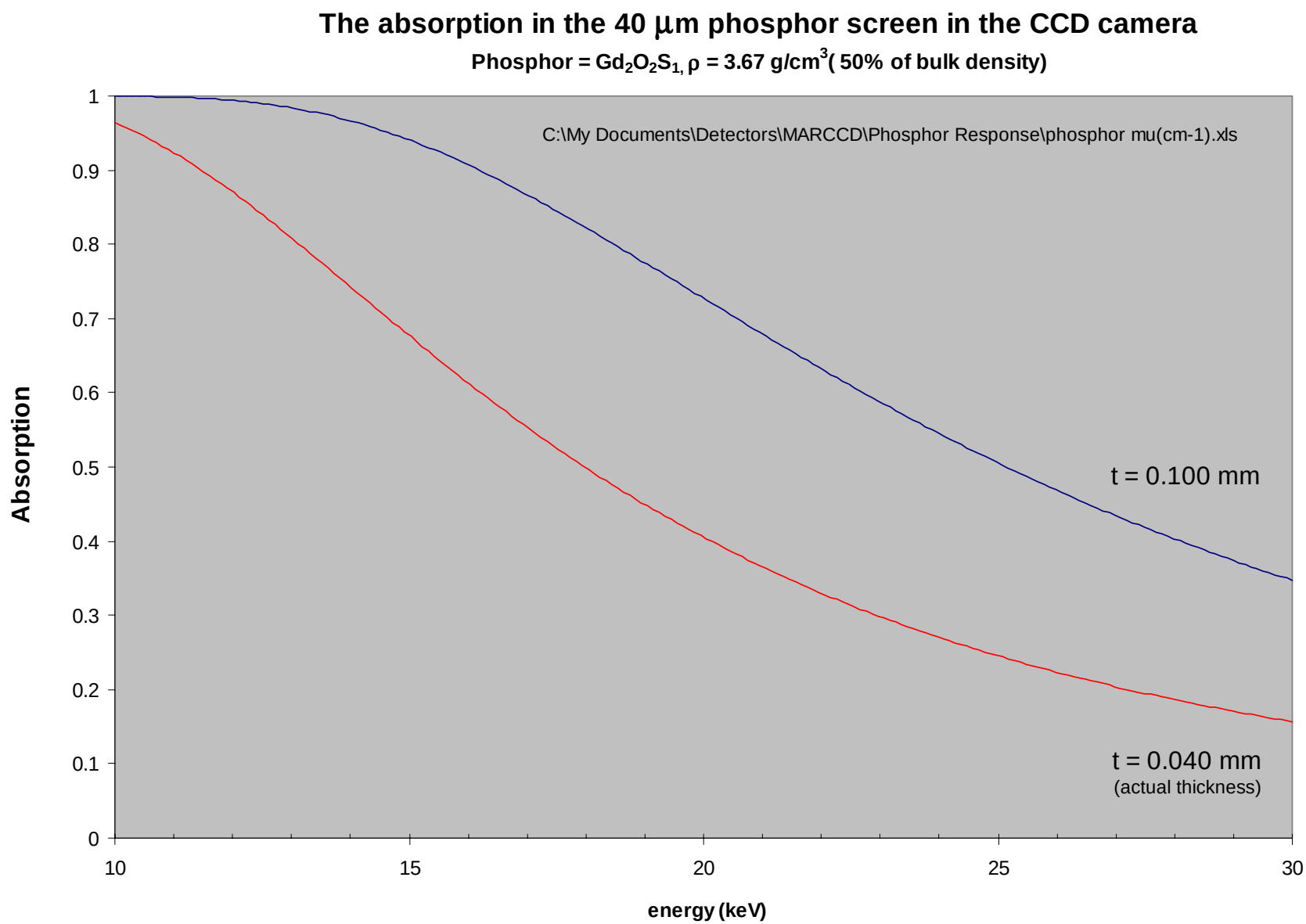
2) The cage structure(I_2..Cl)

3) The bulk solvent structure(CCl_4)

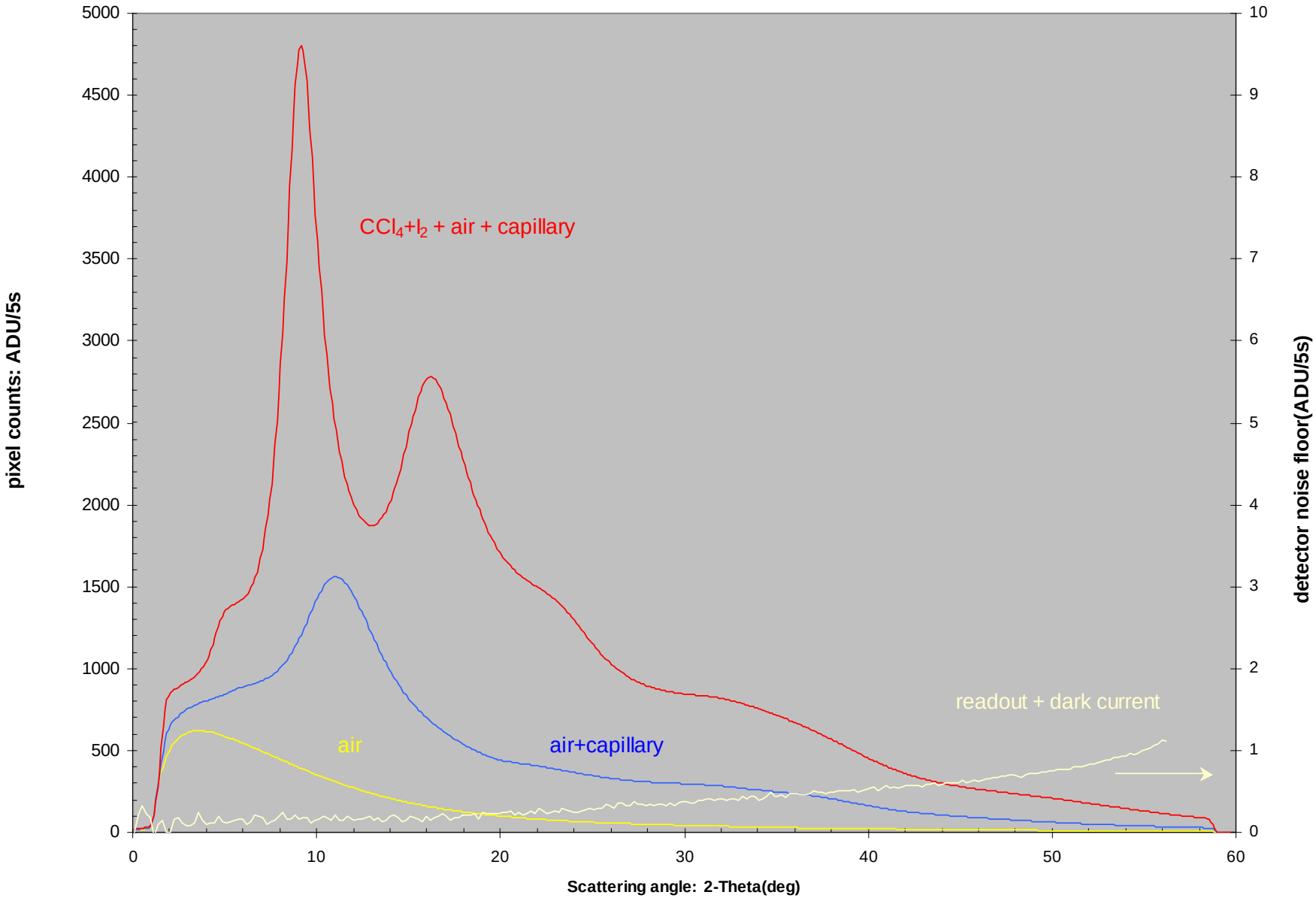
Characteristics of the 133mm diameter MARCCD

