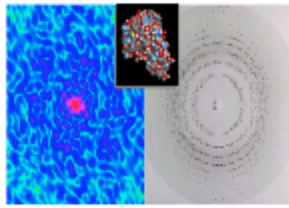
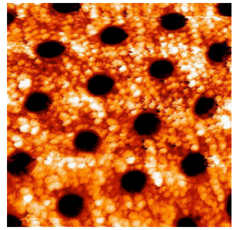
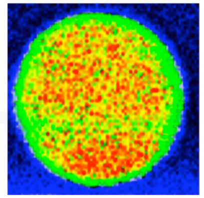
Towards single molecule shape determination with laser generated soft x-rays

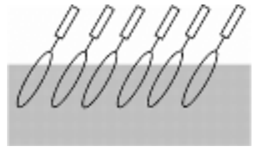
Jeremy Frey

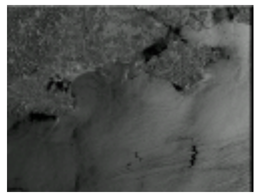
School of Chemistry, University of Southampton

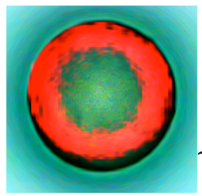












Talk

- Towards Ultra-fast x-ray single molecule studies
- Single molecule shape
- Development of a High Harmonic Source
- Initial Scattering Studies
- Future Experiments

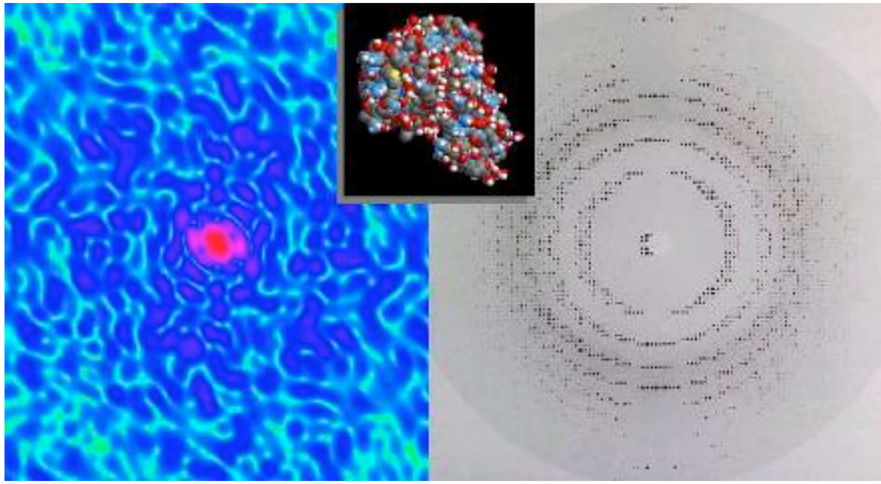
Current ways to find protein structure

- X-ray crystallography
  - Needs a crystal
  - Crystal may influence structure
- Cryo-electron microscopy
  - 2D crystal, but can be small
- NMR
  - No crystals, but size limitations

Avoiding the crystal problem….

Many groups looking at this for work with FELs.

single molecule



Crystal

Huge advantages, huge problems!

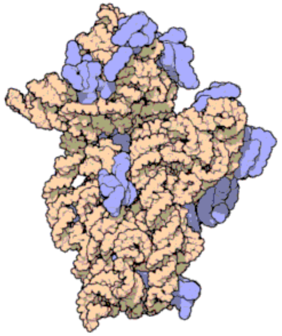
- No need to crystallize
- No averaging

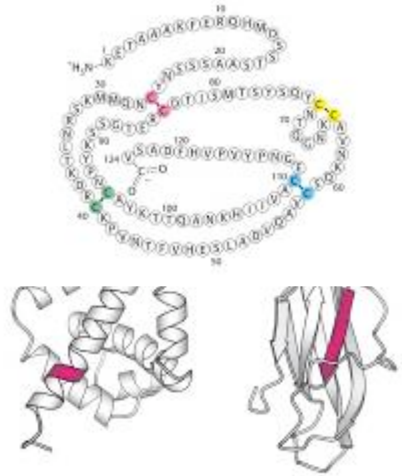
- Damage to molecule from X-rays
- Signal / noise issues

X-ray scattering

X-ray diffraction of crystals

Molecular Cluster ⟷ Shape ⟷ Structure

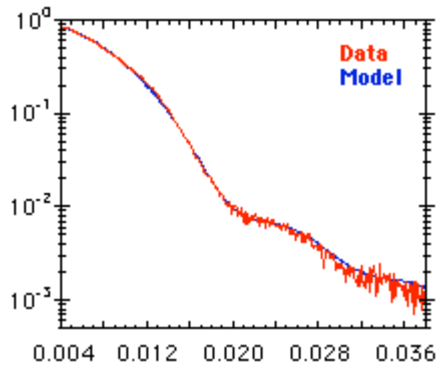




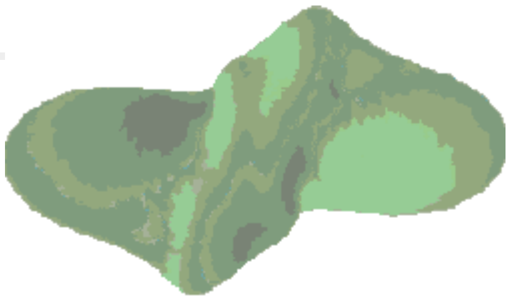
Molecular dynamics simulation

Small Angle Solution X-Ray Scattering

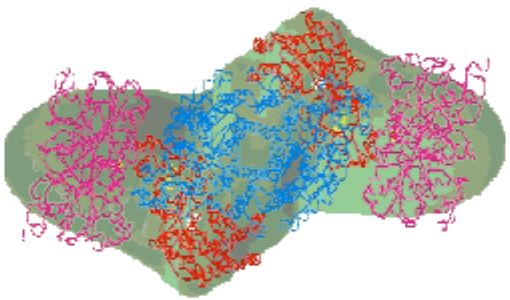
Using Molecular Shape to Characterise Protein Complexes

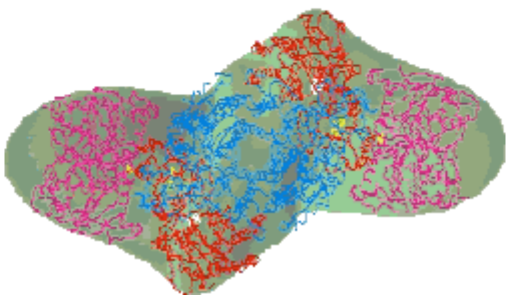


The molecular envelope of the 2:1 complex formed between Mo and Fe component proteins of nitrogenase, derived from solution x-ray scattering.



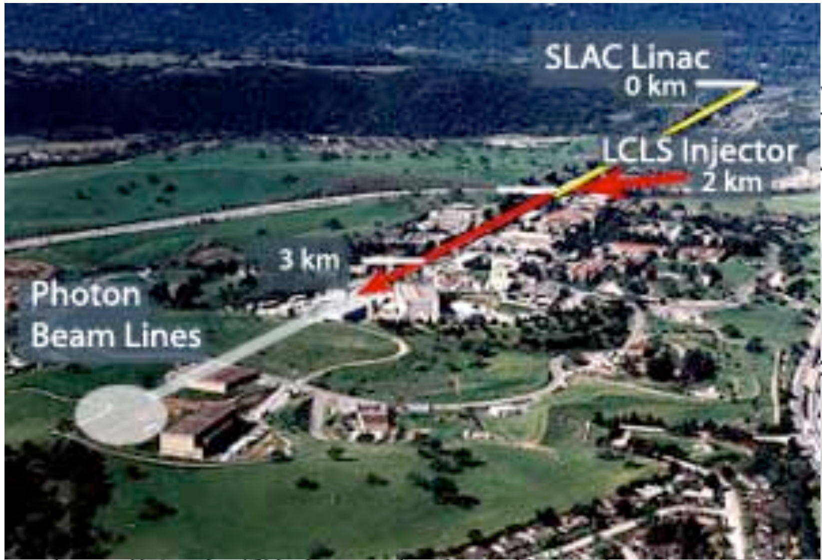
Docking Model of the complex using the crystal structures of the individual proteins: not in agreement with the molecular envelope.

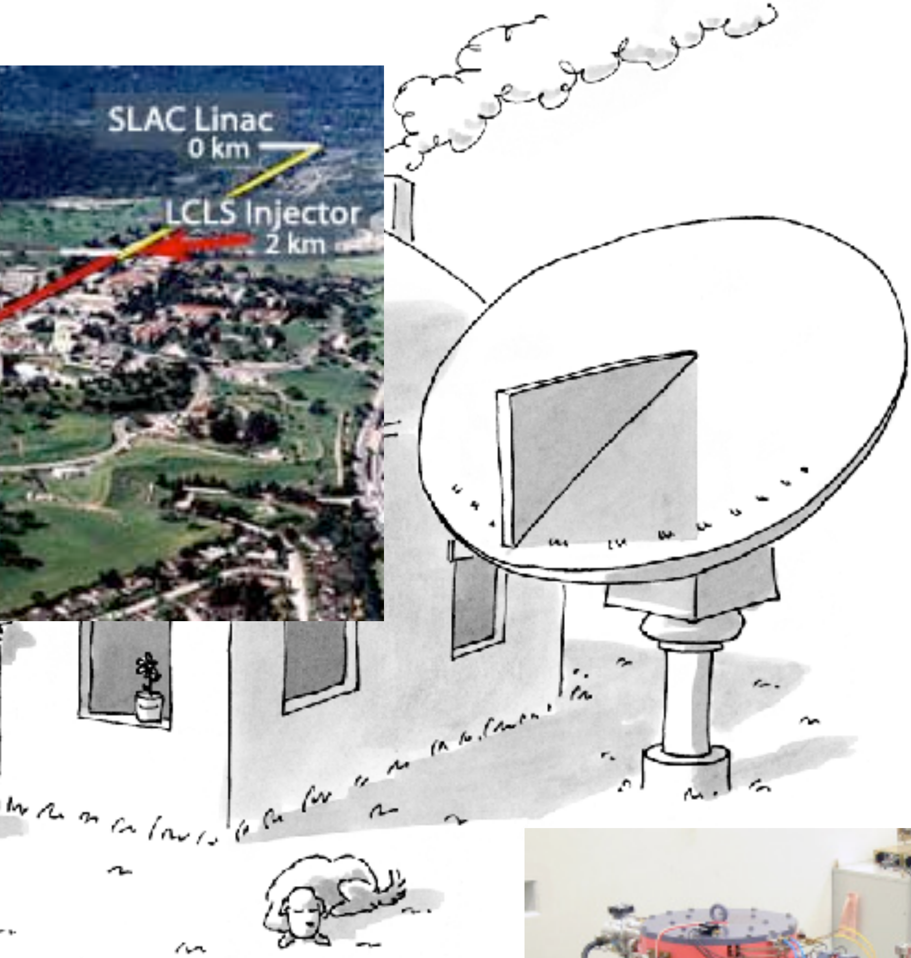




The significant structural changes predicted using solution x-ray scattering are confirmed by the subsequent crystal structure of the complex.

