Towards Soft X-ray Scanning Microscopy Using Tapered Capillaries & Laser-Based High-Harmonic Sources:

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Light

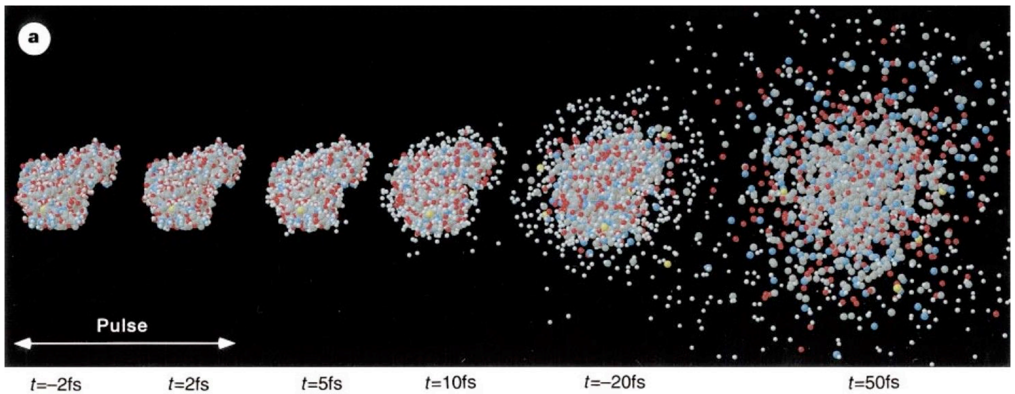
University of Southampton

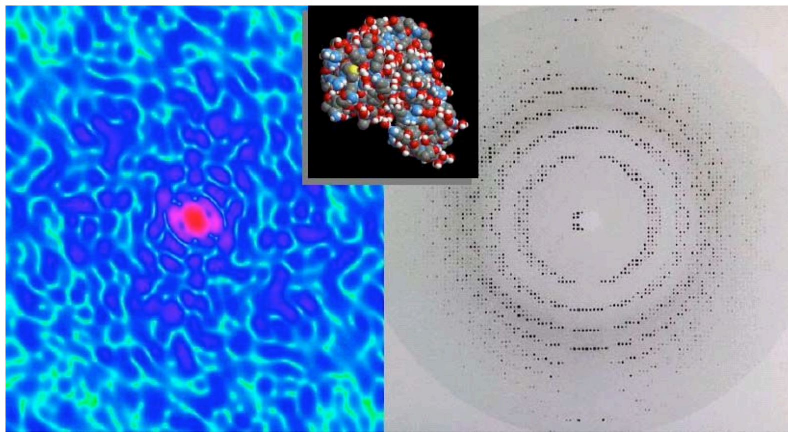
Outline

- Motivations
- Light sources for ultrafast X-rays
- High harmonic generation of soft X-rays
- Properties of high harmonic sources
- Tapers for focusing
- Future developments

Motivation: scattering from nanostructures

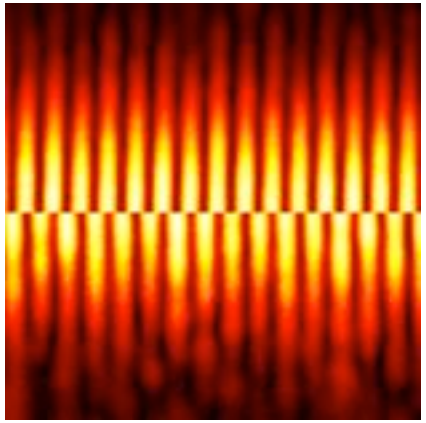
- EUV/soft X-ray nanoprobe
  - – Form of probe ideal for SPM
- Ultimate nanostructure: single protein molecule
  - – Need ultrafast (< 10fs) pulses to overcome damage issues

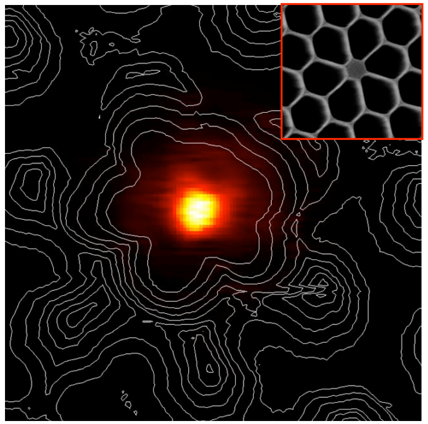


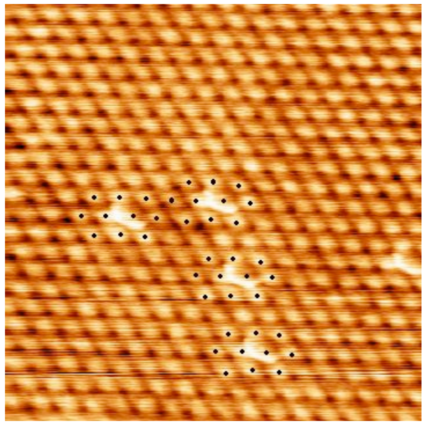


Group research: NSOM

- Interferometric IR (1.5μm) NSOM of telecomms devices
  - Fibre Bragg gratings
  - Photonic crystal fibres
- Femtosecond NSOM of nonlinear optical devices
- IR laser/STM studies of molecules on surfaces
  - 3-5μm excitation of SAMs with
  - simultaneous STM imaging and spectroscopy





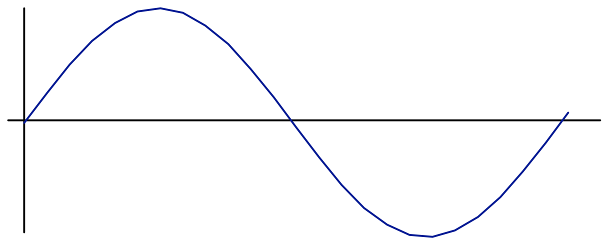


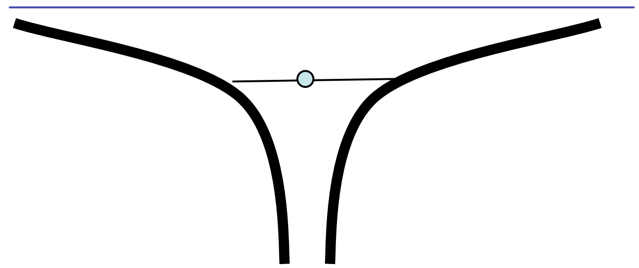
Why High Harmonic Generation?

- femtosecond X-ray sources:
  - Upcoming FEL sources (LCLS, TESLA, 4GLS)
    - High flux ✔
    - Tunability, hard x-rays ✔
    - Time structure may be too long ✖
    - Impatience (?) ✖
  - Laser-produced plasma
    - Hard X-rays ✔
    - slow ✖
    - omnidirectional ✖
  - High Harmonic Generation (HHG)
    - Good time structure ✔
    - Source availability ✔
    - Beam quality ✔
    - Compatible with fibre sources ✔
    - Low flux ✖
    - Long wavelength ✖

HHG - simple 3 step 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





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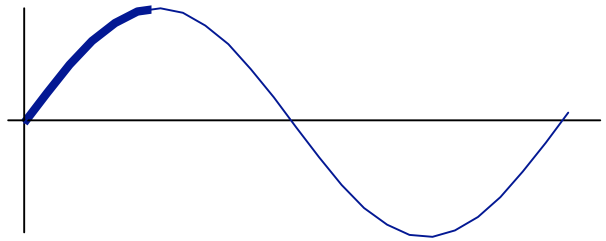

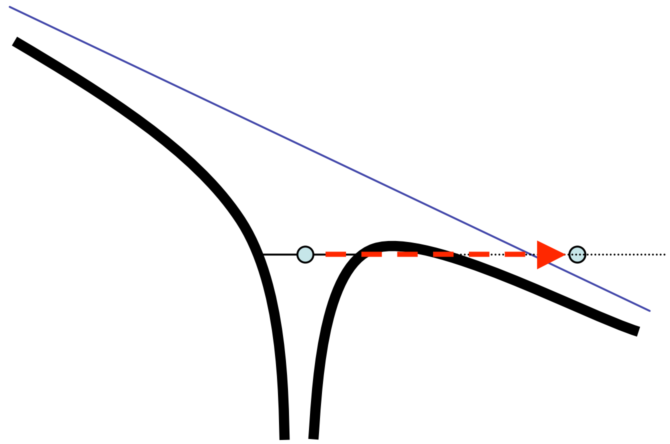

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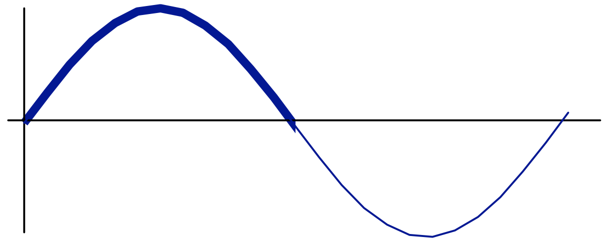