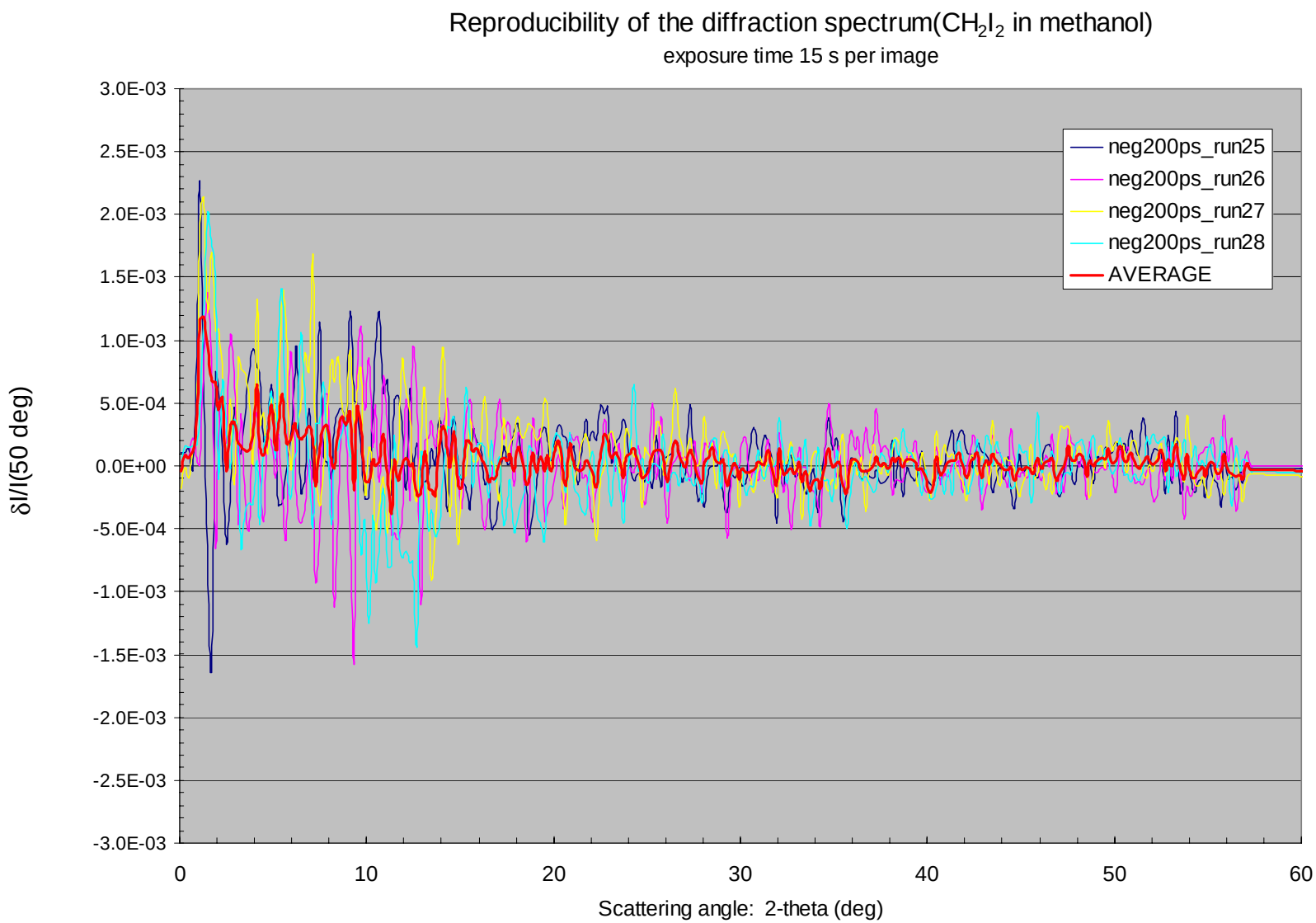
Input diameter(mm)	133
Pixel array	2048 x 2048
CCD dimension(mm)	61 x 61
Taper defocusing	46%
Pixel size(um)	64.276
Thickness of phosphor screen(um)	40
DQE(18keV)	52%
Dynamic range(bits)	16
Readout speed(s)	6.3
Image correction speed(s)	0.4
Image saving speed(s)	0.9
Noise levels	
Dark current(ADU/pixel)	0.1
Readout noise(ADU/pixel/s)	0.01




The absorption in the 40 µm phosphor screen in the CCD camera
Phosphor = $Gd_2O_2S_1$, ρ = 3.67 g/cm^3(50% of bulk density)
C:\My Documents\Detectors\MARCCD\Phosphor Response\phosphor mu(cm-1).xls
t = 0.100 mm
t = 0.040 mm
(actual thickness)
Absorption
energy (keV)
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
10
15
20
25
30

Contributions to the radial signal on the CCD detector



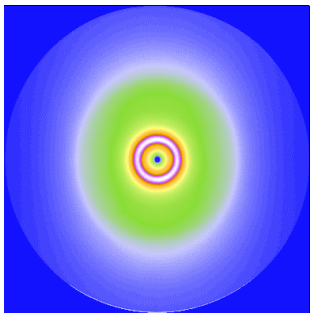

Reproducibility of the diffraction spectrum(CH_2I_2 in methanol)
exposure time 15 s per image
δI/I(50 deg)
3.0E-03
2.5E-03
2.0E-03
1.5E-03
1.0E-03
5.0E-04
0.0E+00
-5.0E-04
-1.0E-03
-1.5E-03
-2.0E-03
-2.5E-03
-3.0E-03
0
10
20
30
40
50
60
Scattering angle: 2-theta (deg)
neg200ps_run25
neg200ps_run26
neg200ps_run27
neg200ps_run28
AVERAGE

Data collection strategy in pump and probe liquid diffraction

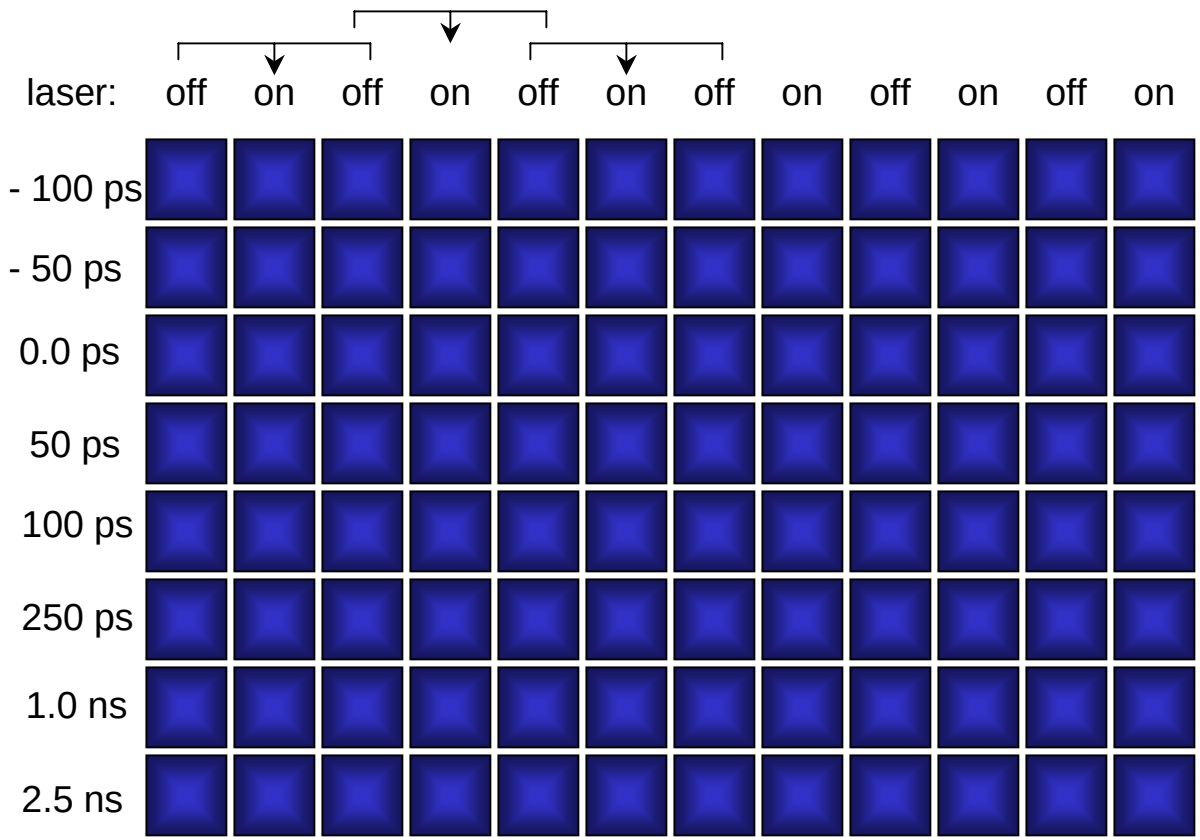
aim: minimize effects of drifts in spectrum, position and sample.