http://srs.dl.ac.uk/mbg/saxstech.html

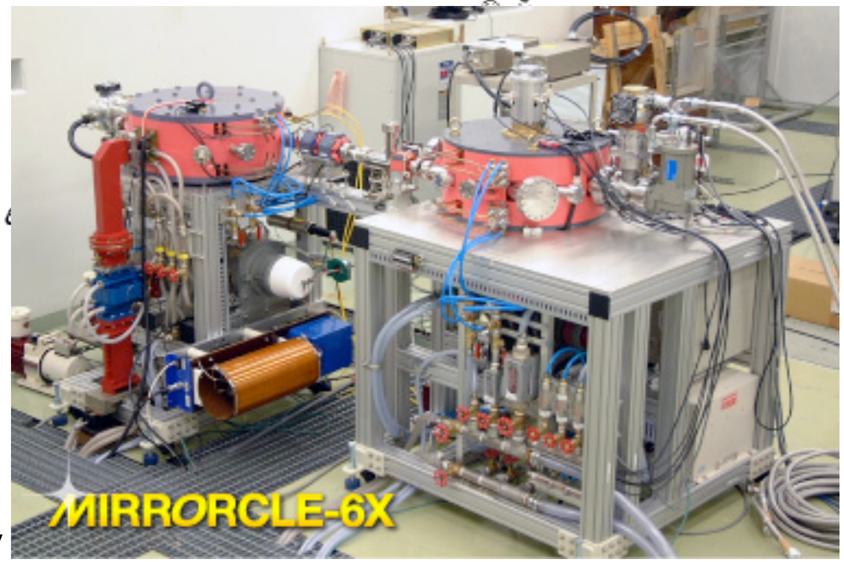




"Sure, it's an eyesore, but we get better time than anyone

But recent develop ments may change this

Synchrotron & FEL technology does not fit in the laboratory

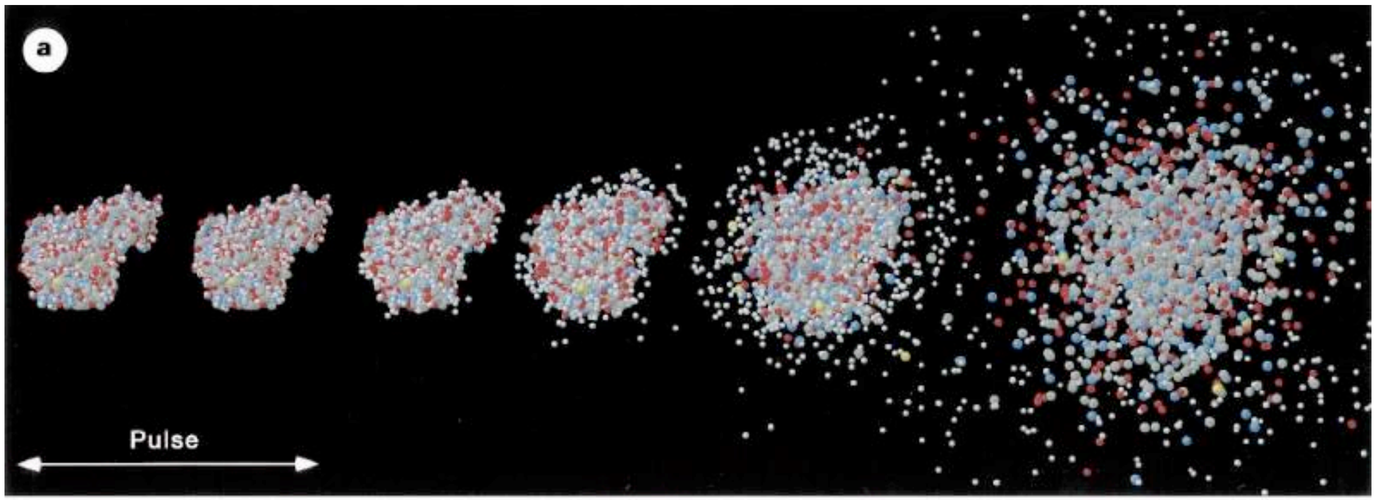


Soft x-rays

- Wavelength of a few nm
- Ideal for probe nm shape of protein complexes, biological machines
- Nanomaterials
- Soft x-rays can be manipulated quite readily
- Absorption is a problem

Damage limitation

- Conventional damage threshold: 200 ph/Å^2
  Damage process
  - inelastic scattering of electrons out of molecule
  - Coulomb repulsion of remainder
- Timescale? fs

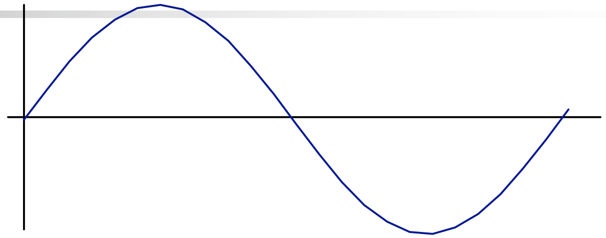


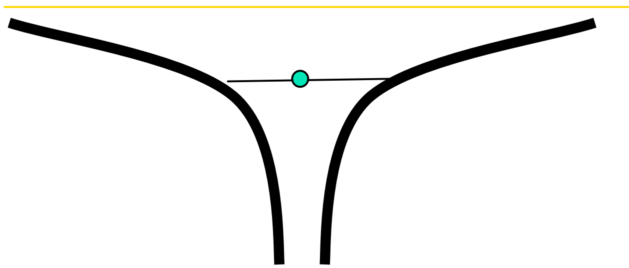
t=–2fs *t*=2fs *t*=5fs *t*=10fs *t*=–20fs *t*=50fs

Neutze *et al.*, Nature (2000) **406**, 752

HHG - the simple model

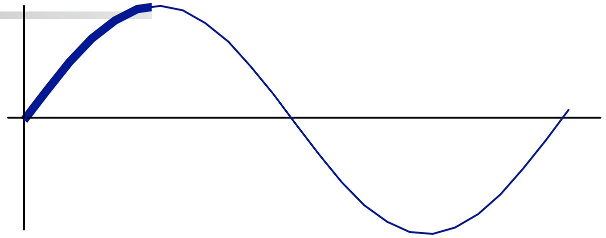
- Electron tunnels out of atom as field increases
- Electron accelerates in laser field as free particle
- Electrons which come back to the atom can recombine and emit an energetic photon

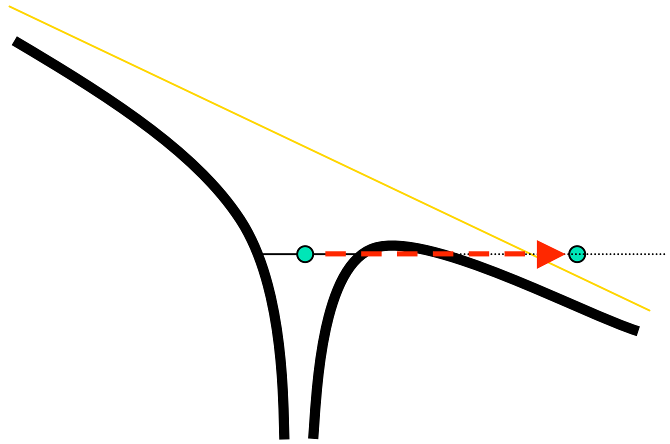




HHG - the simple model

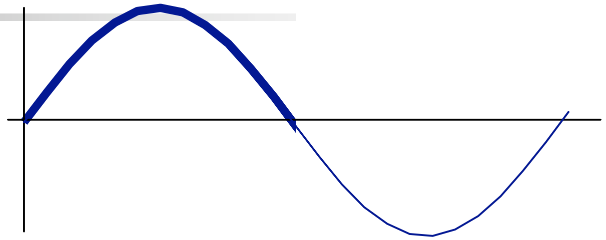
- Electron tunnels out of atom as field increases
- Electron accelerates in laser field as free particle
- Electrons which come back to the atom can recombine and emit an energetic photon

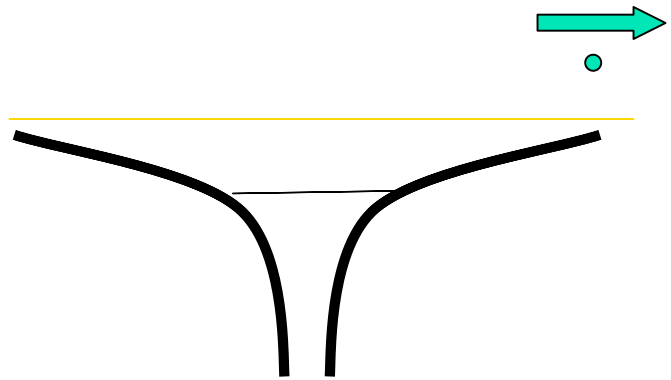




HHG - the simple model

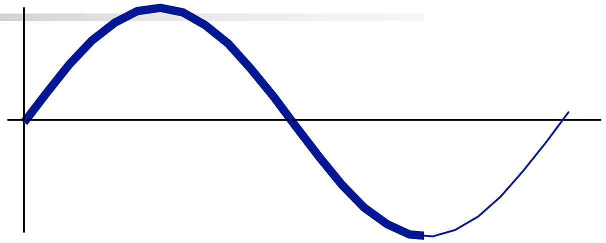
- Electron tunnels out of atom as field increases
- Electron accelerates in laser field as free particle
- Electrons which come back to the atom can recombine and emit an energetic photon

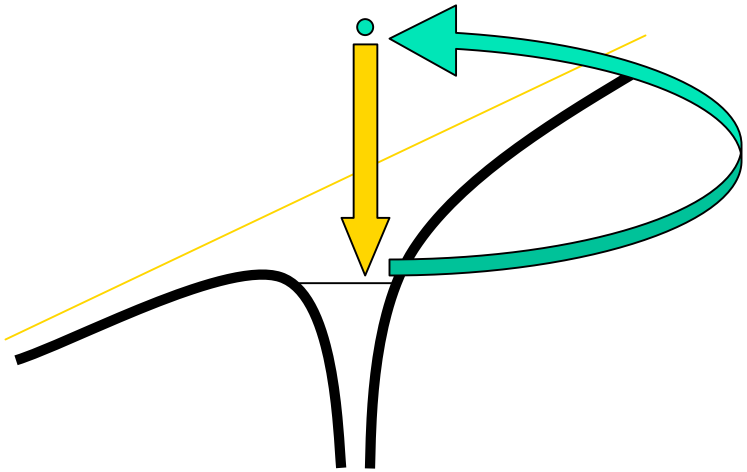




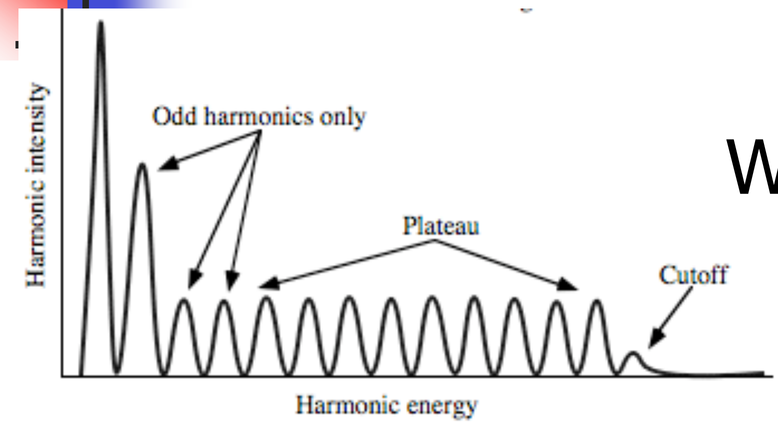
HHG - the simple model

- Electron tunnels out of atom as field increases
- Electron accelerates in laser field as free particle
- Electrons which come back to the atom can recombine and emit an energetic photon