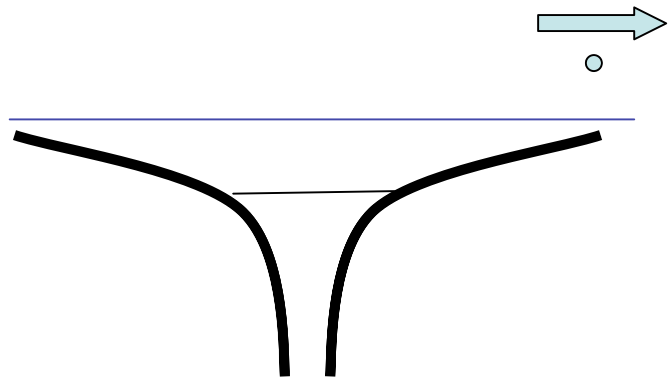

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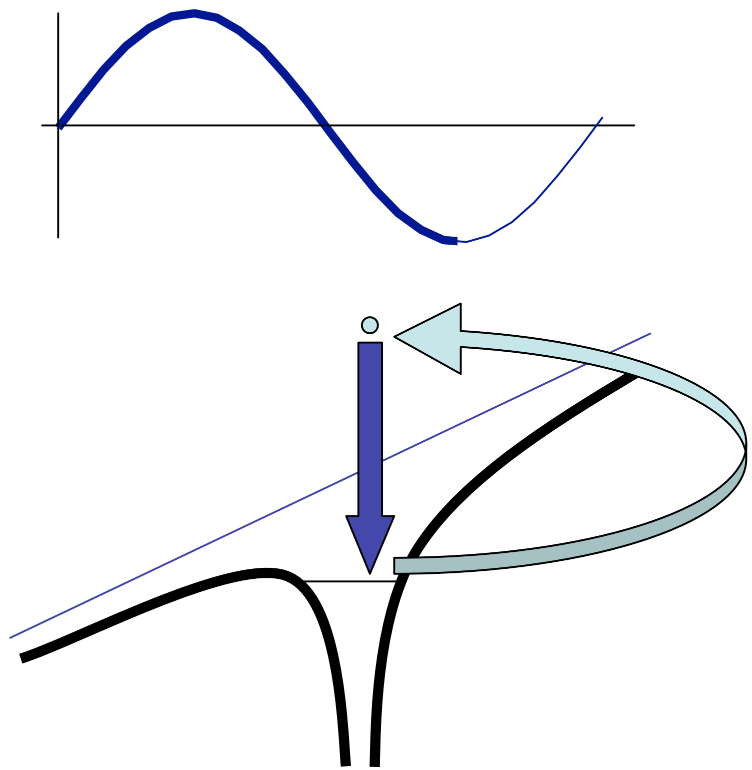
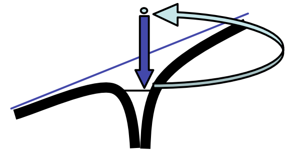


$$E = h\nu \approx I_p + 3U_p$$

Motion of electron after ionization



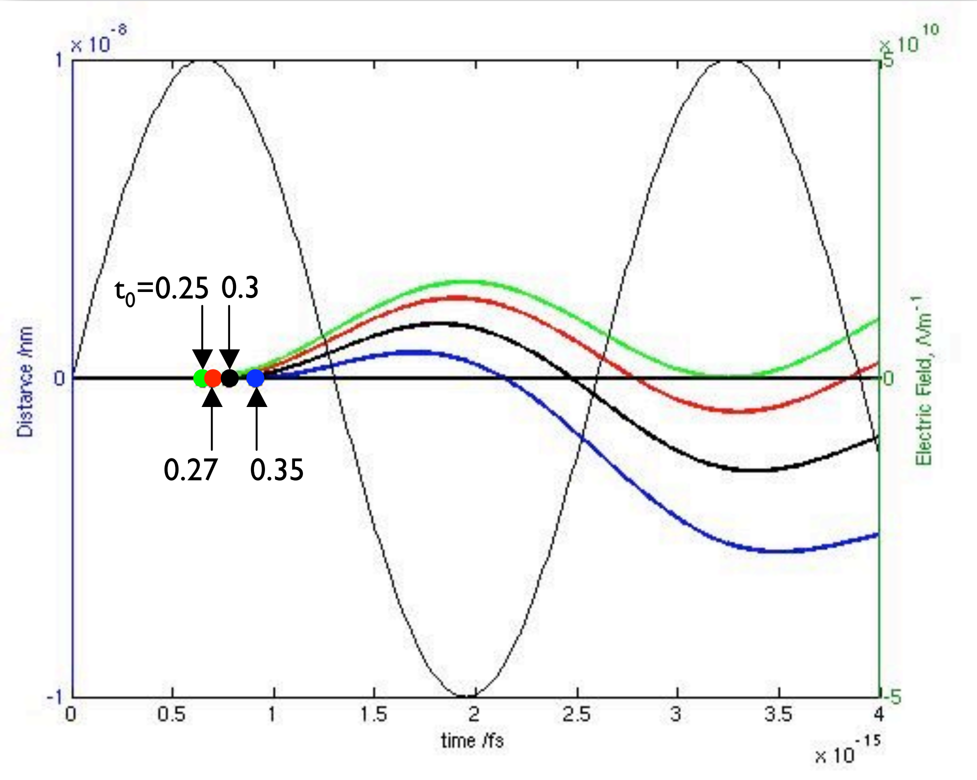
- Simple model - classical equations of motion, force due to electric field of laser:

$$F = eE_0 \sin \omega t = m\ddot{x}$$

$$x = v_0 \left[t \cos \omega t_0 - \frac{1}{\omega} \left(\sin \omega t_0 - \sin \omega t \right) \right]$$

$$\text{where } v_0 = \frac{eE_0}{m\omega}, \; t_0 = \text{time of ionization}$$

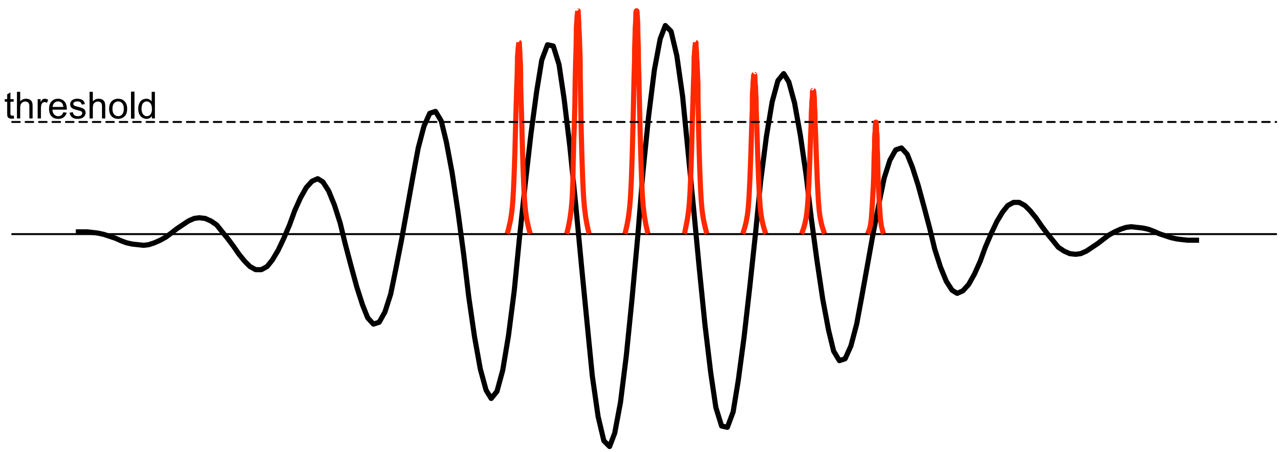
Motion of electron after ionization

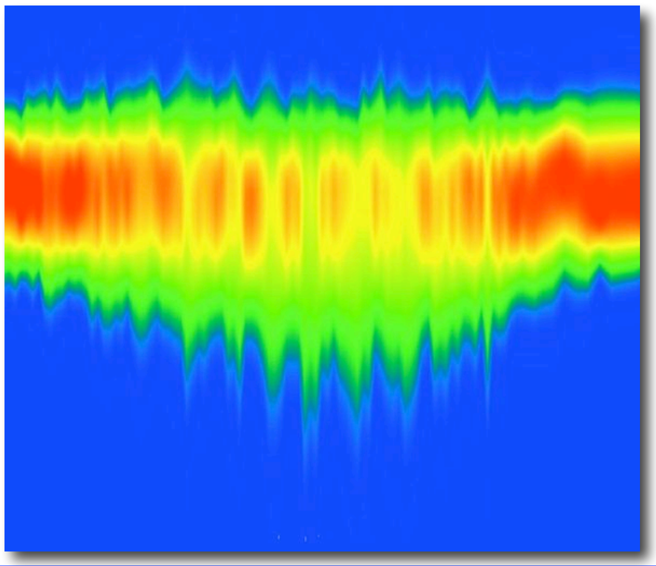


- Electron 'wiggles' in laser field after ionization
- Trajectory depends on ionization time, t_0
- Some trajectories **return** to ion core
- KE on return defines X-ray emission energy