diff = on - ½ (off1 + off2)

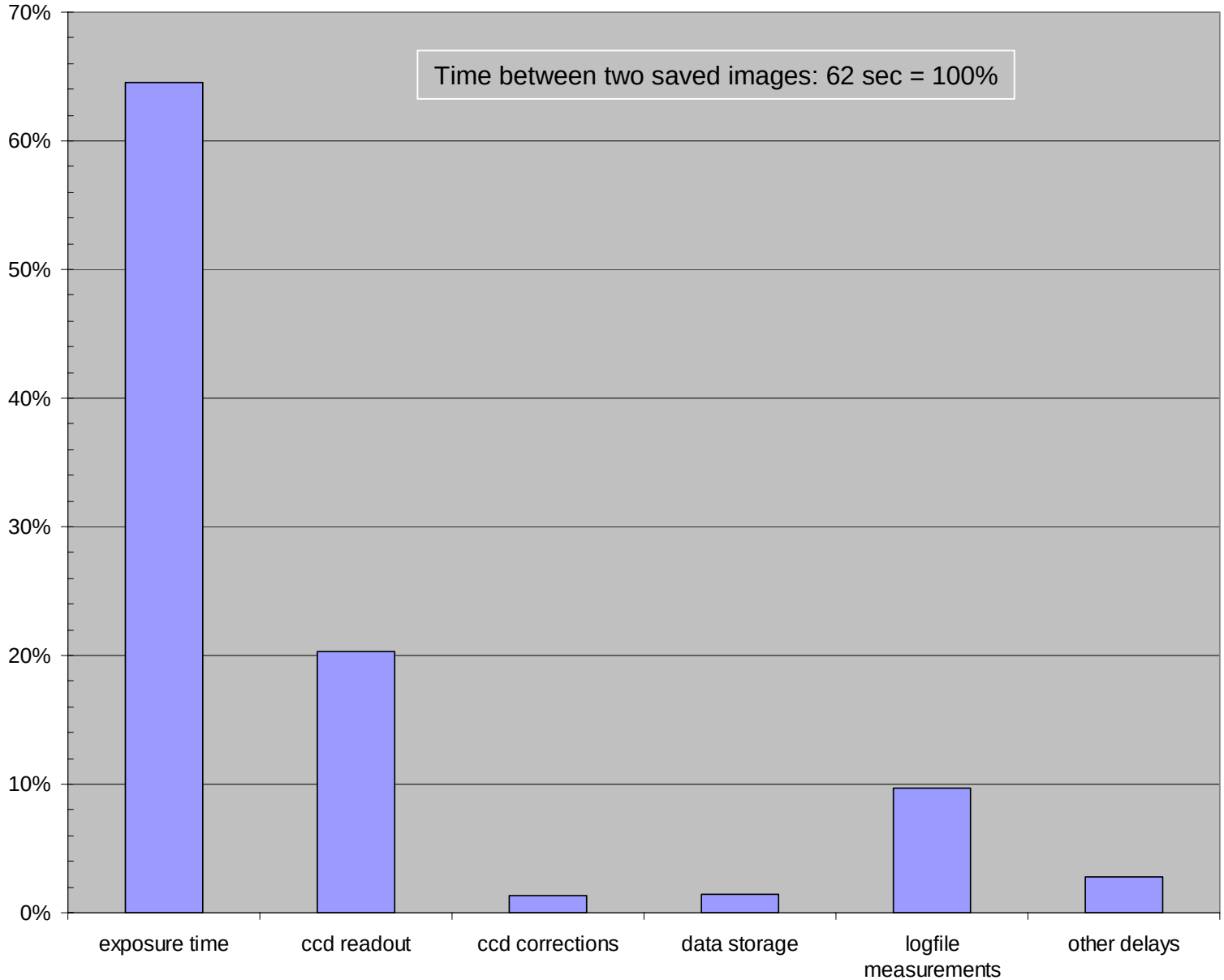
CCD-frame

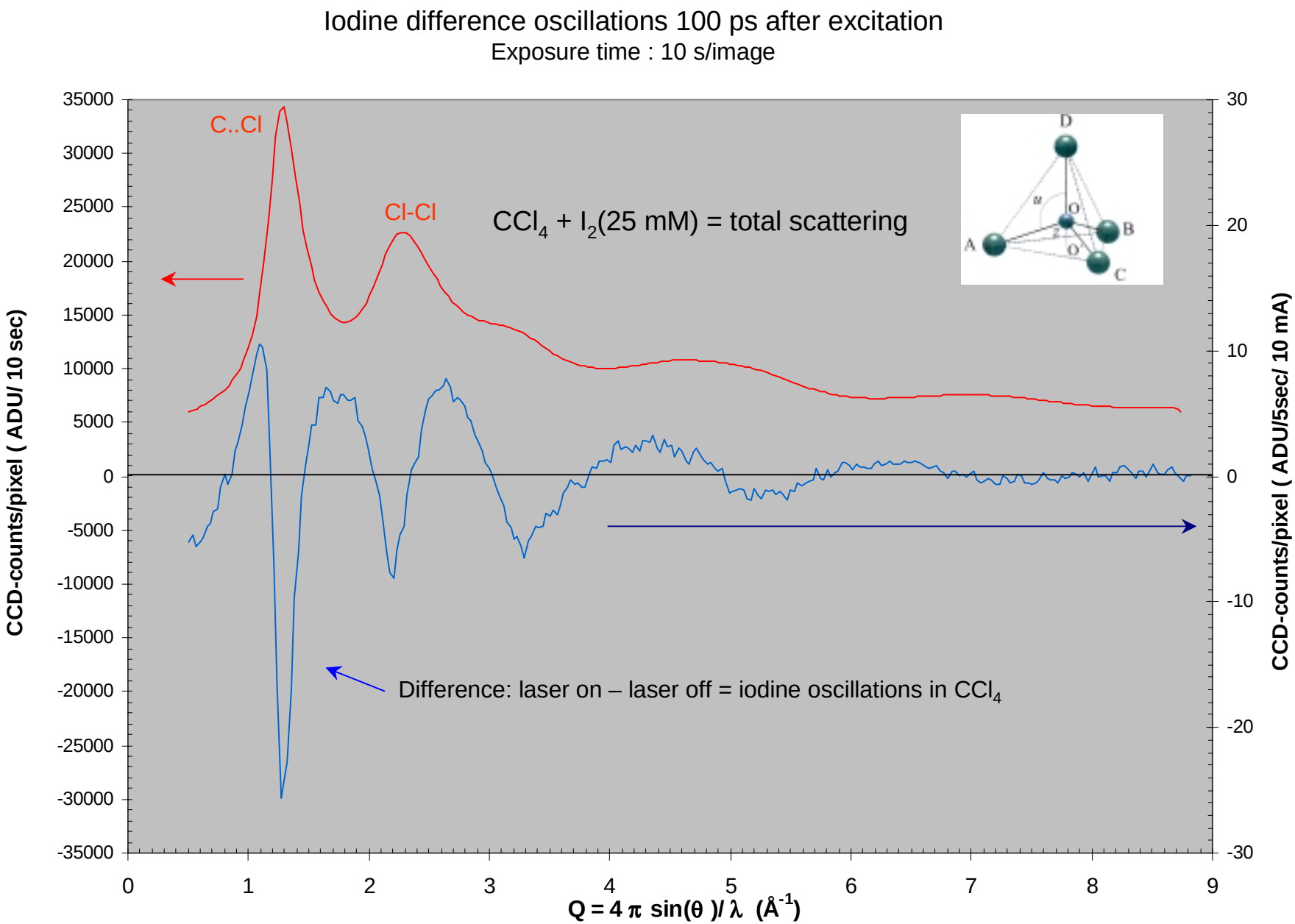


laser - xray delay

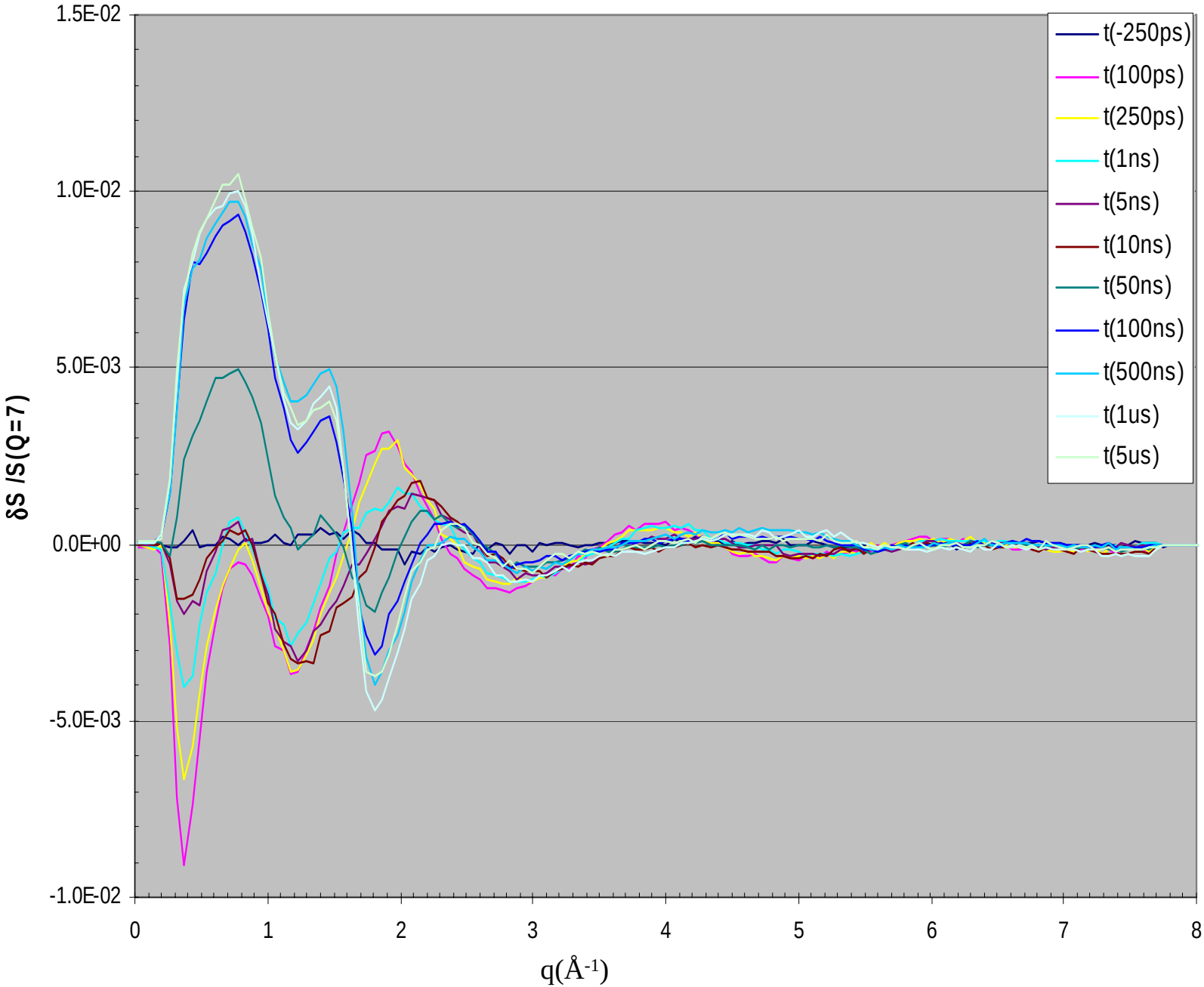


The efficiency of the MARCCD in a liquid diffraction experiment(with zinger suppression)
(from: $C_2H_4I_2$ in methanol, 16 bunch, December 2004)

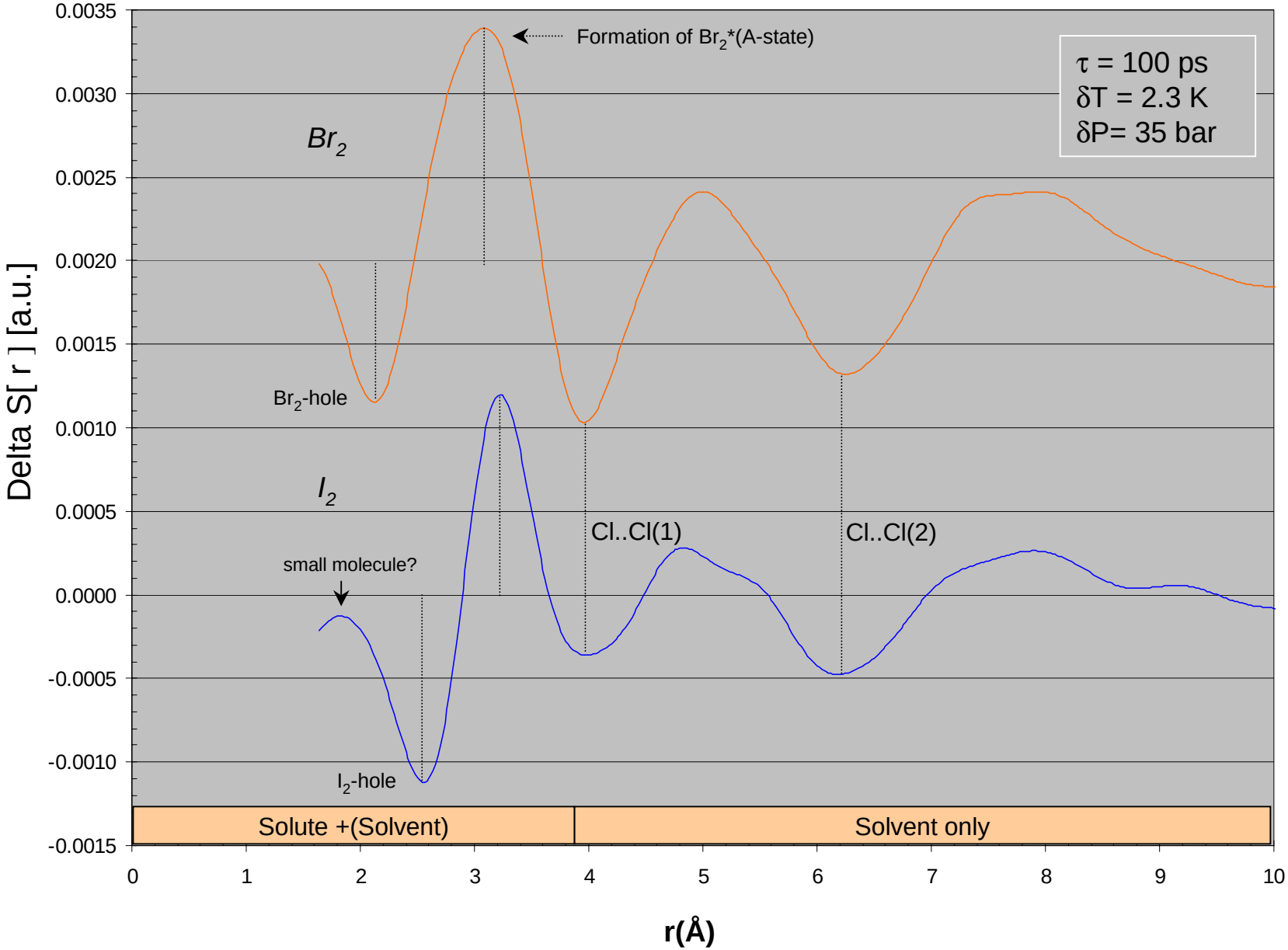



Iodine difference oscillations 100 ps after excitation
Exposure time : 10 s/image
C..Cl
Cl-Cl
CCl4 + I2(25 mM) = total scattering
Difference: laser on – laser off = iodine oscillations in CCl4
CCD-counts/pixel (ADU/ 10 sec)
CCD-counts/pixel (ADU/5sec/ 10 mA)
Q = 4 π sin(θ)/ λ (Å-1)
35000
30000
25000
20000
15000
10000
5000
0
-5000
-10000
-15000
-20000
-25000
-30000
-35000
30
20
10
0
-10
-20
-30
0
1
2
3
4
5
6
7
8
9
D
O
A
B
C