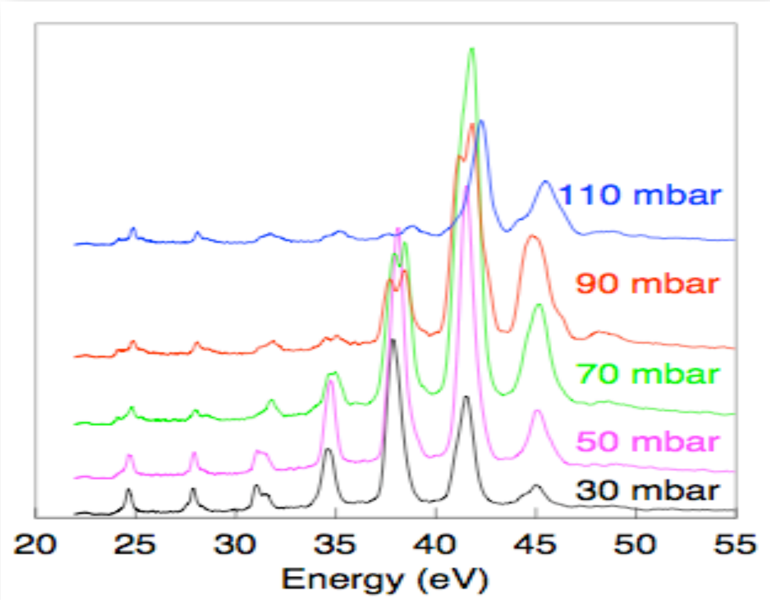


$$E = h\nu = I_p + 3U_p$$

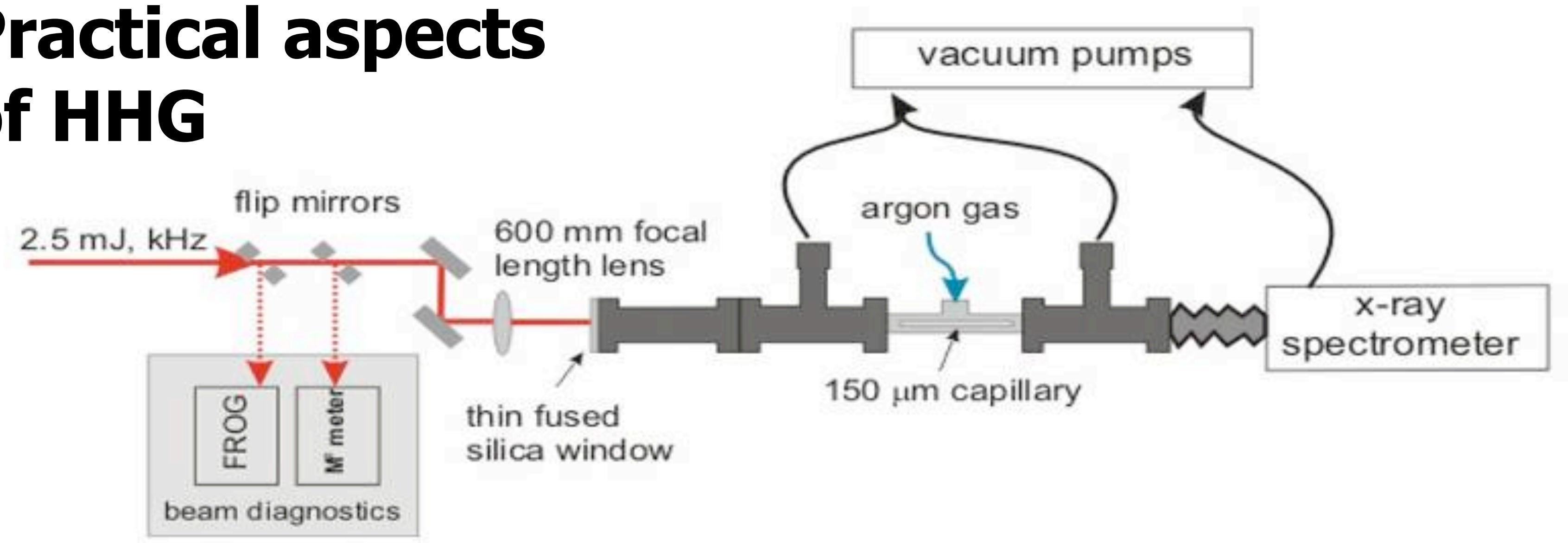


From"Ultrafast Dynamics of Quantum Systems: Physical processes and spectroscopic techniques". NATO ASI International School of atomic and molecular spectroscopy. Ed. B. di Bartolo, pp611-623 (plenum press, NY, 1999)

- Observed spectrum mainly due to Phase Matching
- The theoretical cutoff is not reached
- With modified capillaries we can reach in to the water window



Practical aspects of HHG

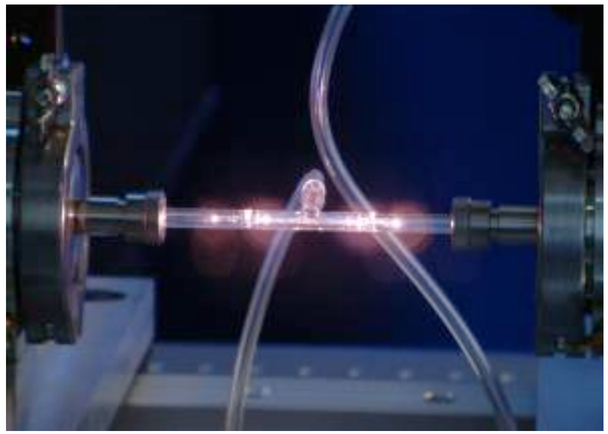


•Capillary tube holds *Ar* gas at low pressure (also Ne, N_2 and N_2O)

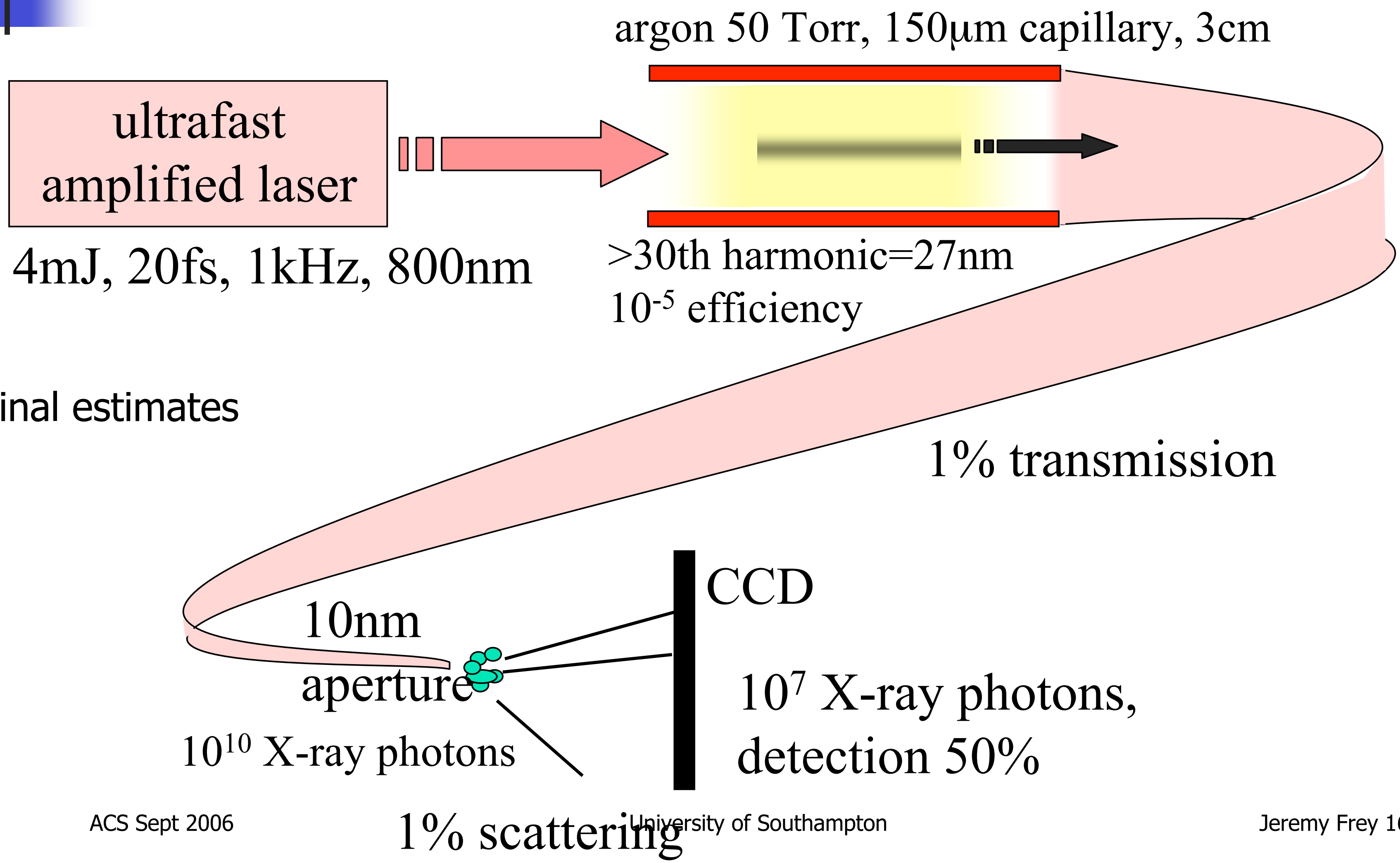
•Need **very high** laser powers, Peak intensity ~10^{15} W/cm^2 & Peak E-field ~100 GV/m