X-ray emission during a pulse

Ionization & recollision repeats every half-cycle





M. Hentschel et al, **Nature** 414, 509 (2001)

X-ray emission during a pulse

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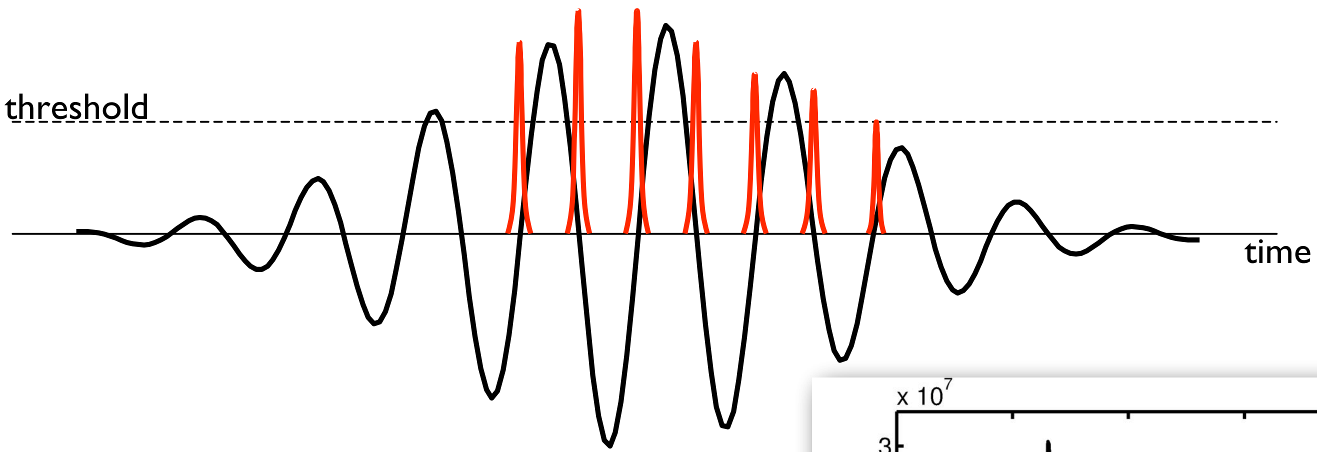
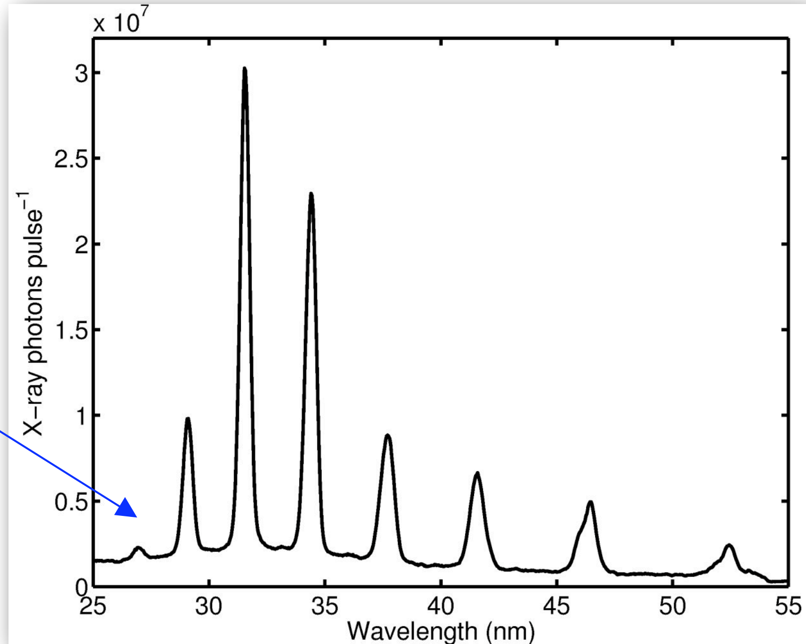
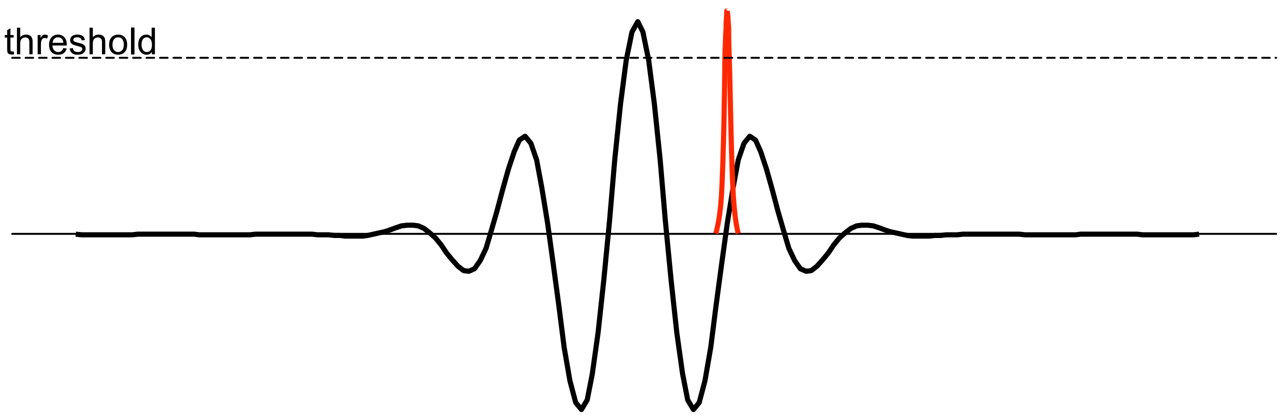


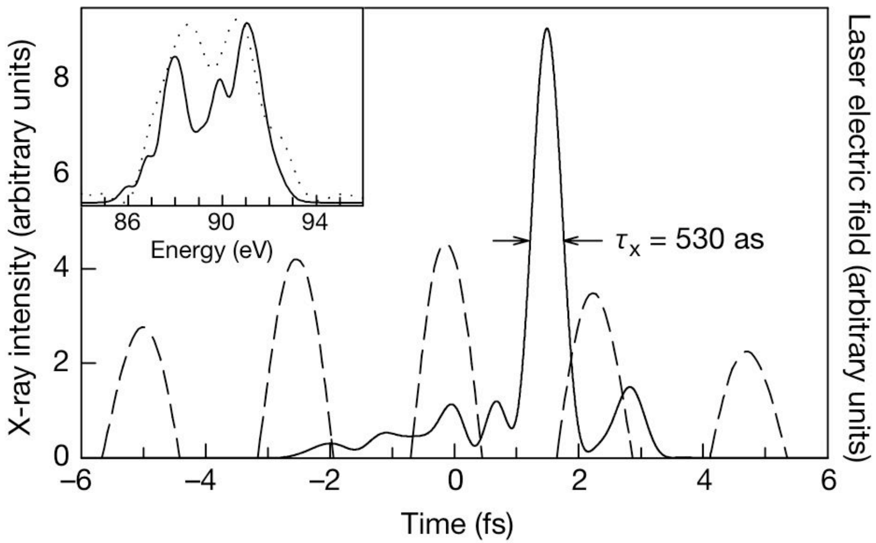
Spectrum:

Repeated pulses in time produce comb of frequencies up to cutoff



Aside: single attosecond X-ray pulses





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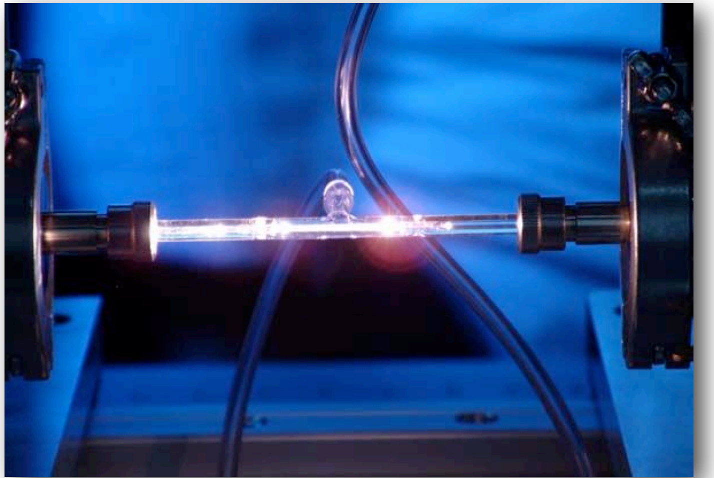
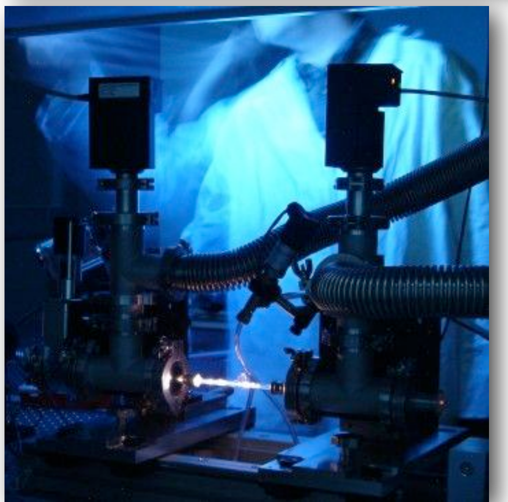
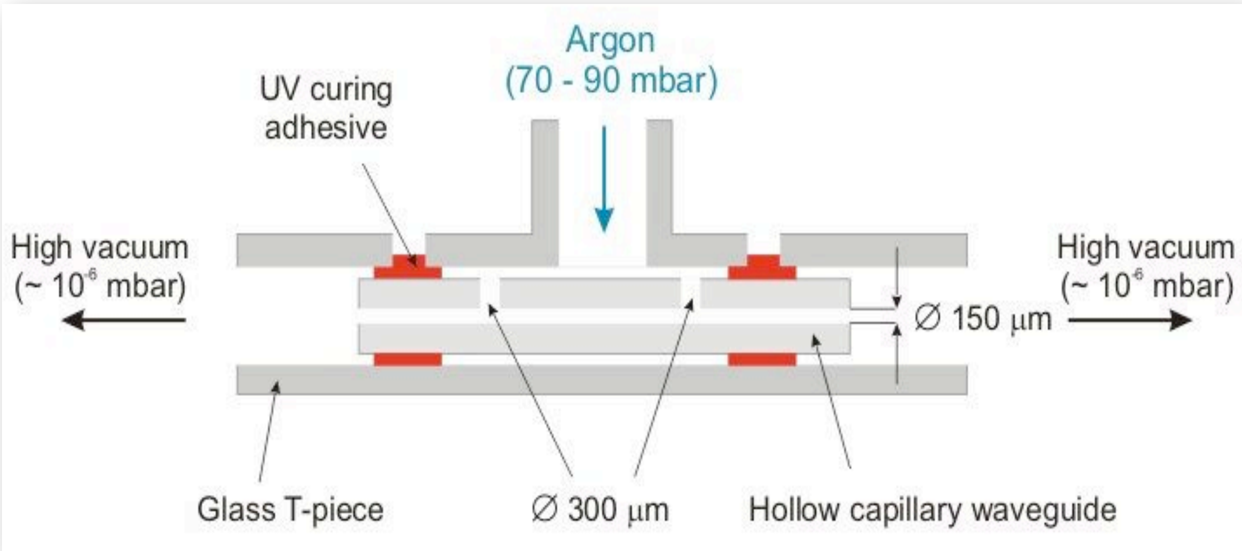
Practical X-ray generation

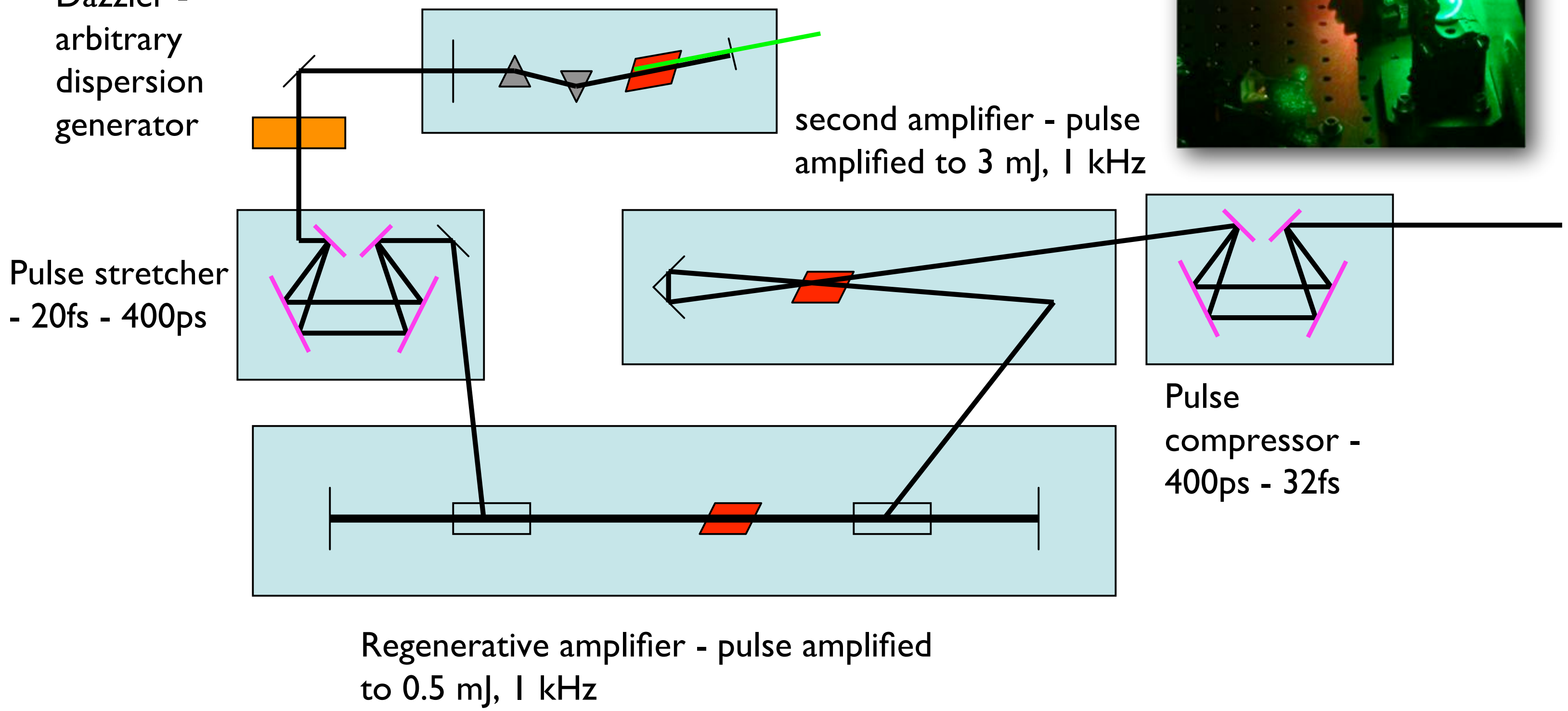


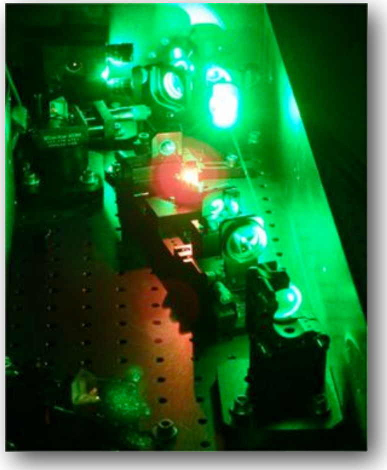


- Argon gas is a suitable medium, capillary tube holds gas at low pressure
- Laser focused into capillary, guided along 150µm bore
- X-rays generated as a coherent beam along capillary