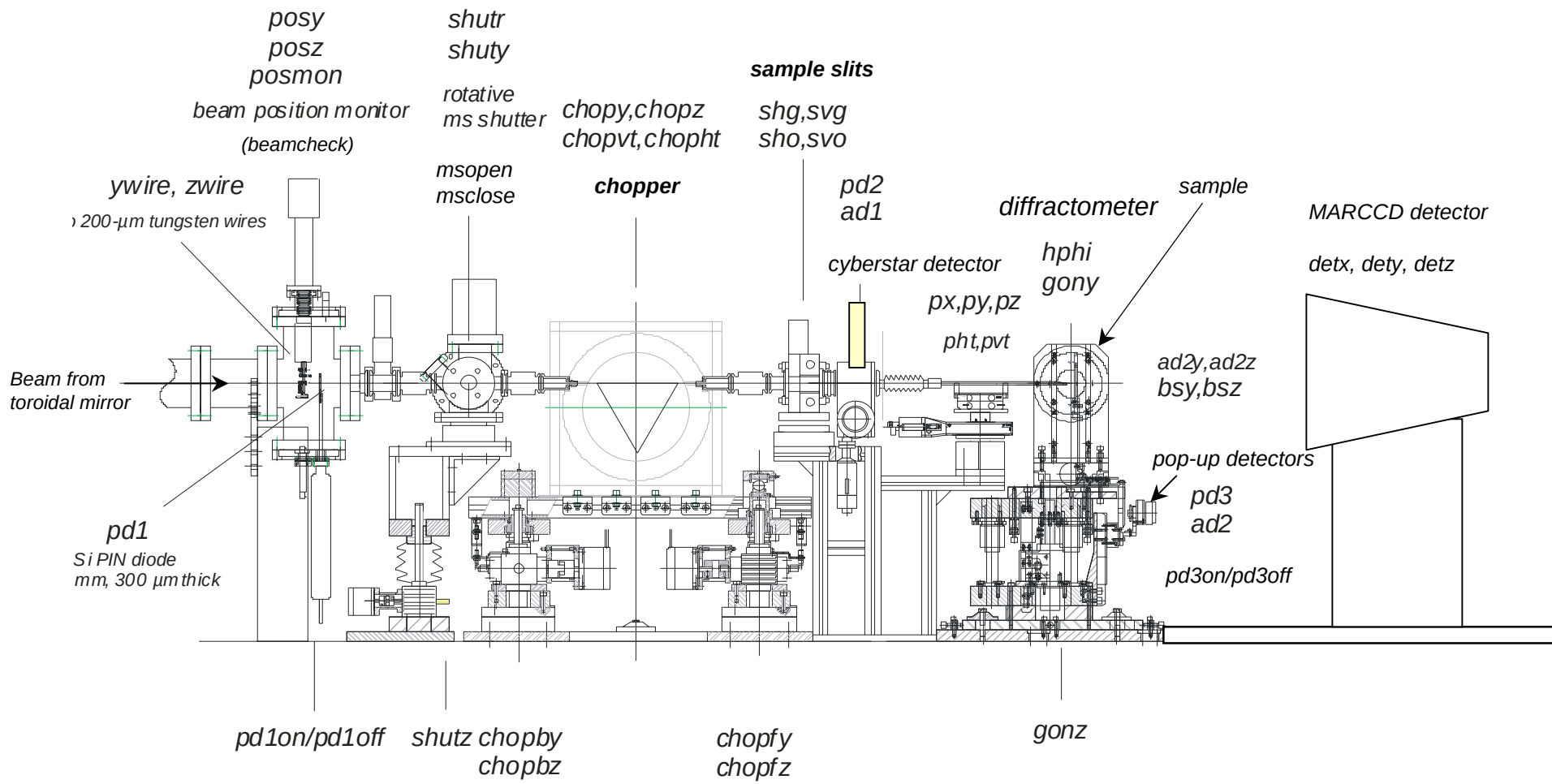
Difference oscillations from I_2* in Methanol(CH_3OH)