•Laser focused into capillary, guided along bore

•X-rays generated as a coherent beam along capillary



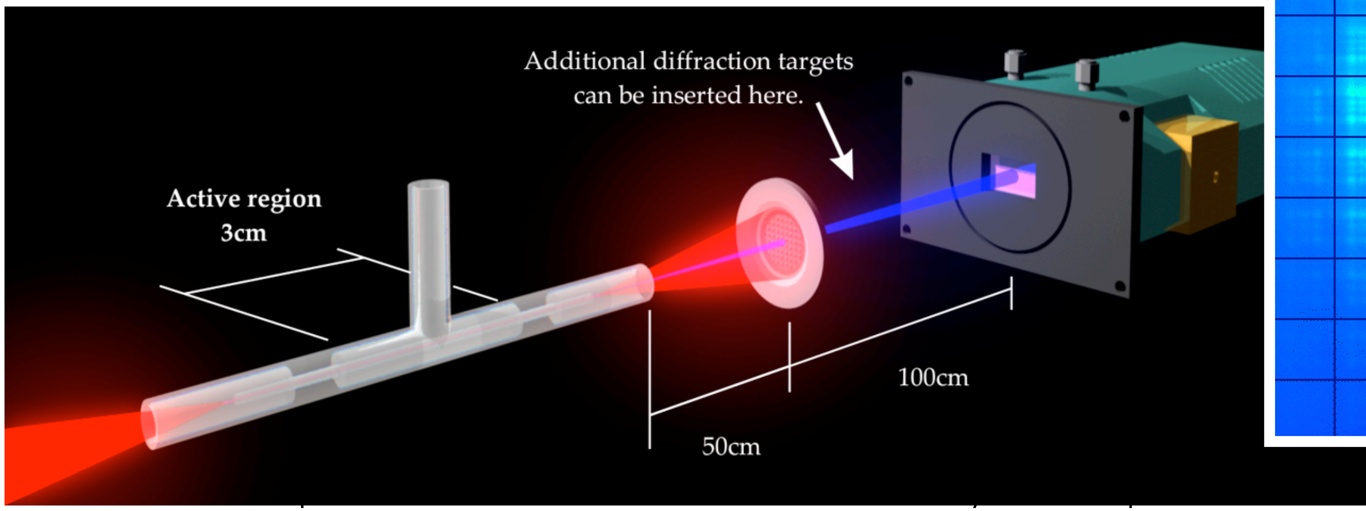
Xray throughput for high harmonic generation

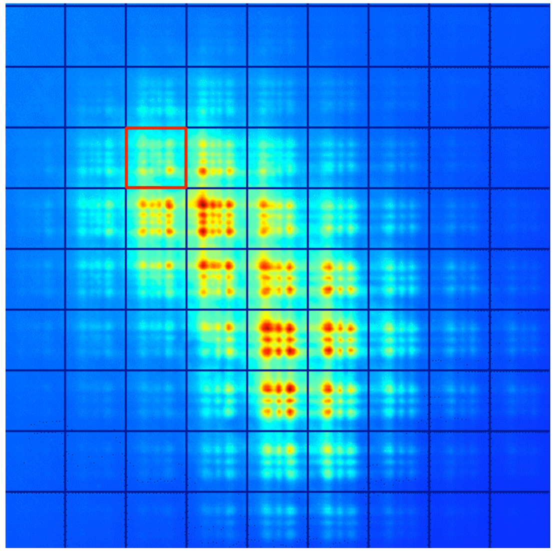


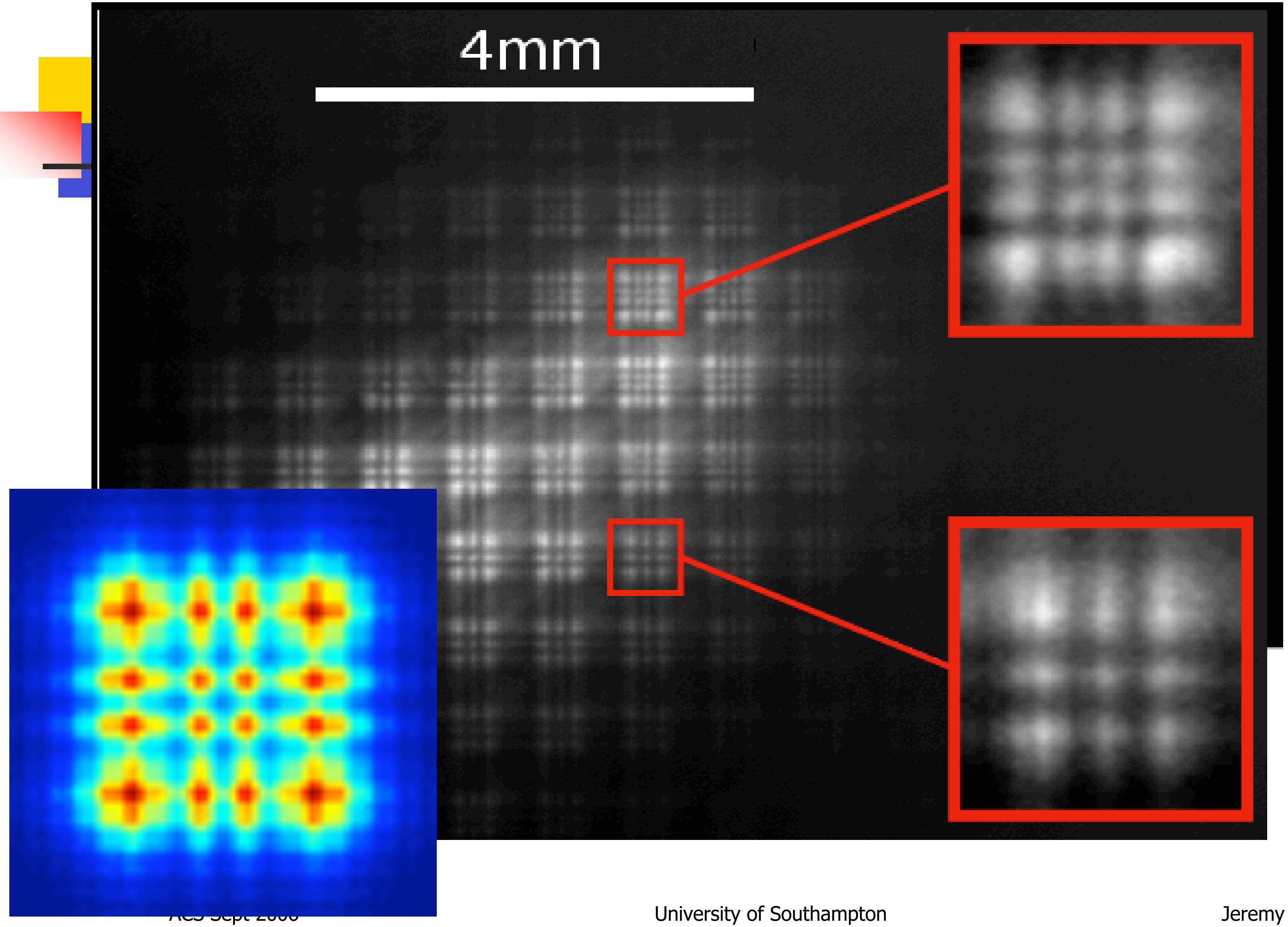
Original estimates

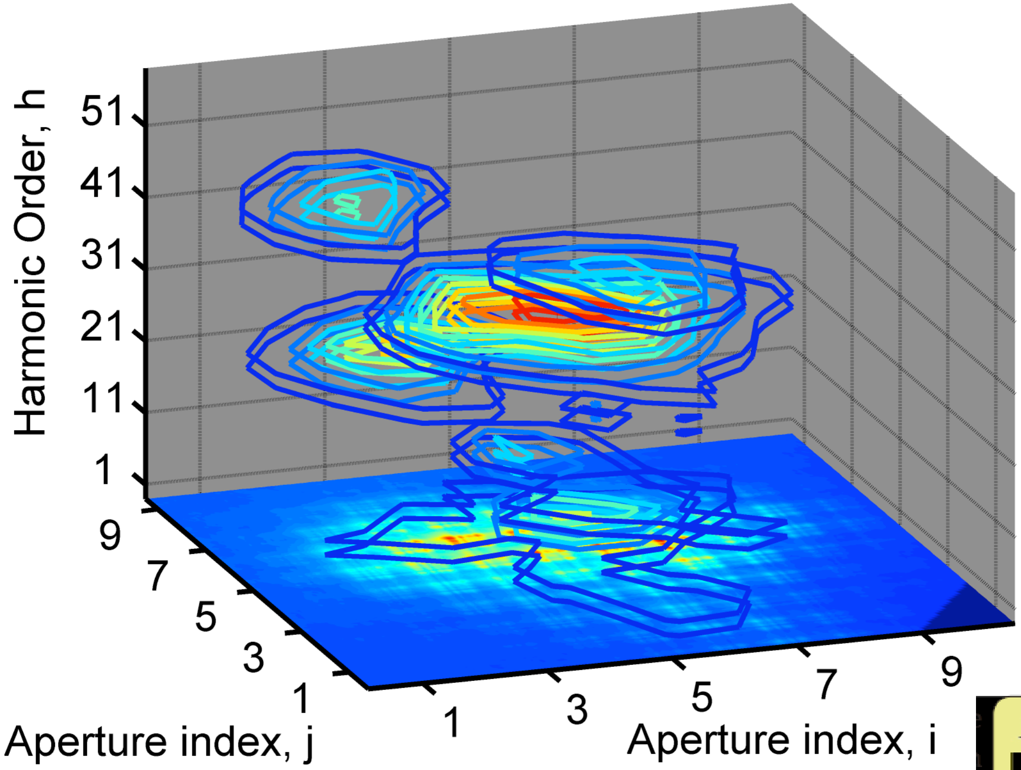
Square Grid Diffraction

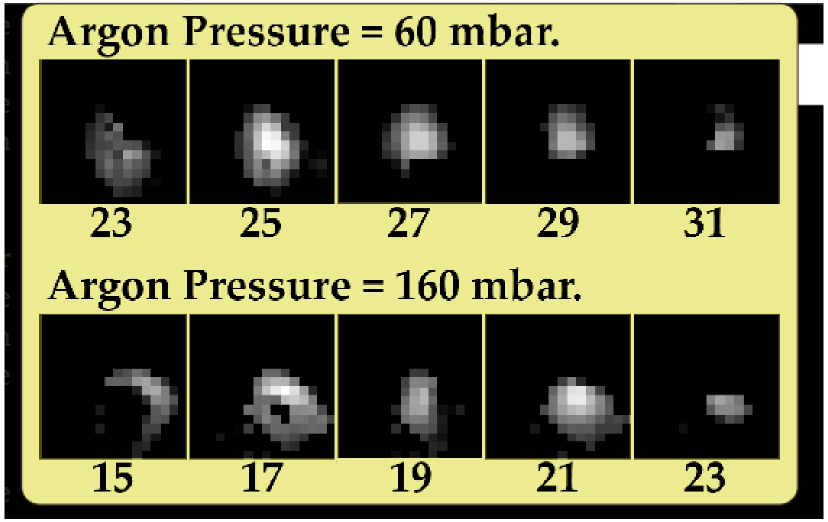
- Record x-ray diffraction by filter support grid.
- 340μm square apertures, 18μm bars.
- 50cm source to grid, 100cm grid to camera.
- Observe spatial variation of diffraction patterns.
- Can we extract x-ray spectrum?