Ti: sapphire laser system

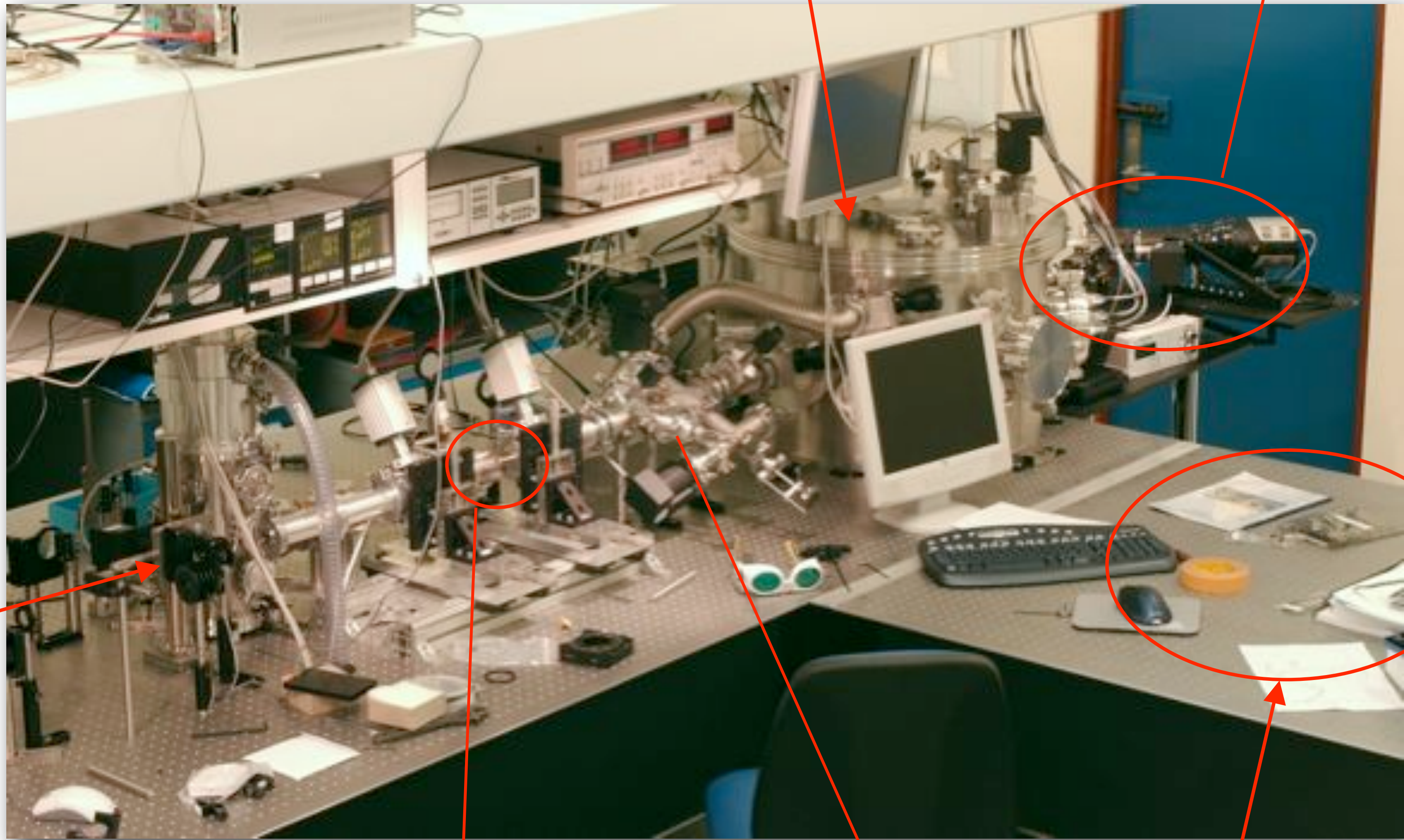




Output: 32fs pulse @780nm, 2.5mJ @1kHz
Focused intensity ~ 10^{15} W/cm^2
E field ~ 80GV/m (a.u. = 500GV/m)

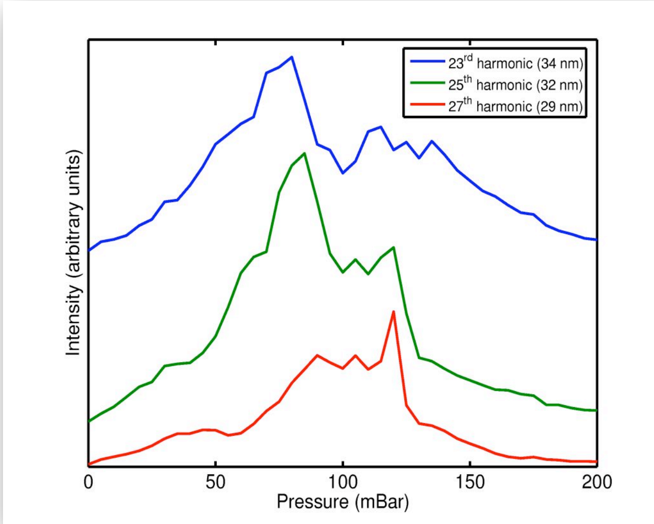
Real experiment - 2 years ago

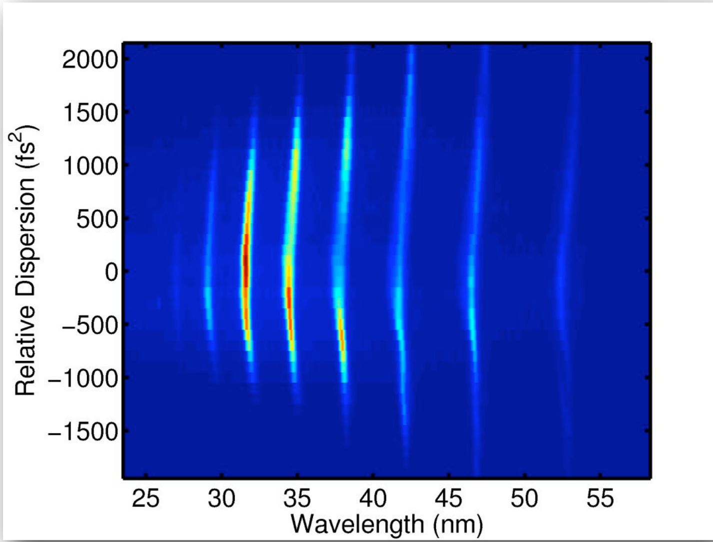
Real experiment

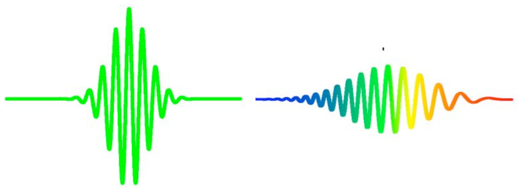


Source properties: controlling HHG

- *Phase matching* changes spectrum:
  - Gas pressure
  - Ionization level (via laser intensity)
- Laser chirp
  - Can tune over some fraction of harmonic interval