Radial maps of the recombination of Br_2 and I_2 in CCl_4 after 100 ps

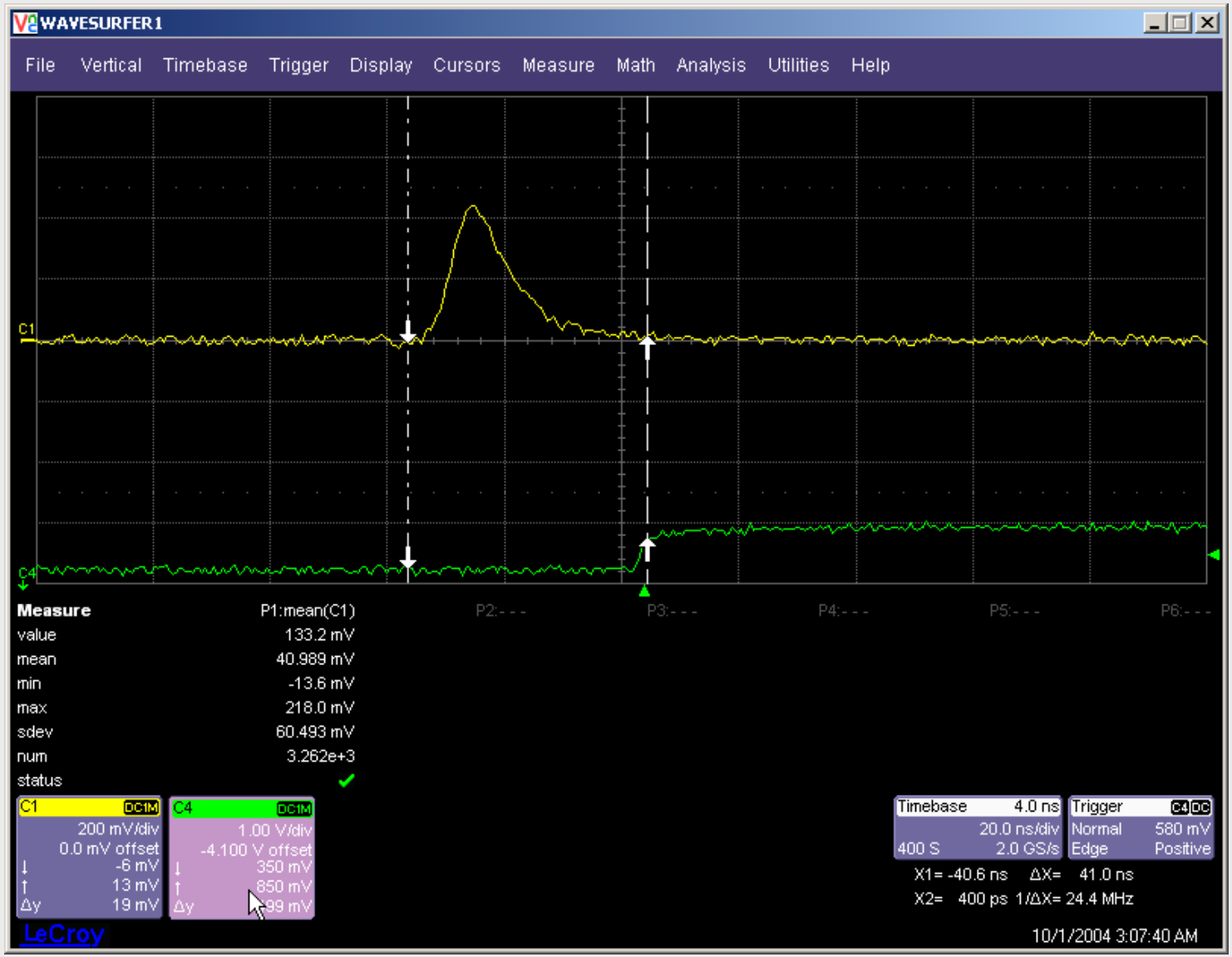


Components on the experimental table in EH2



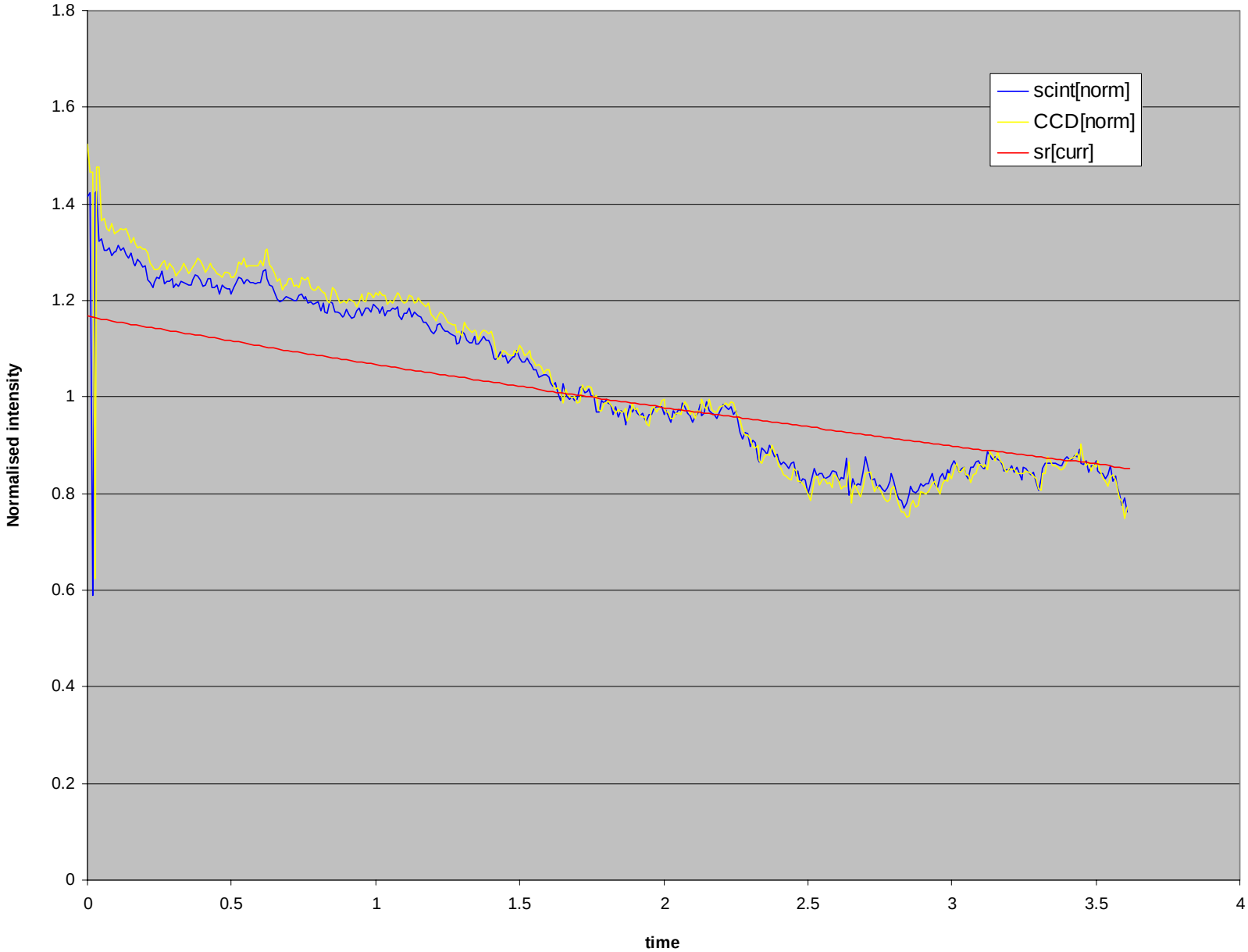
Measuring the intensity of the 1kHz x-ray beam with a Cyberstar scintilator(air scattering from white beam).

The Cyberstar/oscilloscope integrates a 120 Hz sub-train of pulses.

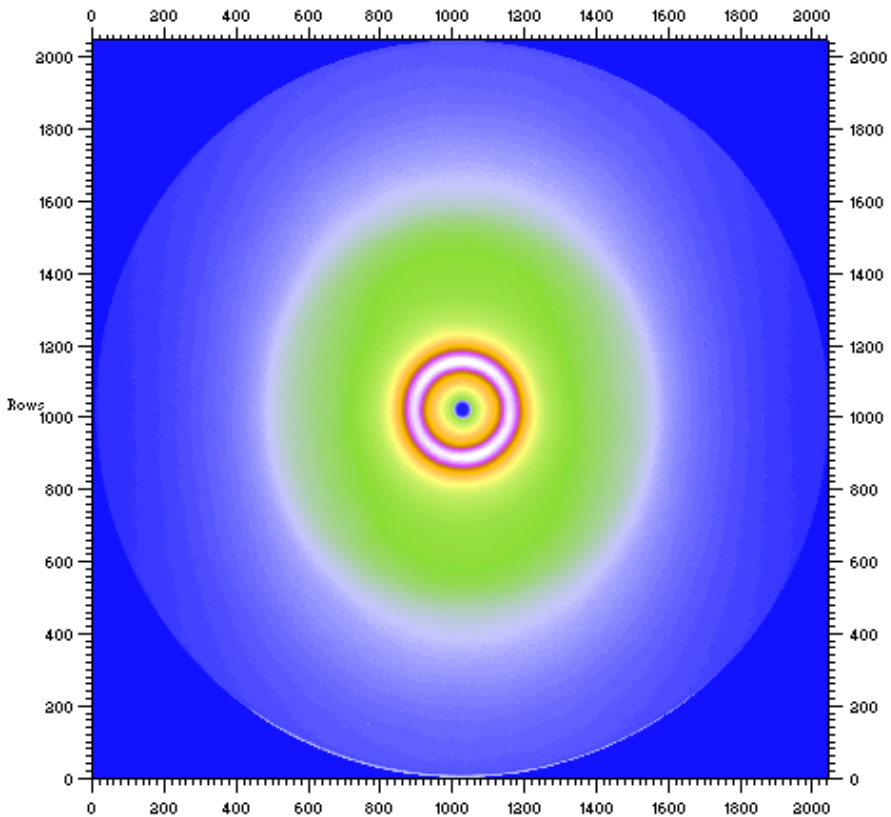


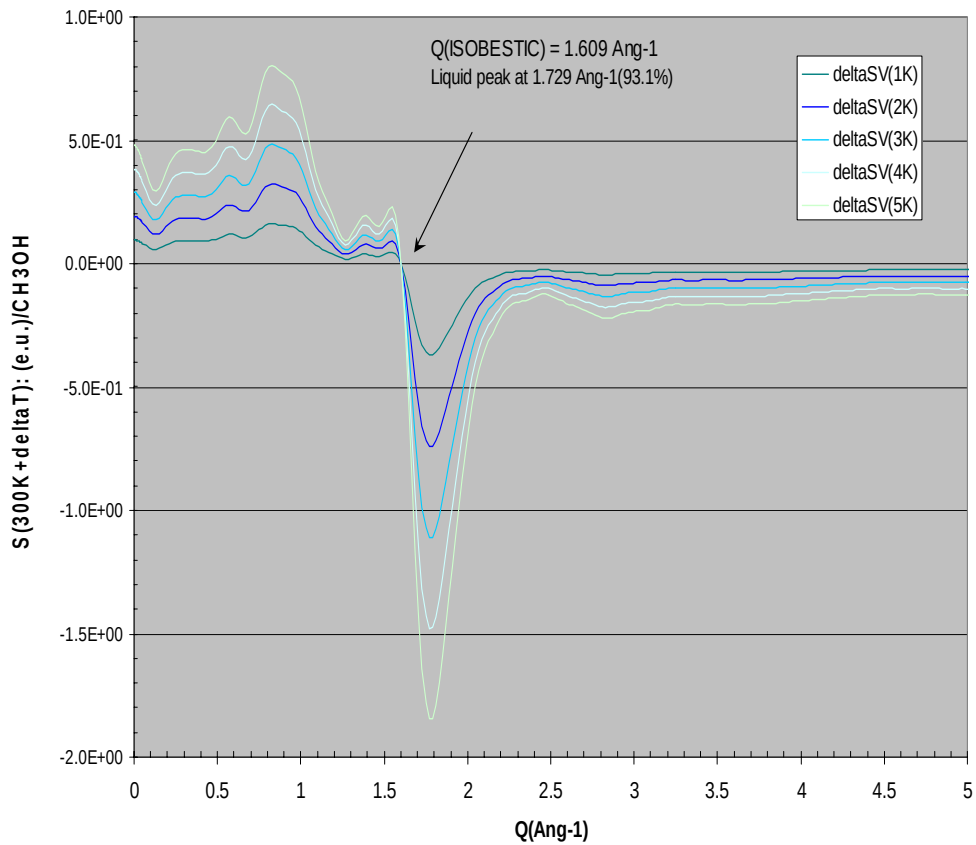
Trying to keep the CCD images at constant amplitude during the data collection:

scaling the exposure time to the intensity of the actual beam on the sample.