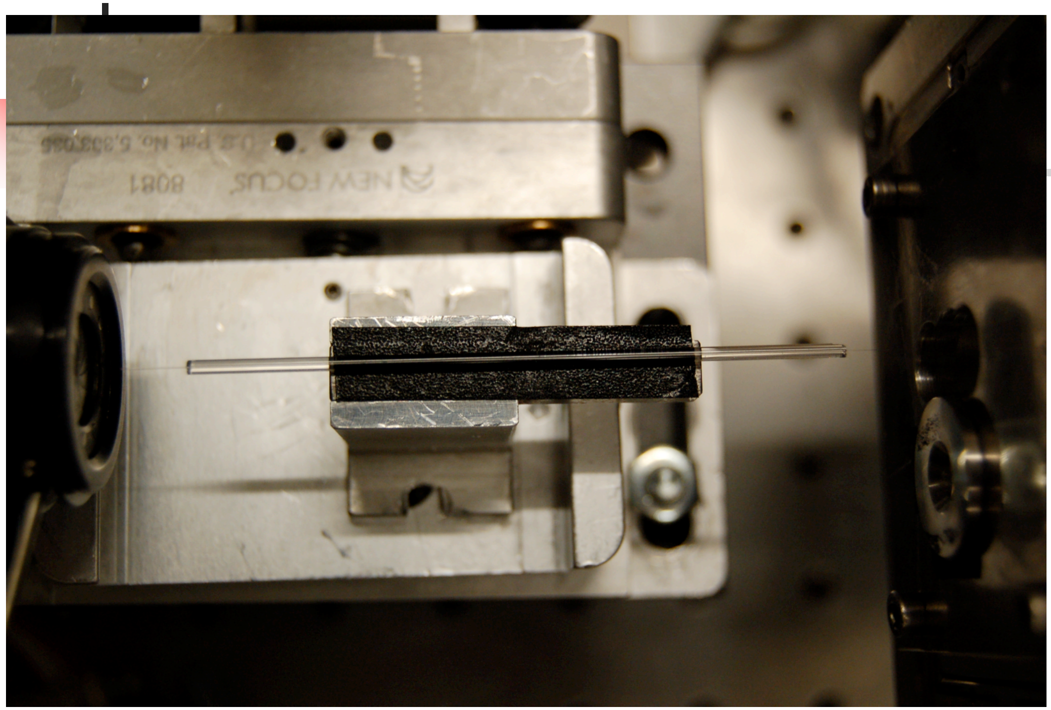

4mm



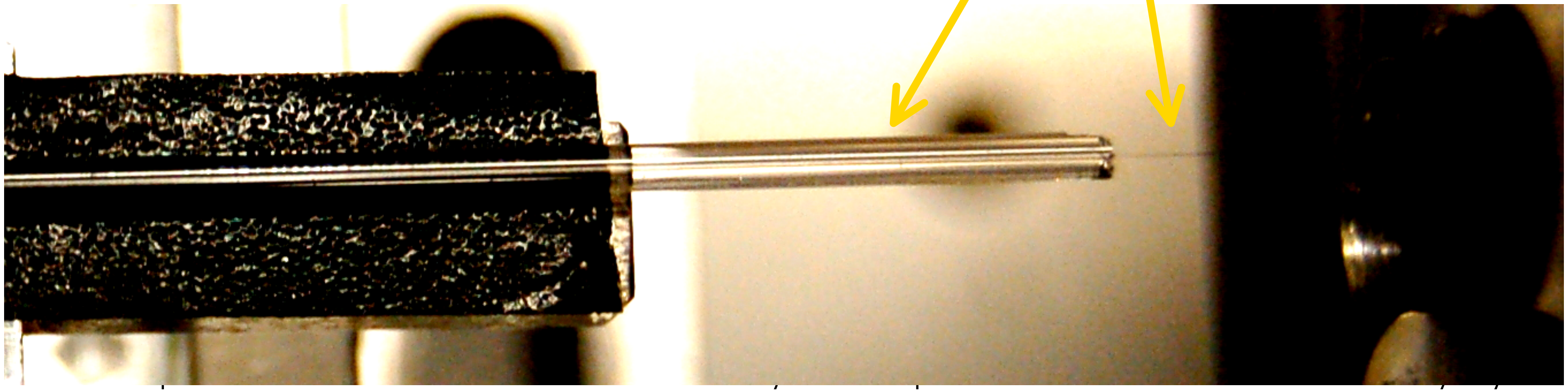


Characterize and to control the wavelength and spatial profile of the x-ray beam

Taper mounted inside a holding capillary

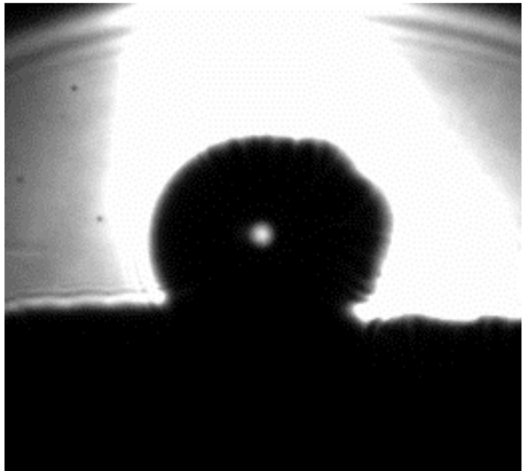


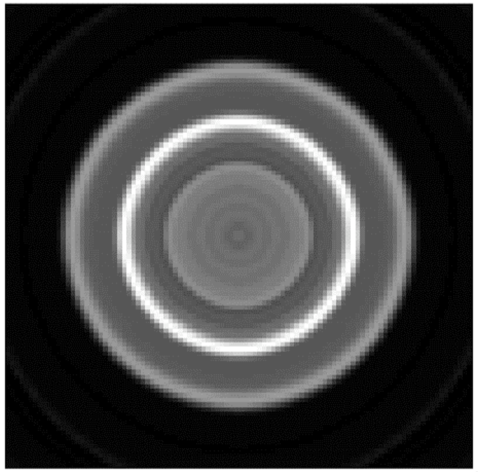
50 μm

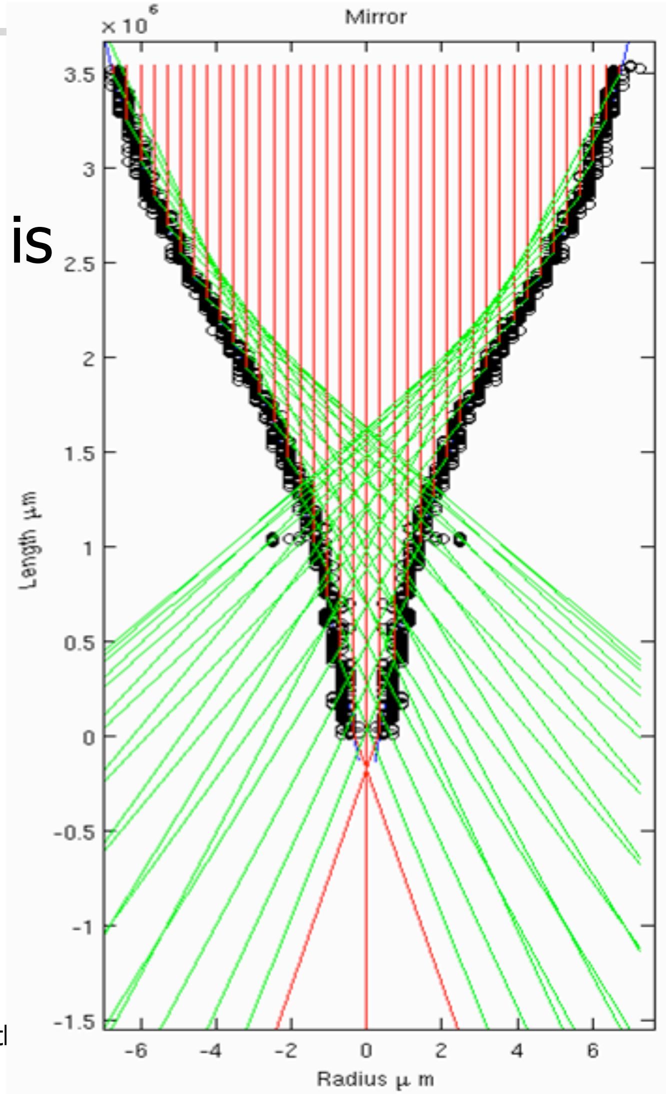


Tapered Capillary Focussing

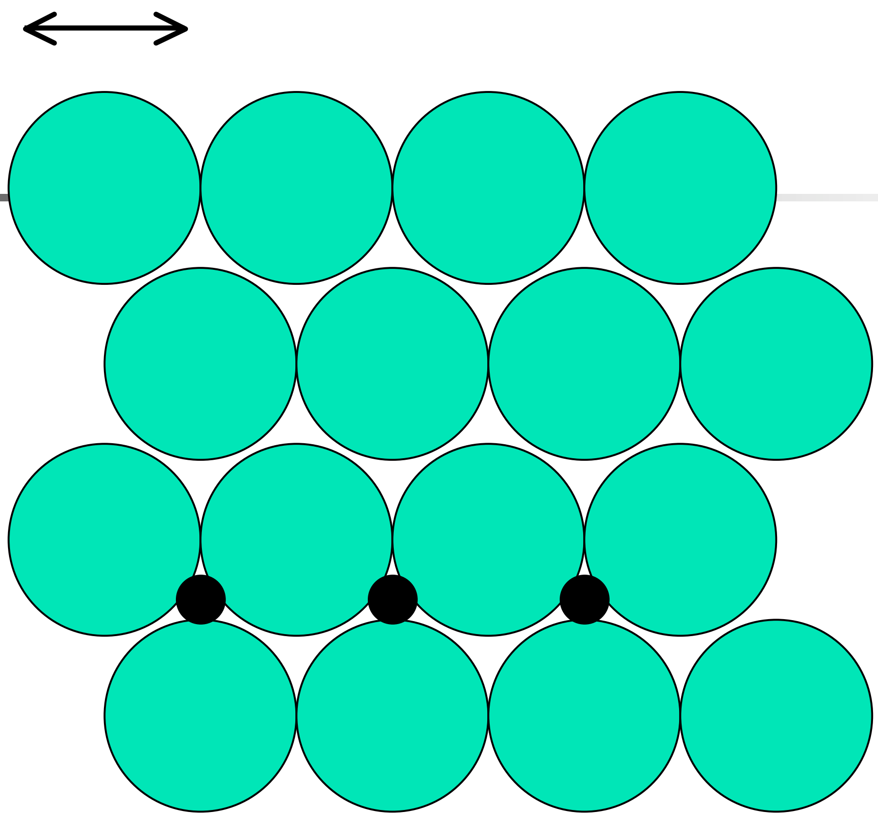
- Grazing incidence reflection.
- Parabolic or elliptical profile.
- Not a true focus but x-ray beam is intensified by a factor of
- Study individual objects or small part of larger objects.