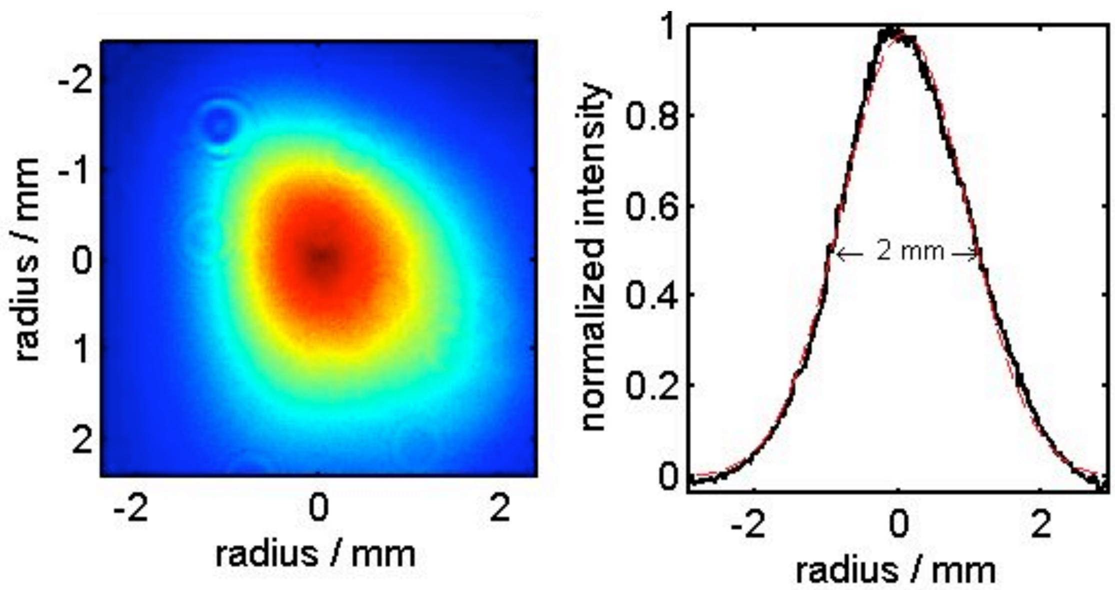




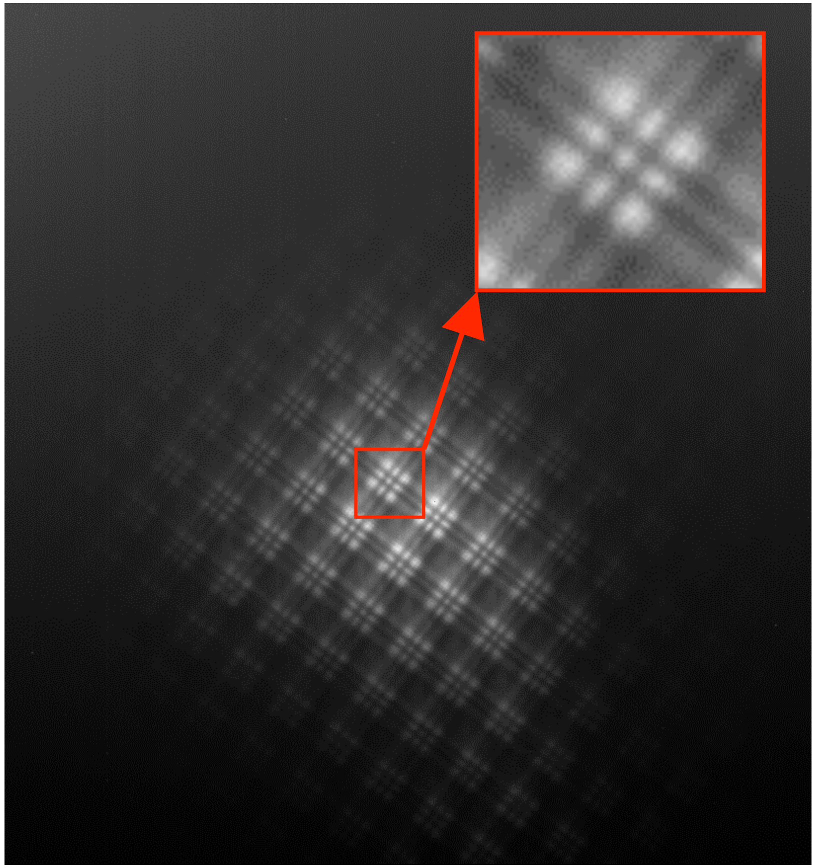


EUV/soft X-ray beam characteristics

- 10^{13} photons /harmonic /pulse /steradian
  - 10^{7} per pulse per harmonic (1kHz rep rate)
- divergence ~1 mrad, size at capillary ~30μm
- Beam profile measured 1.5 m from capillary:

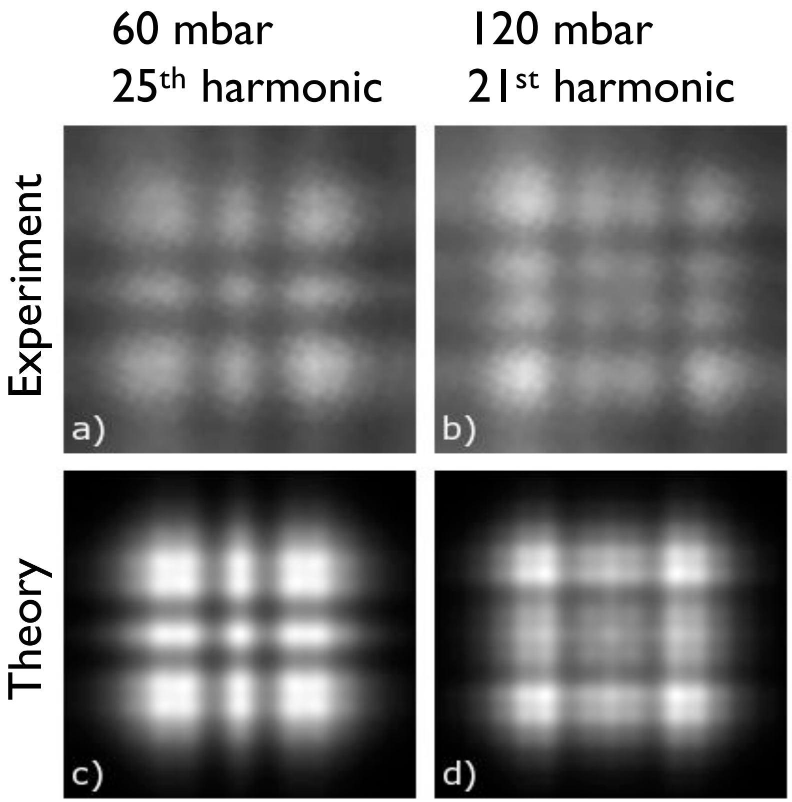


Fresnel diffraction of EUV beam



- Wire mesh: 18μm bars, 340μm spacing (Al filter support)
- Experiment and theory agree.
- Incoherent sum of all harmonics.

Fresnel diffraction of EUV beam



- Wire mesh: 18μm bars, 340μm spacing (Al filter support)
- Experiment and theory agree.
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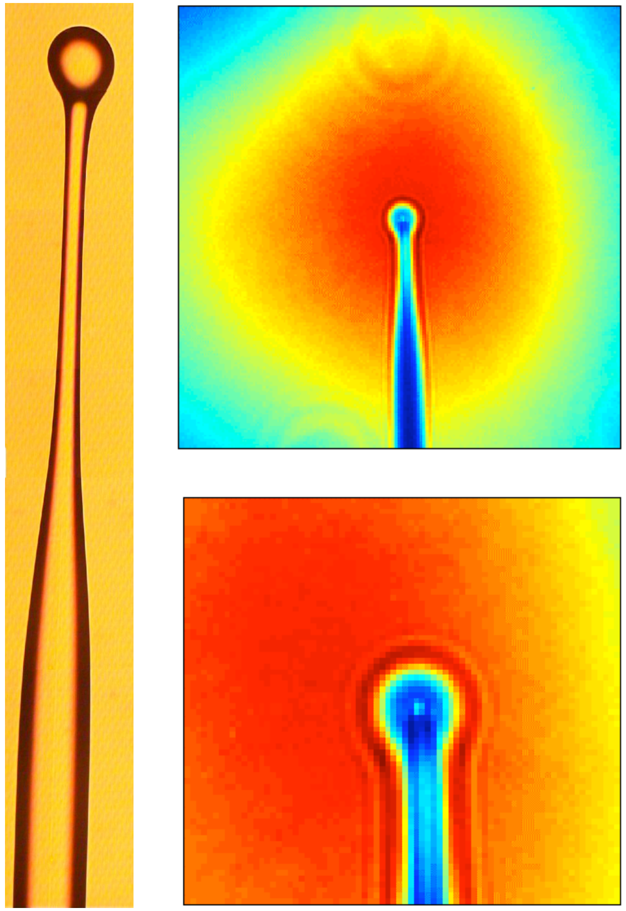
Fresnel diffraction: Poisson spots

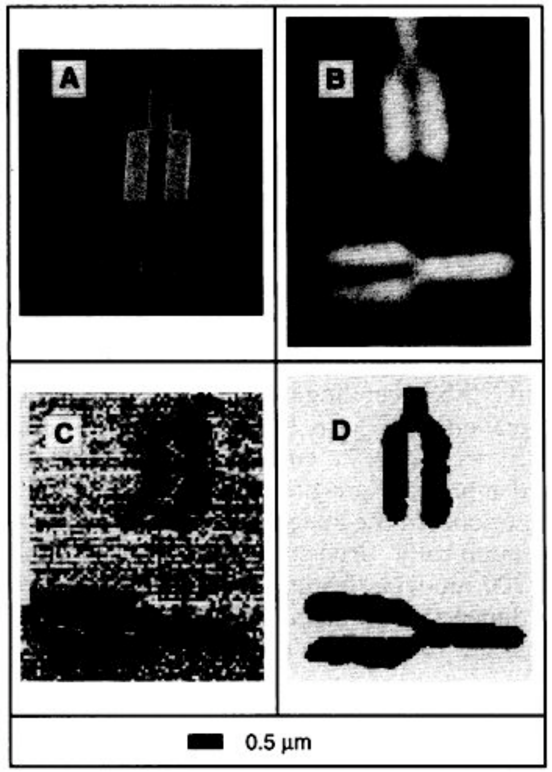
- Use fibre splicer to melt the end of fibre tapers into ~100 μm beads.
- Poisson spot formed in diffraction pattern of glass bead held in the x-ray beam.
- More accurate masks needed

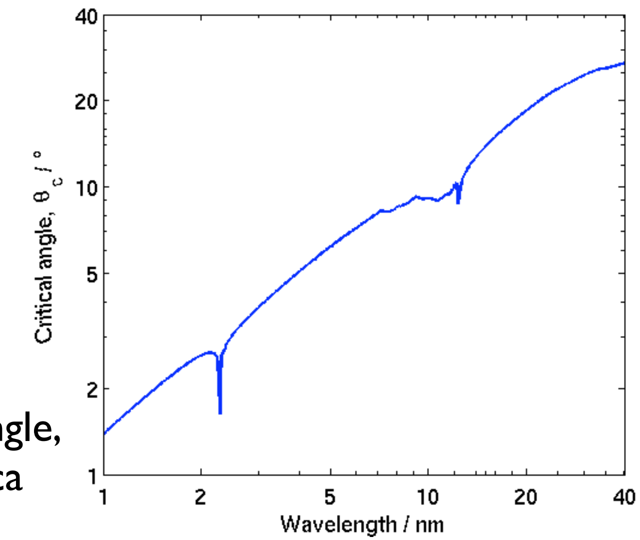


Focusing with tapered capillaries

- Successful at hard X-ray wavelengths
- Use total external reflection
  – critical angle @ 30 nm ~ 25°
- Can pull with a parabolic profile
  – single bounce focus for all incident rays