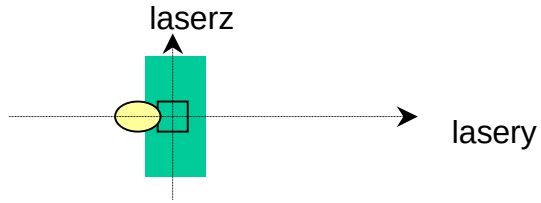
Using ring integrals to check the spatial overlap of the laser excitation in real time.

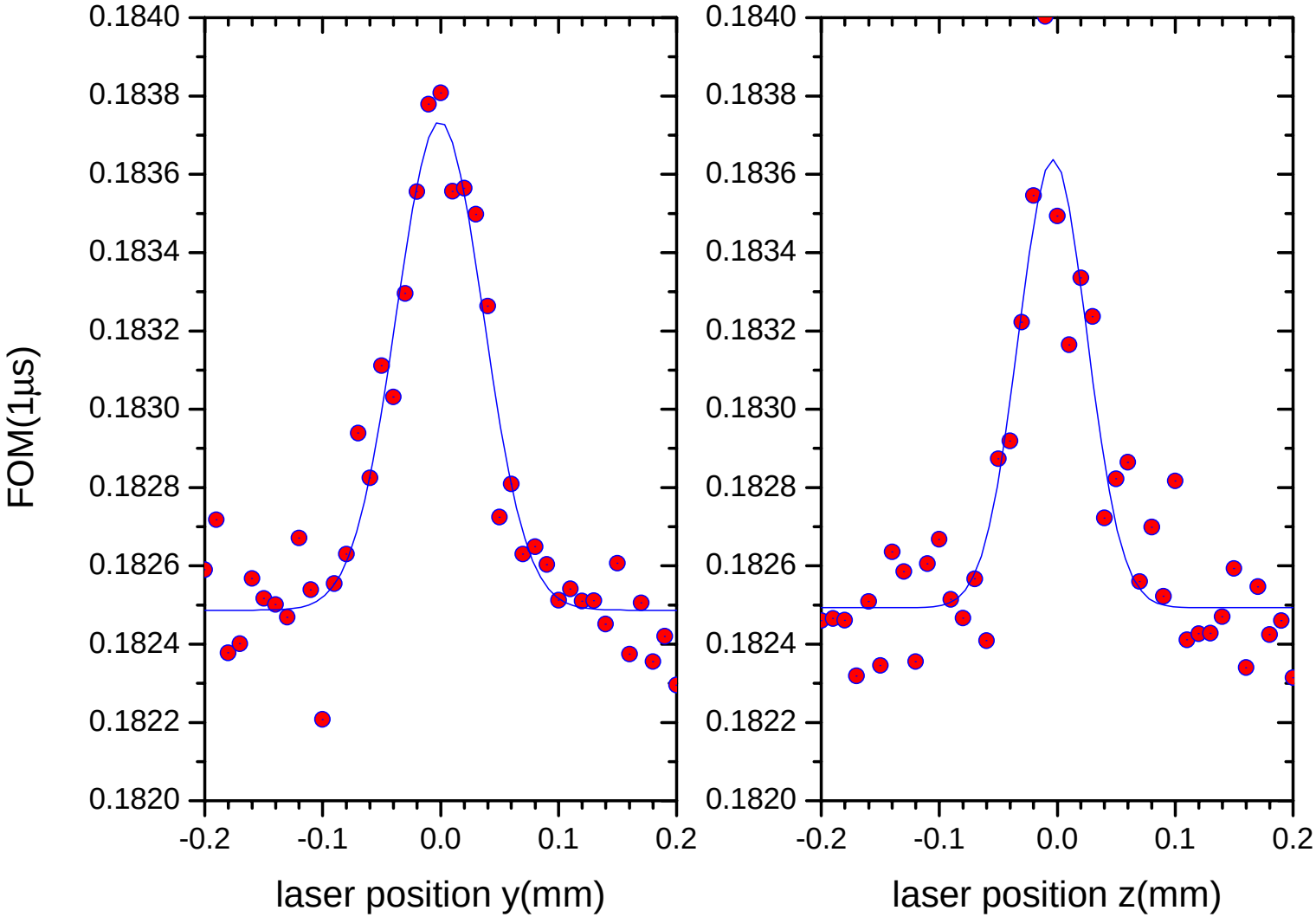




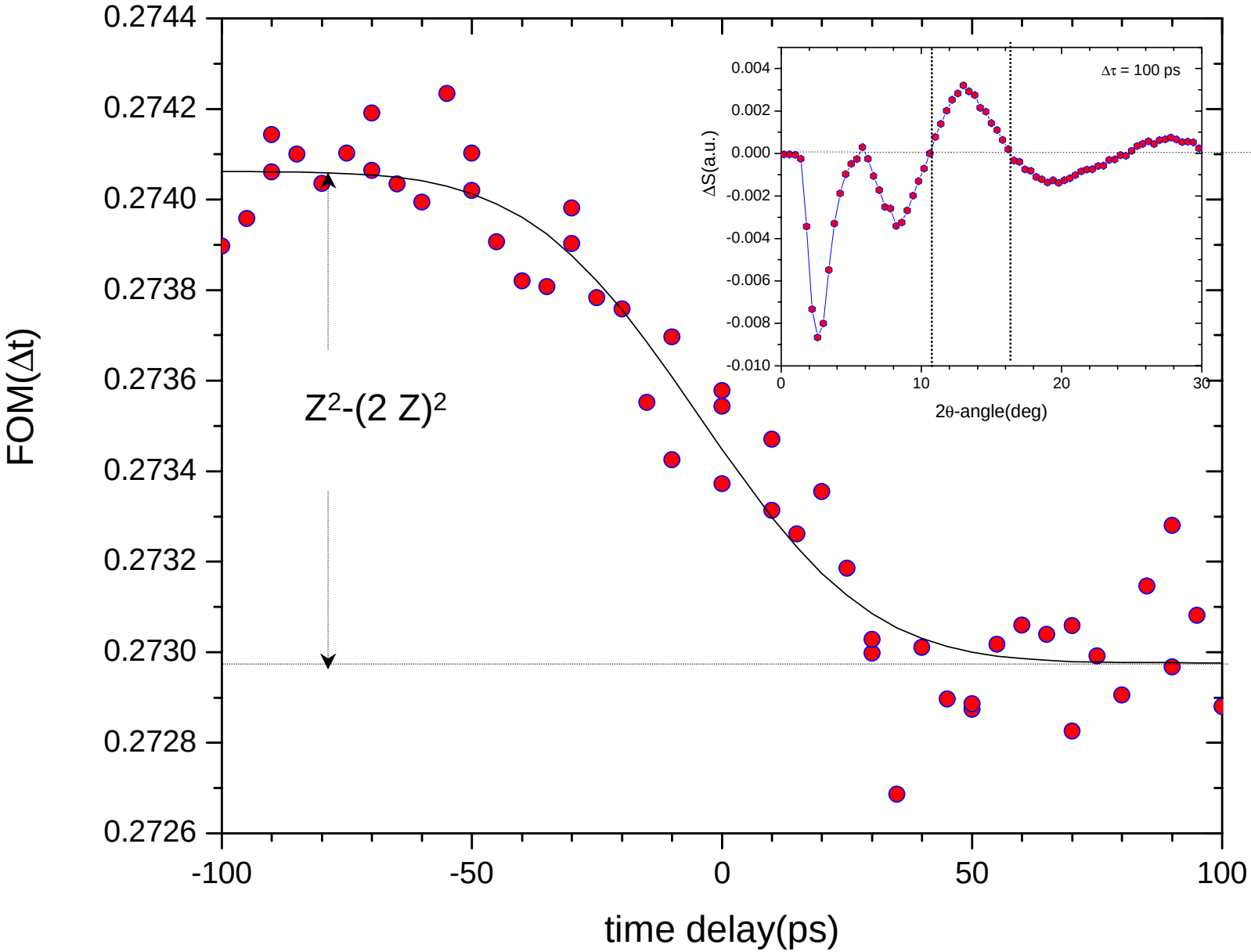
Bringing the laser spot onto the x-ray volume