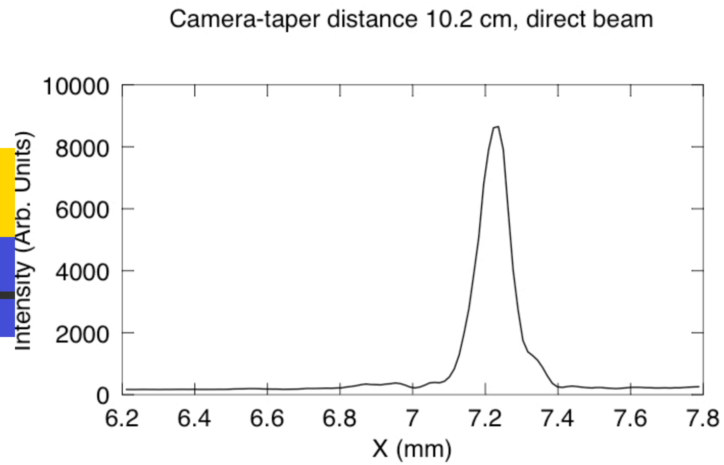
200 nm

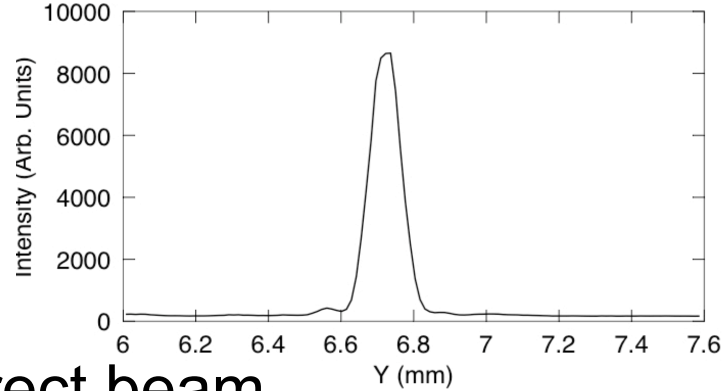


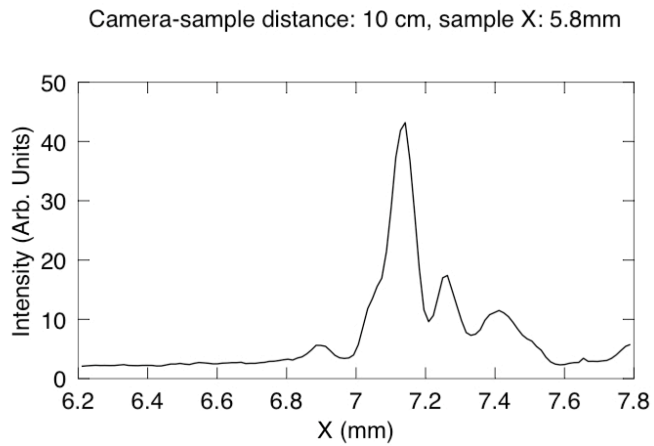
2D lattice of
200 nm latex
spheres

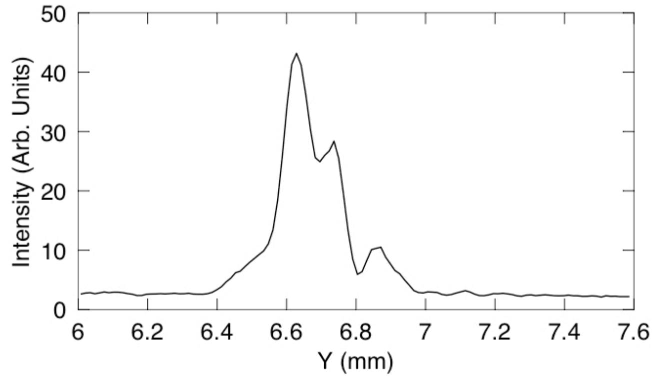
It is the gaps that
transmit the x-rays

Scattering close to the direct beam

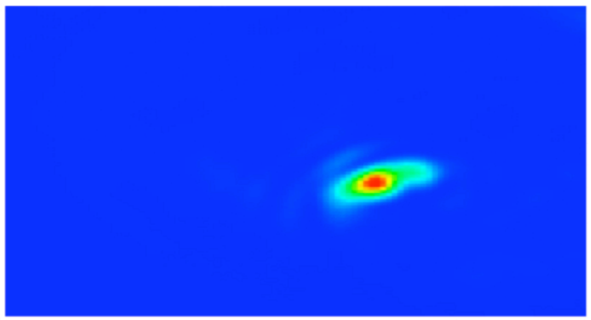


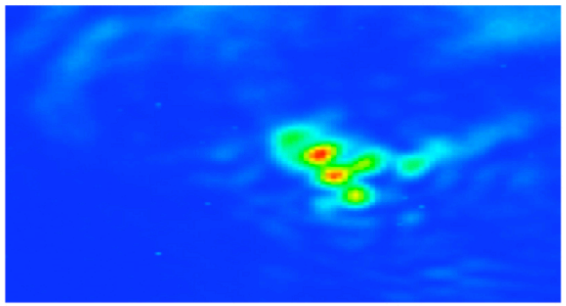




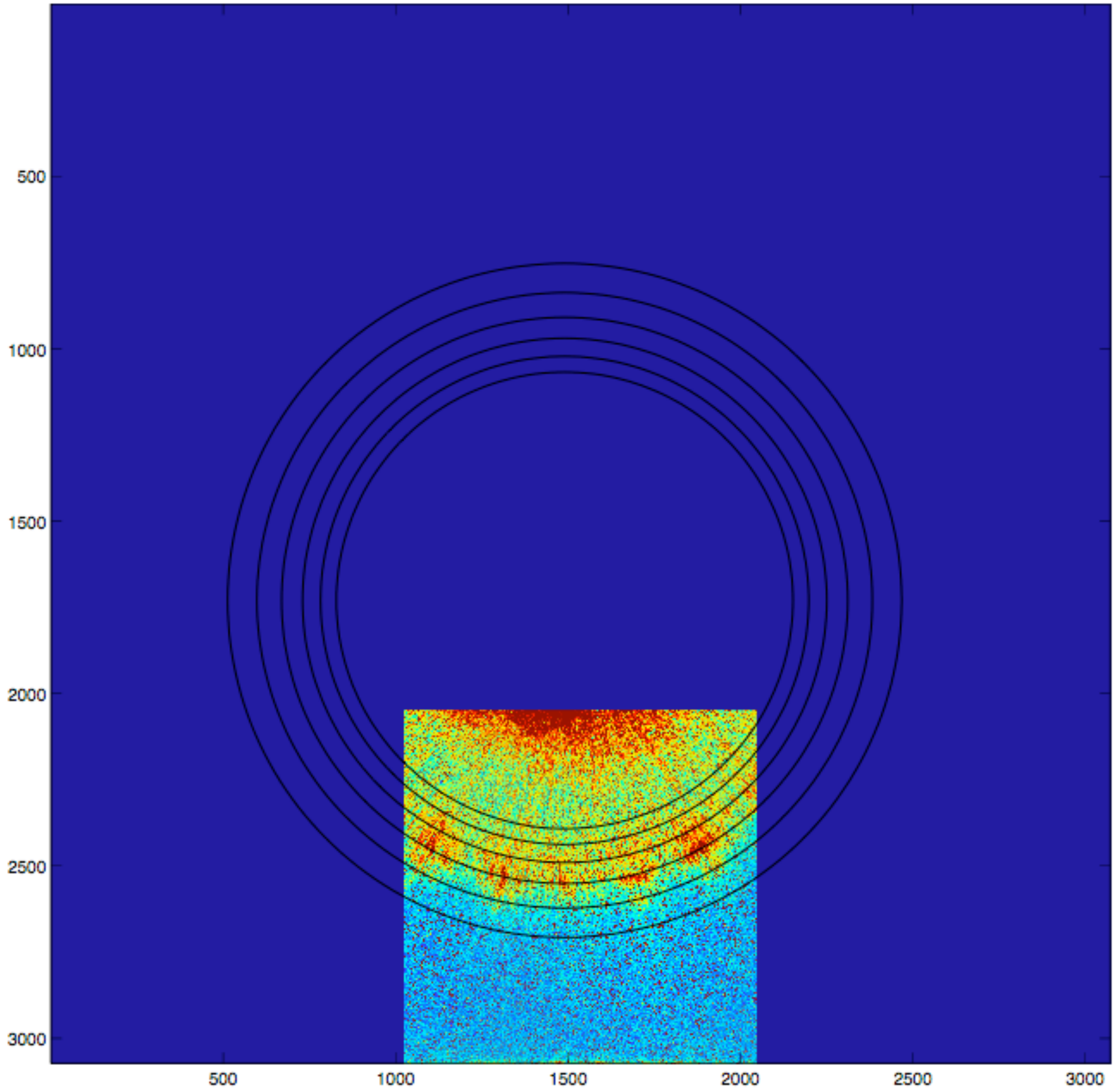