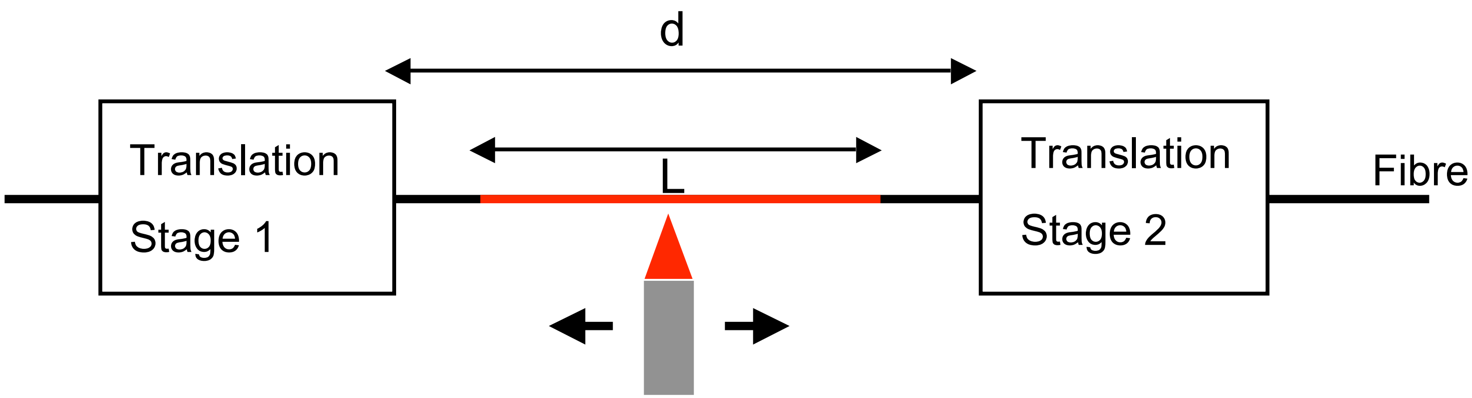
Tapered capillary x-ray concentrator (Bilderback et al, Science **263**, p5144, 1994)





Critical angle, θc for silica

Fabricating Capillary Tapers



- Technology from fibre coupler production
- Gas burner heats fibre along a length, L.
- Translation stages separation, d, increased as a function of time; burner travel, L decreased, resulting in parabolic taper.
- Model developed by Birks *et al.*, predicts parameters required for fabricating any reasonable shape taper.

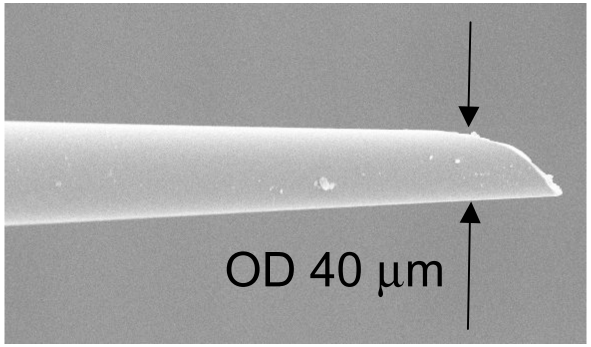
T.A. Birks et al, J Lightwave Technology 10(4), p.432 (1992)

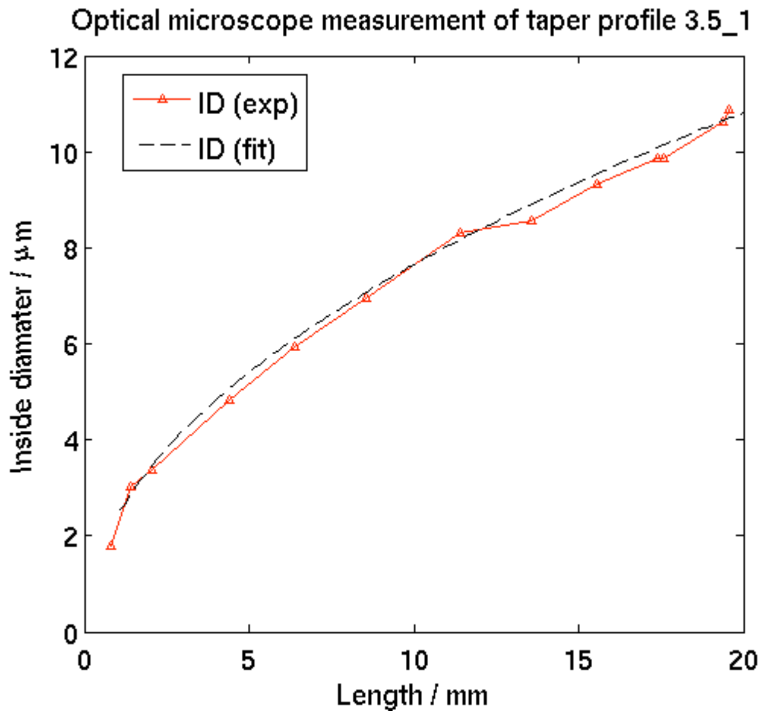
ORC fabrication facilities

- But we'll be fabricating again within weeks!

Taper profiles

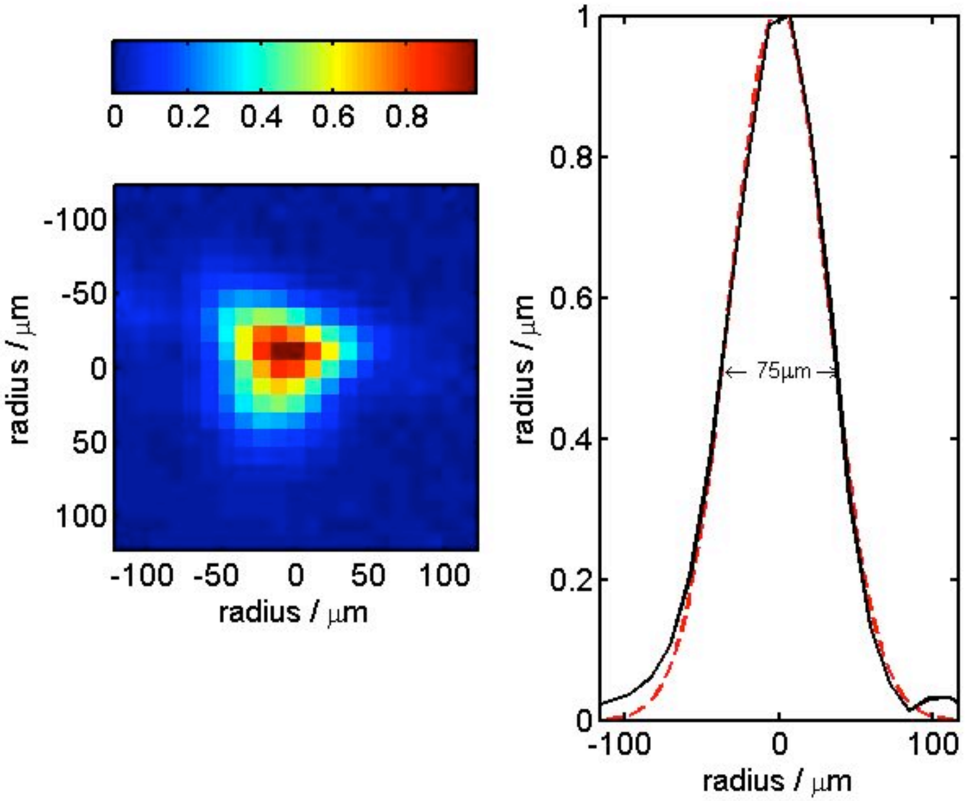
- Characterisation
  - White light microscopy
  - Scanning electron microscopy
- Variety of taper profiles manufactured
  - Smallest output aperture size currently ~ 2µm
  - Aim to reduce the tip size to create 'nano' focus
  - sub-100nm feasible