$I_2 + h\nu \rightarrow I_2^* \rightarrow \ldots \rightarrow I_2$ + heat transfer to solvent

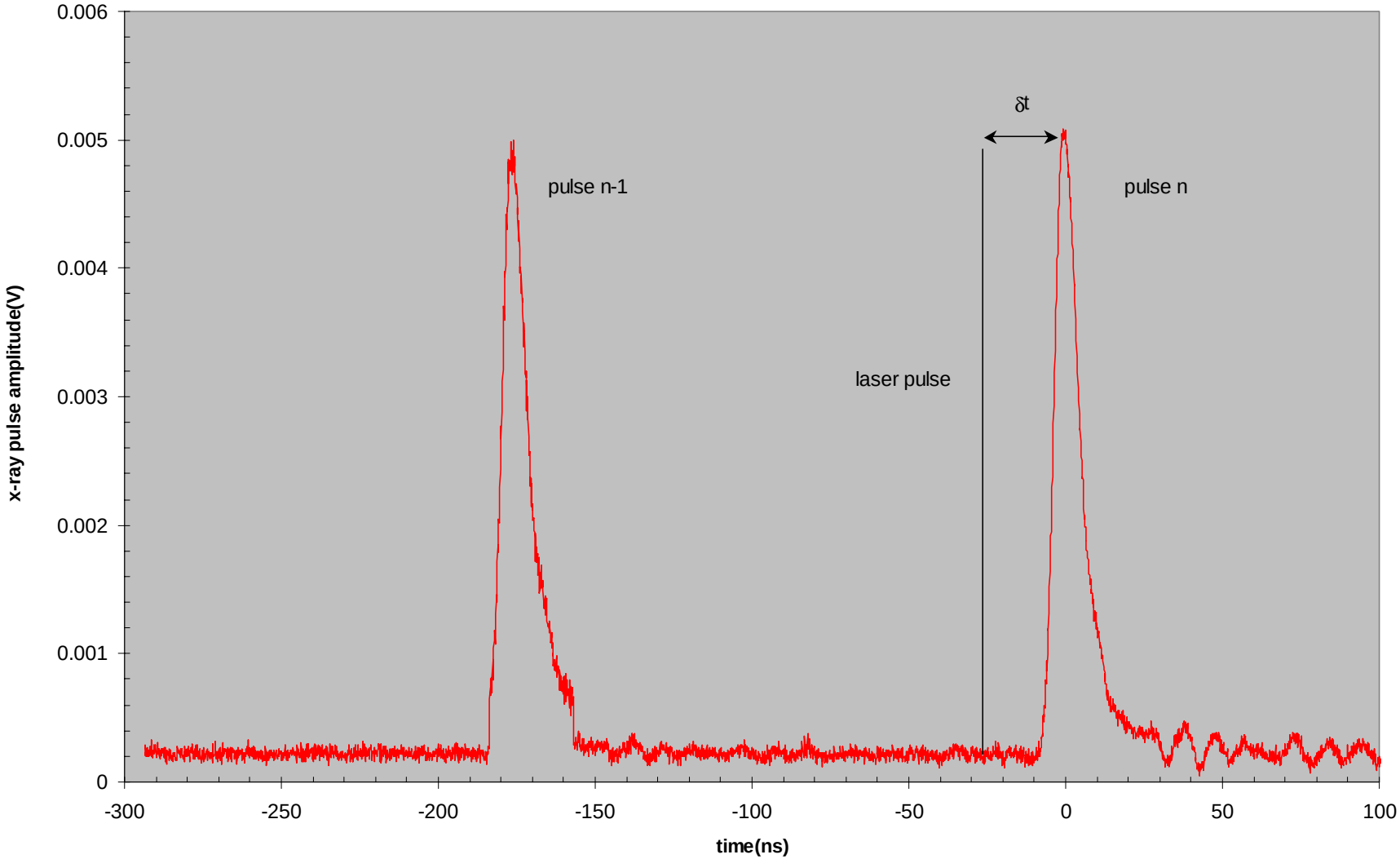




Finding time-zero with a live-signal from the sample



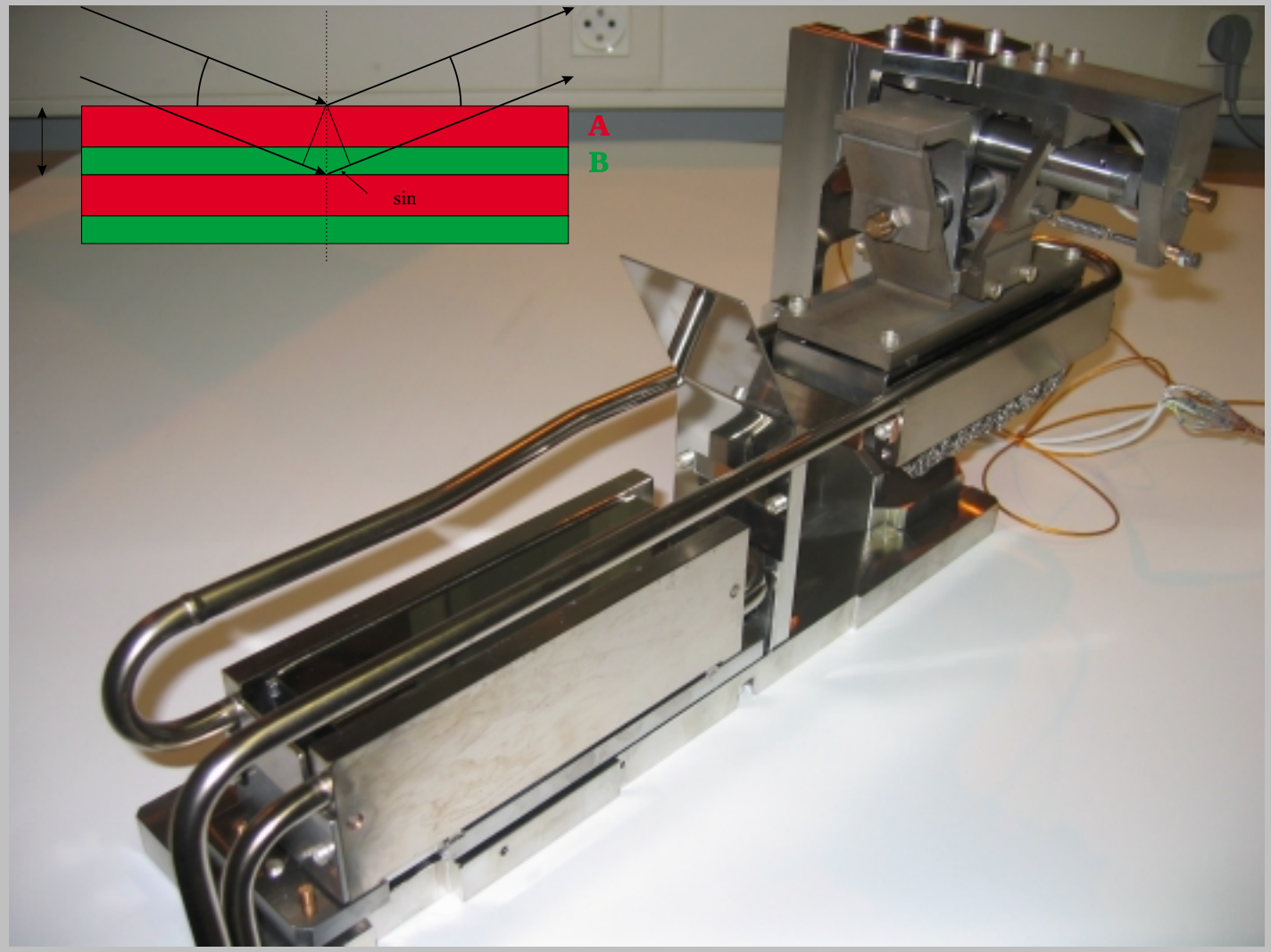
Future area detector: chopper selects two pulses in 16 bunch mode:
The non-excited and excited spectra are recorded on the same sample and (nearly) the same x-ray beam



Multilayer optics(cryogenically cooled)

(Ru / B4C)$_{51}$: d = 39.20 Å, 10-20 keV, delta E/E = 3.1%

(Ir / Al2O3)$_{100}$: d = 25.66 Å, 20-30 keV, delta E/E = 1.9%



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