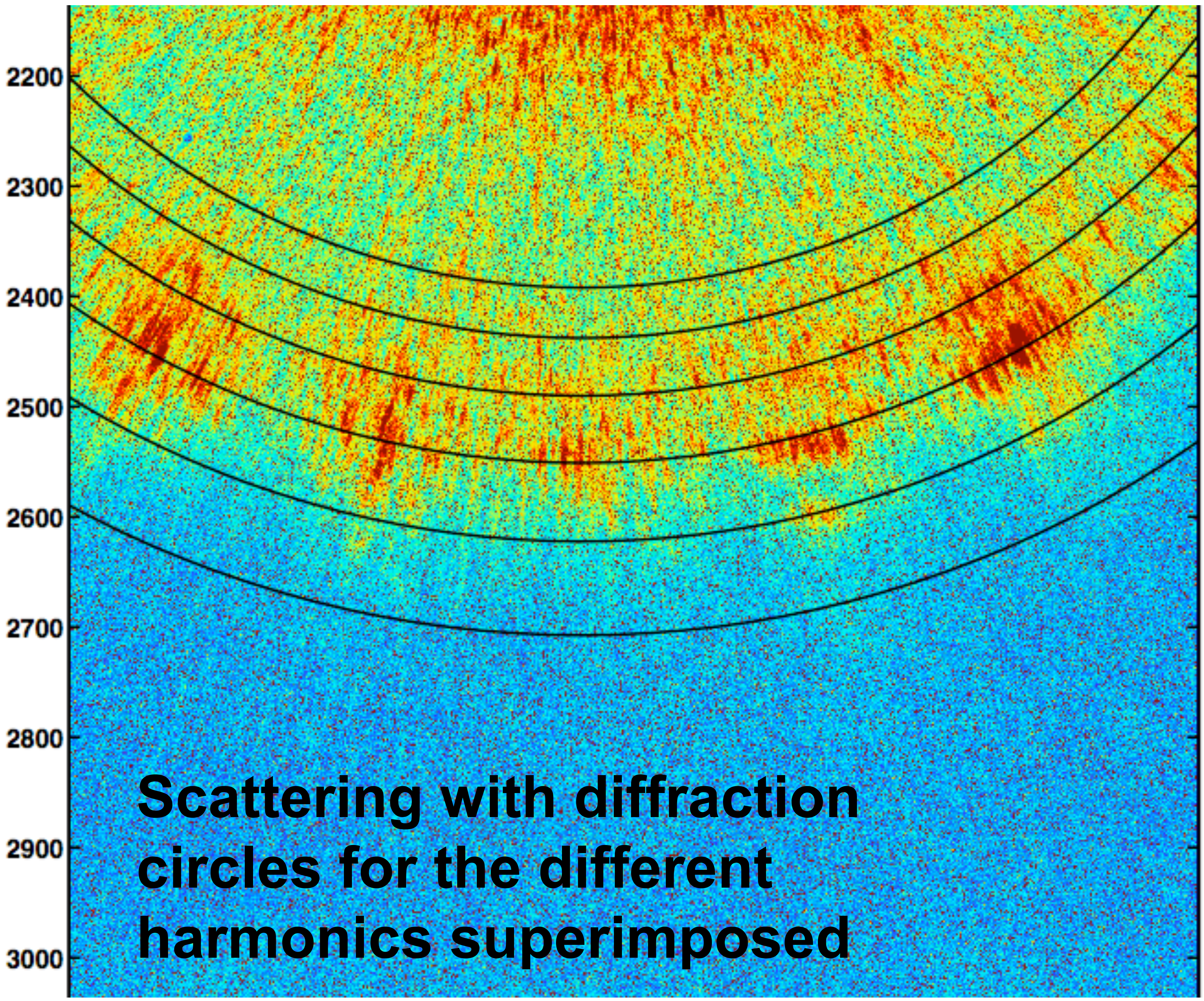
Direct beam





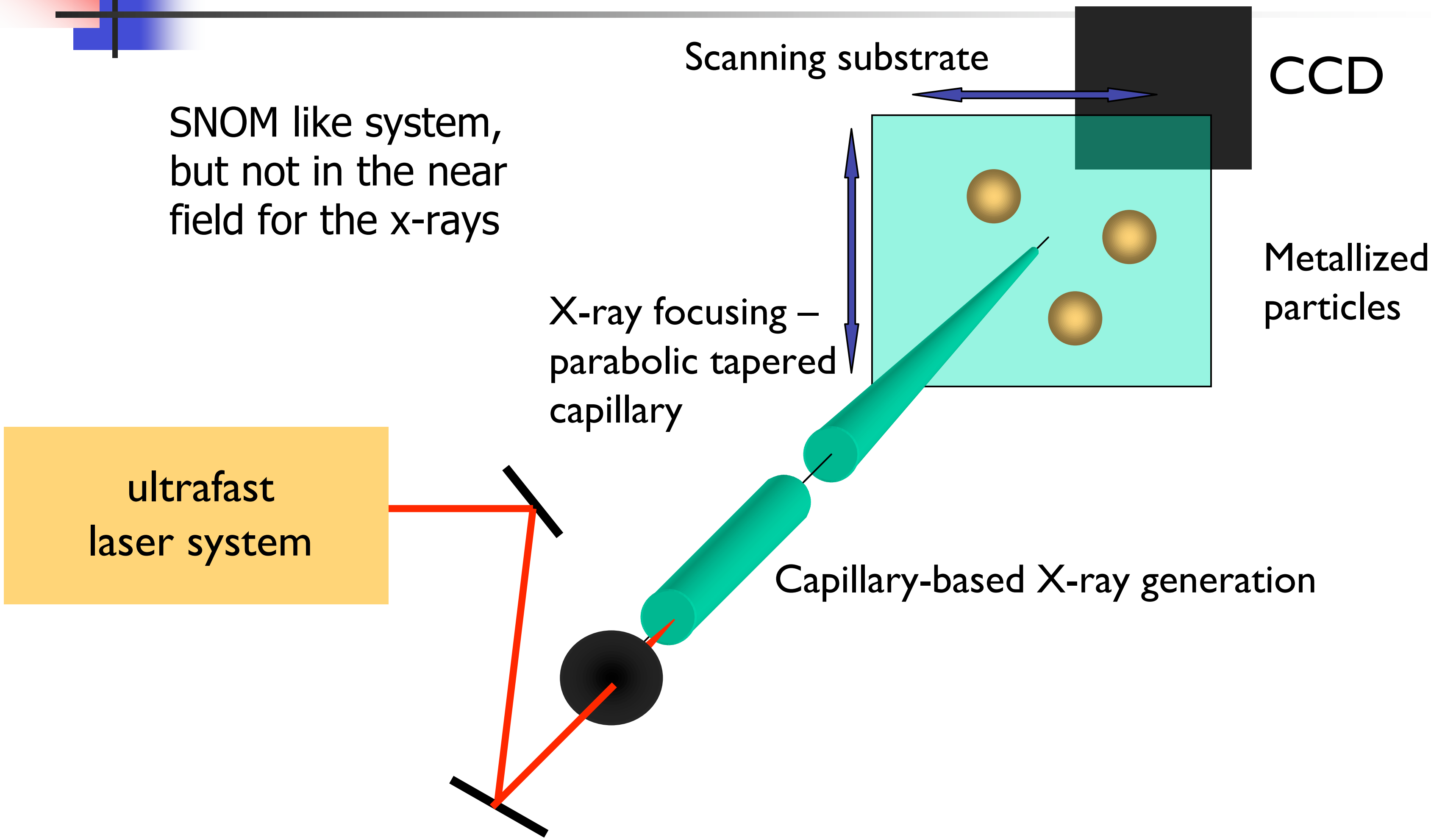
Scattering image showing the location of the CCD image



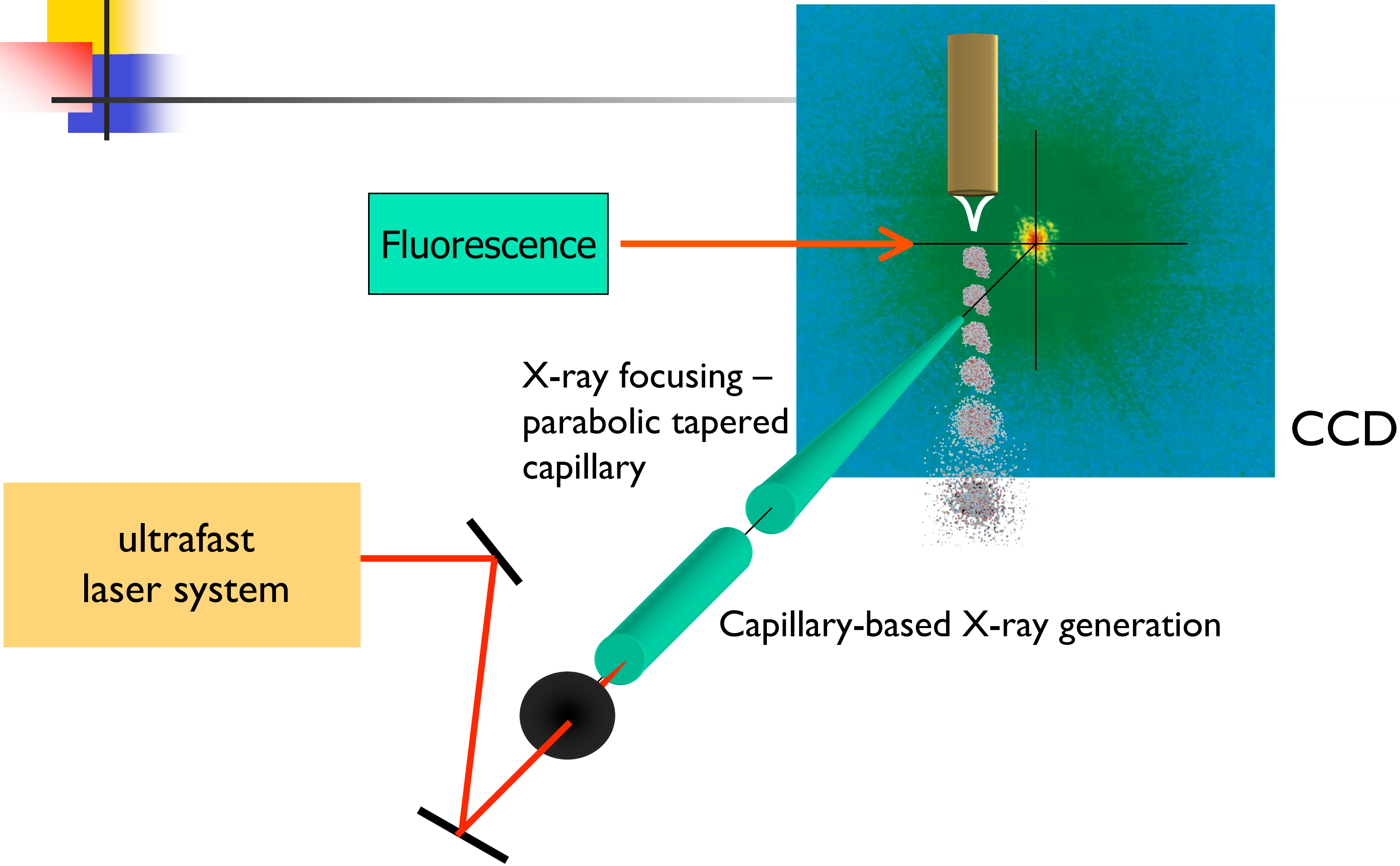

2200
2300
2400
2500
2600
2700
2800
2900
3000
Scattering with diffraction circles for the different harmonics superimposed

- Scanning X-ray nanoprobe

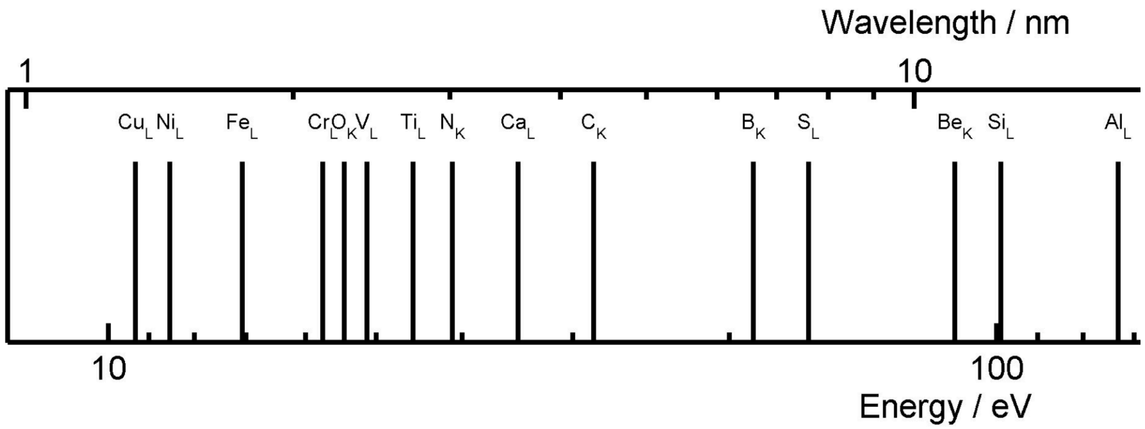
SNOM like system, but not in the near field for the x-rays