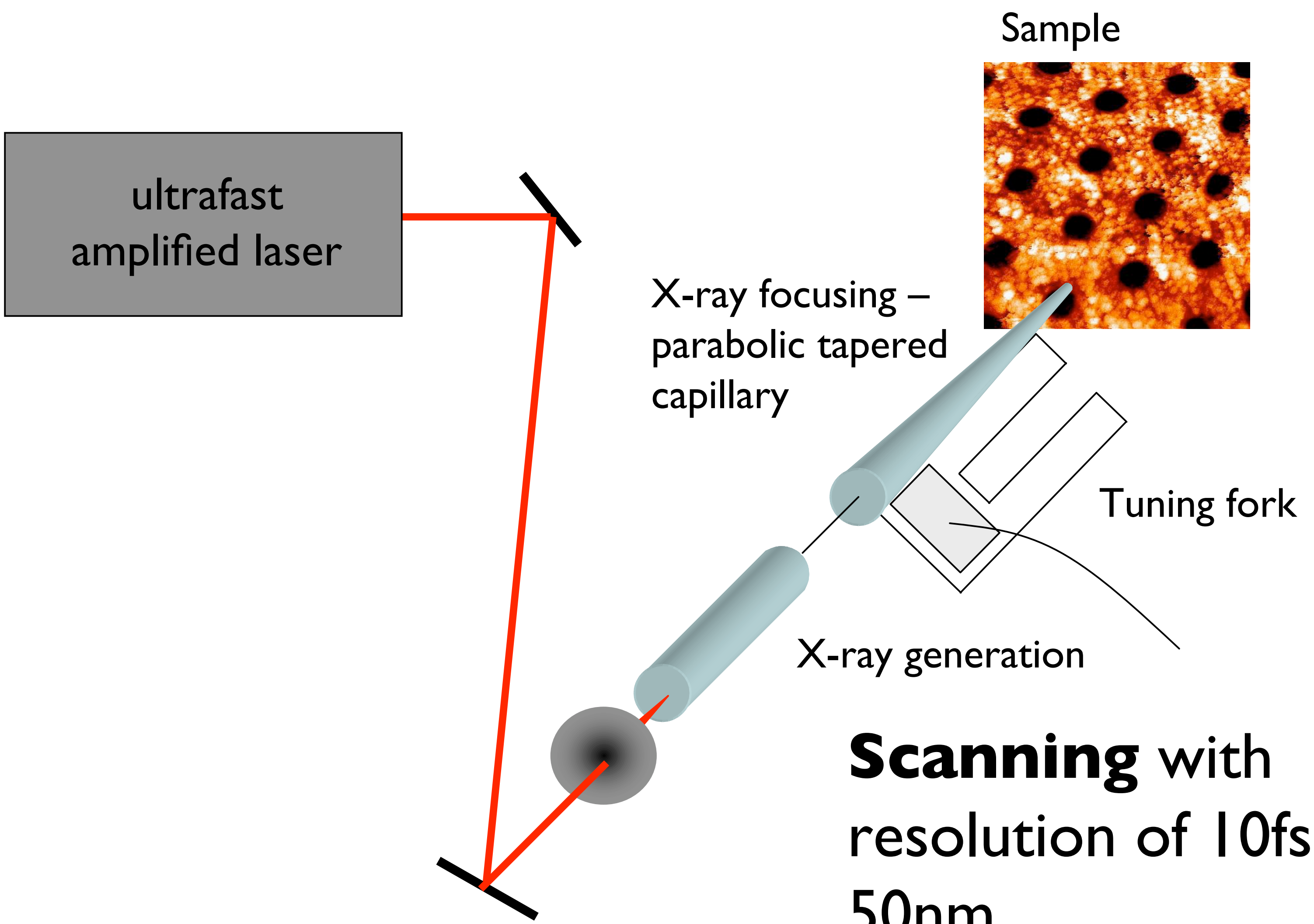


First taper results - large spot sizes

- Using large bore taper (500 μm – 100 μm)
- X-ray spot through taper, 75 μm FWHM.



Planned experiments:



Scanning with resolution of 10fs and 50nm

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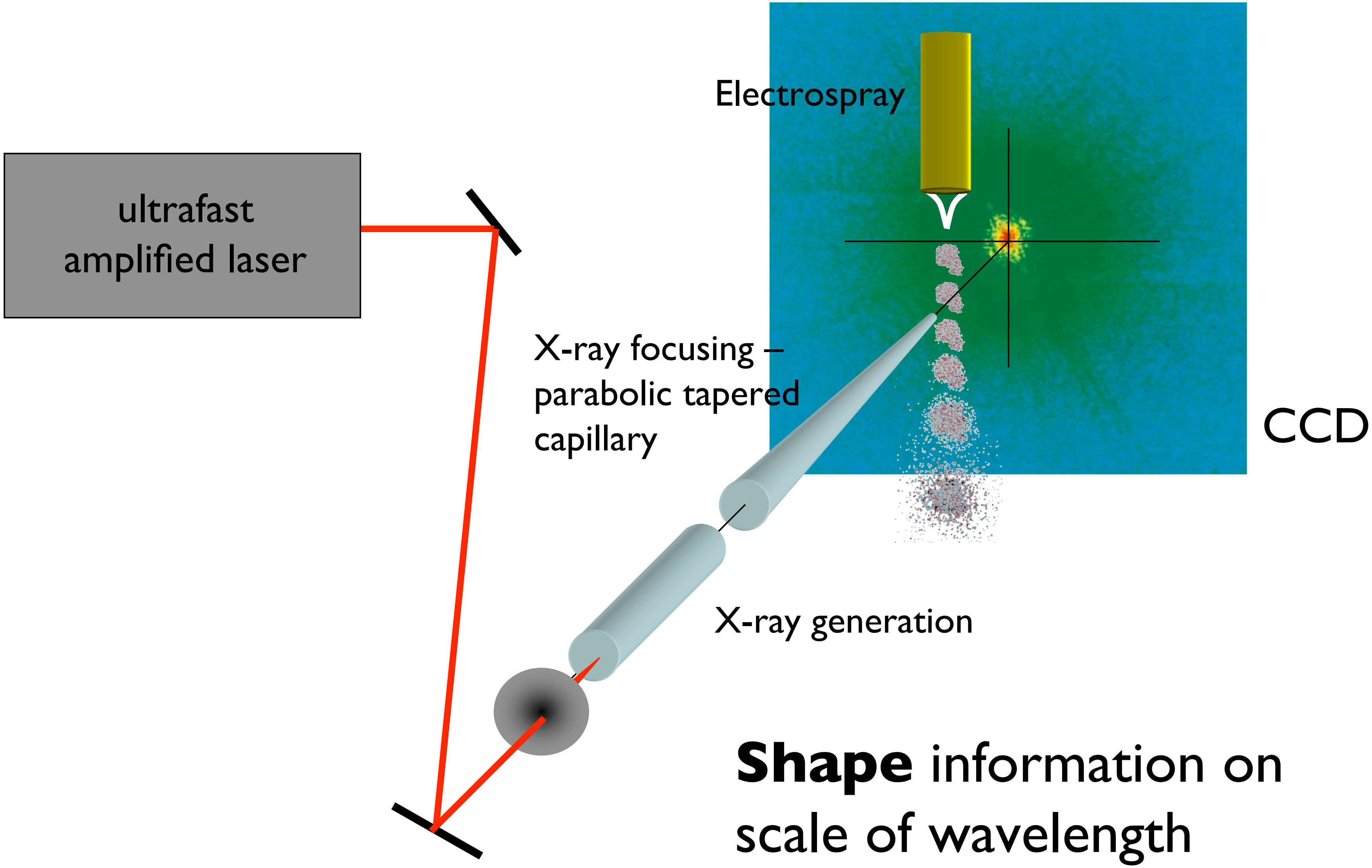
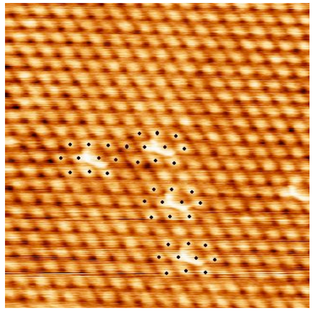
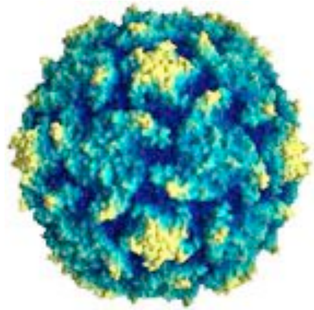


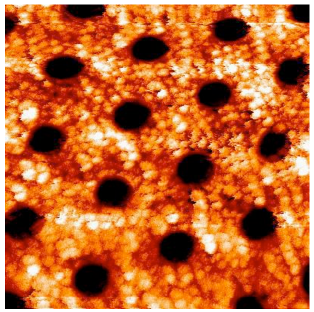
Shape information on scale of wavelength

What samples for scattering?

- Initially investigate larger, uniform systems
- Metal/DNA clusters - radiation stable, so could scatter from the same sample several times
- Large virus particles (metallised)
- Metal-labeled protein complexes
  - Back to “old school” crystallographic techniques







Future directions: X-ray source

- Shorter wavelengths - water window
  - Quasi-phasematching
    - Gets round Δk=0 condition - no ionization limitations
    - Increases shortest wavelength available - 250eV demonstrated
- More flux
  - Fibre-based pump laser systems
    - Compact solid state systems
    - high rep rate for flux increase
  - Multiple colours
    - 100-fold increase recently demonstrated using fundamental and 2nd harmonic together

Summary

- HHG provides versatile source for XUV/soft X-ray production
- Ideal beam for sub-micron focusing/positioning using tapered capillaries
- Capillary geometry ideal for SPM experiments
- Protein / nanostructure shape information on X-ray wavelength scale from Mie-like scattering
- Many possibility for future experiments, including time-resolved (fs) pump-probe studies

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Places

University of Southampton:

- School of Chemistry
- School of Physics & Astronomy
- Optoelectronics Research Centre

CCLRC Rutherford Appleton Laboratories