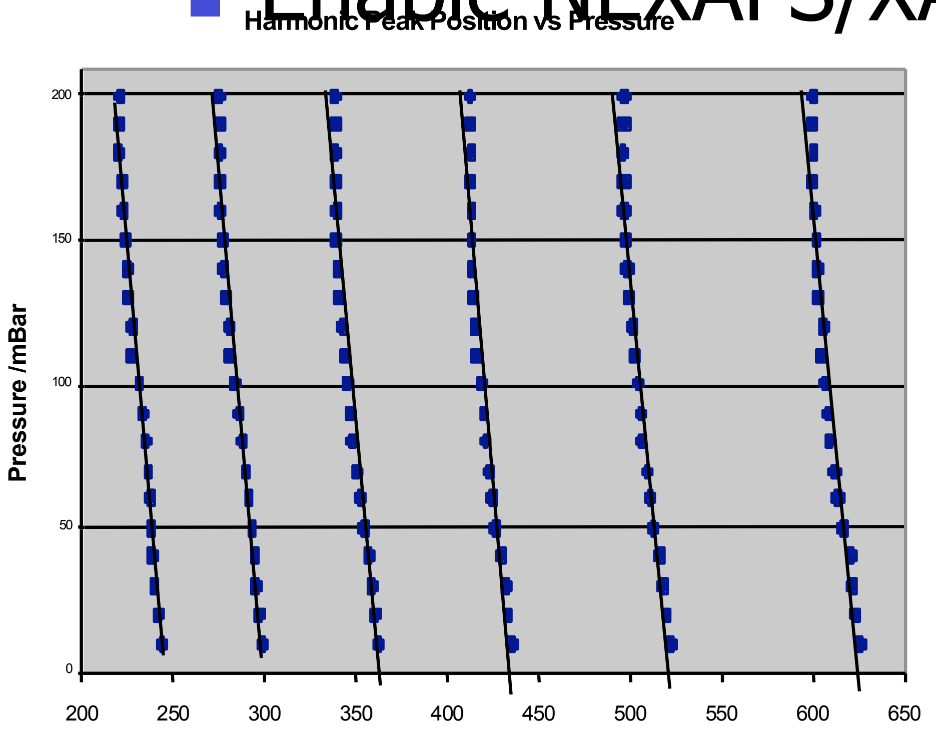
Electrospray or laser desorption



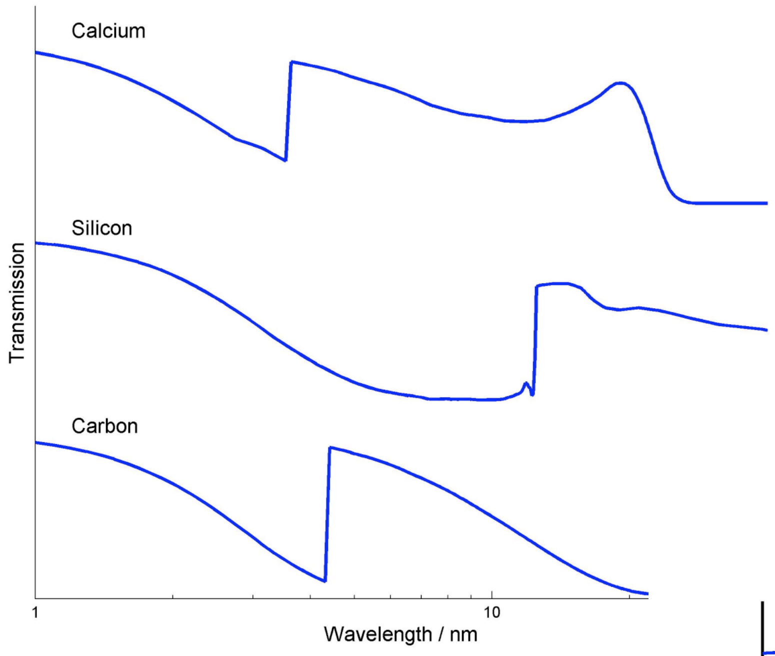
Tuning



- Enable NEXAFS/XANES

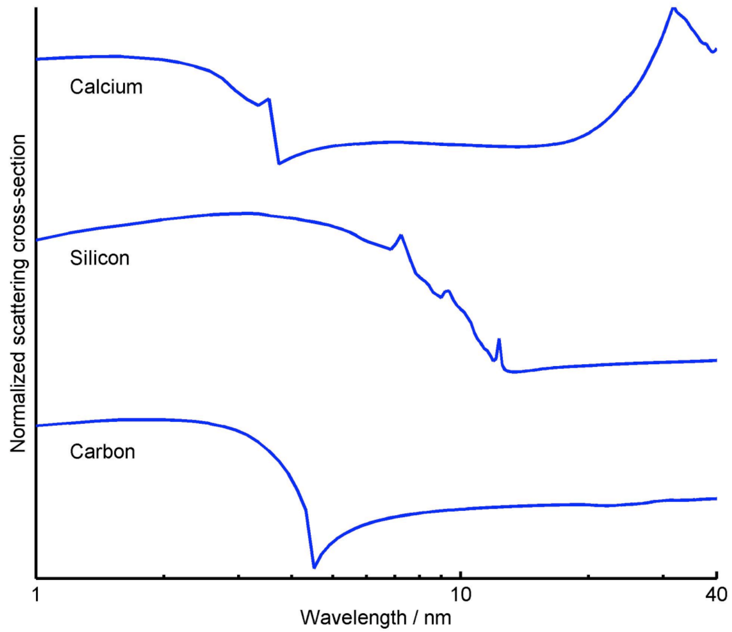


Pressure tuning, & tune by using the Dazzler to change the fundamental dispersion altering



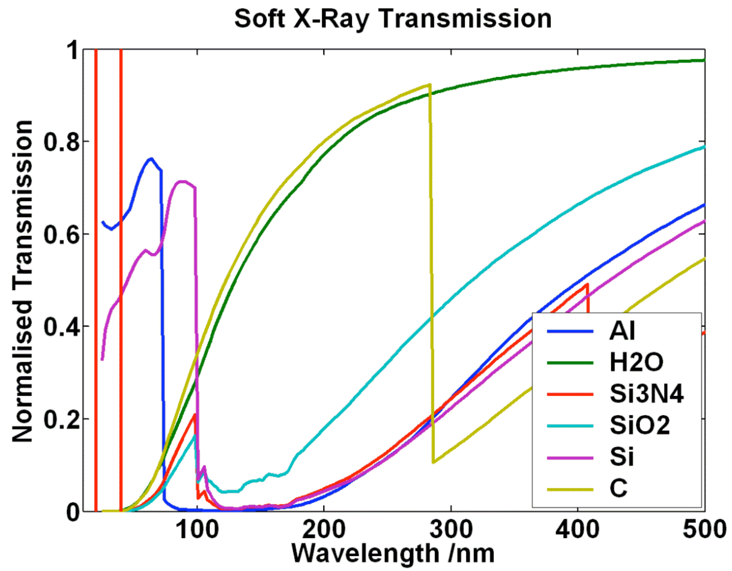
Absorption

Scattering



Non-linear x-ray spectroscopy

- Focussed fs beam of attosecond pulse train
- Very high peak power
- Non-linear spectroscopy and microscopy should be possible
- Extend the range of resonances that can be probed using our wavelength range

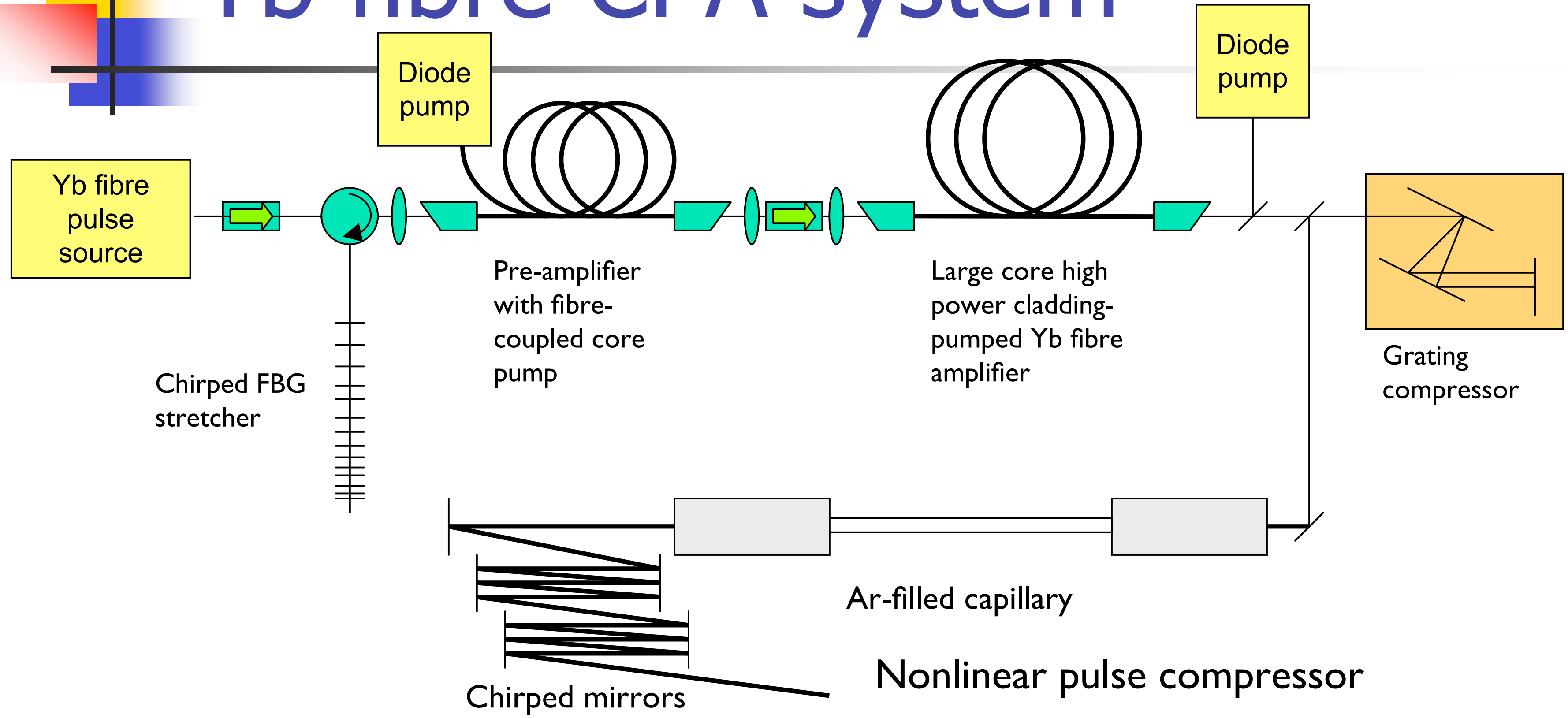


Data from: www-cxro.lbl.gov

Detectors

- Need highly efficient detector
- Xray photon counting
- Single pulse
- 3D detector
- Use conventional and innovative designs
  - Nanoscale lithography?

Yb fibre CPA system



Ti:sapphire replacement to provide a more compact source requiring less maintenance

On track for 30fs, 0.1mJ/pulse, with high average power (100's Watts)

HHG vs Other Sources

- Tabletop source
- Short pulse duration
- Well defined time structure
- Good coherence
- Relatively inexpensive
- BUT
- Lower energy
- Lower flux

Nanoscale X-Ray Basic Technology Group














School of Chemistry
School of Physics
ORC
CCLRC-RAL

Credits:

Funding

Basic Technology Programme (Research Councils UK)

People

Chris Froud, Matthew Praeger, Edward Rogers, Ana de Paula, Jonathan Price, Jeremy Frey, Bill Brocklesby, Dave Hanna, Jeremy Baumberg, David Richardson, John Evans, Mike Hursthouse, Graeme Hirst, Gareth Derbyshire John Dyke

Places

School of Chemistry

School of Physics & Astronomy

Optoelectronics Research Centre

CCLRC RAL

University of Southampton