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XUV generation, scattering, and imaging at 29nm with an HHG source

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University of Southampton

UK workshop on Ultrafast Dynamics, 11-12/01/10

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Talk Overview: "applied ultrafast dynamics"

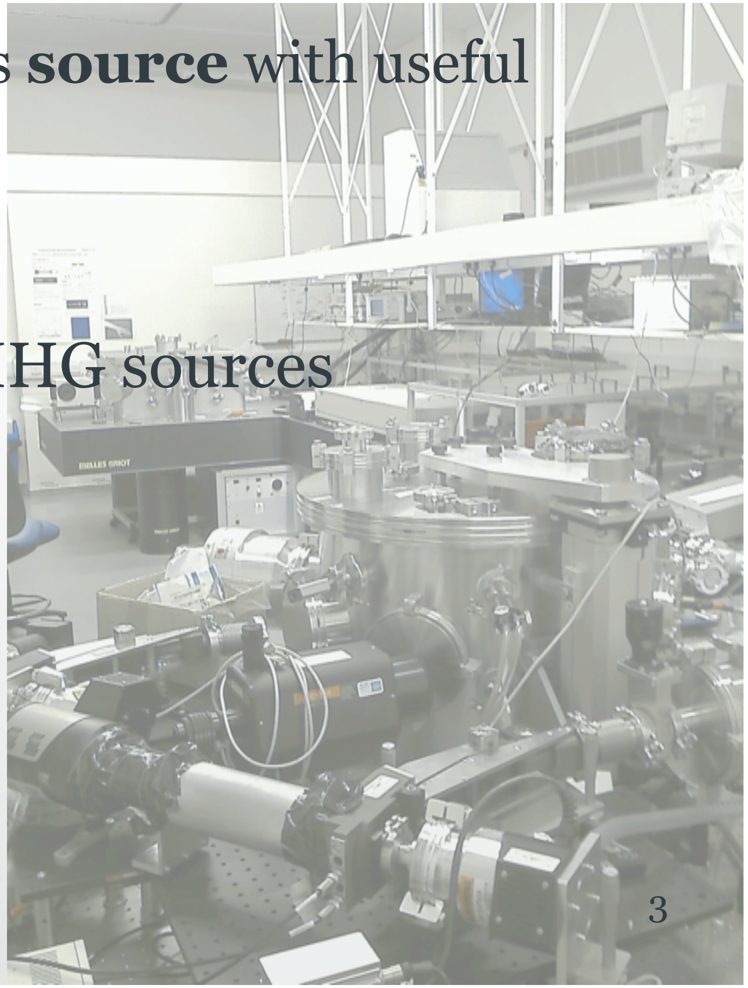
- Generation of XUV for applications using high harmonic generation:
  - Nonlinear propagation in capillary sources
  - Effects of structured gas density in jet sources
- Applications of HHG-based XUV source
  - Diffraction of 29nm light from self-assembled strutures
  - Imaging using phase retrieval techniques
- Future work

Group Research aims

- XUV/soft X-ray generation using ultrafast pulses
  - High harmonic generation (HHG) as **source** with useful characteristics

Research programs

- Development of efficient, tractable HHG sources
- Use of HHG sources in:
  - **imaging**
  - spectroscopy



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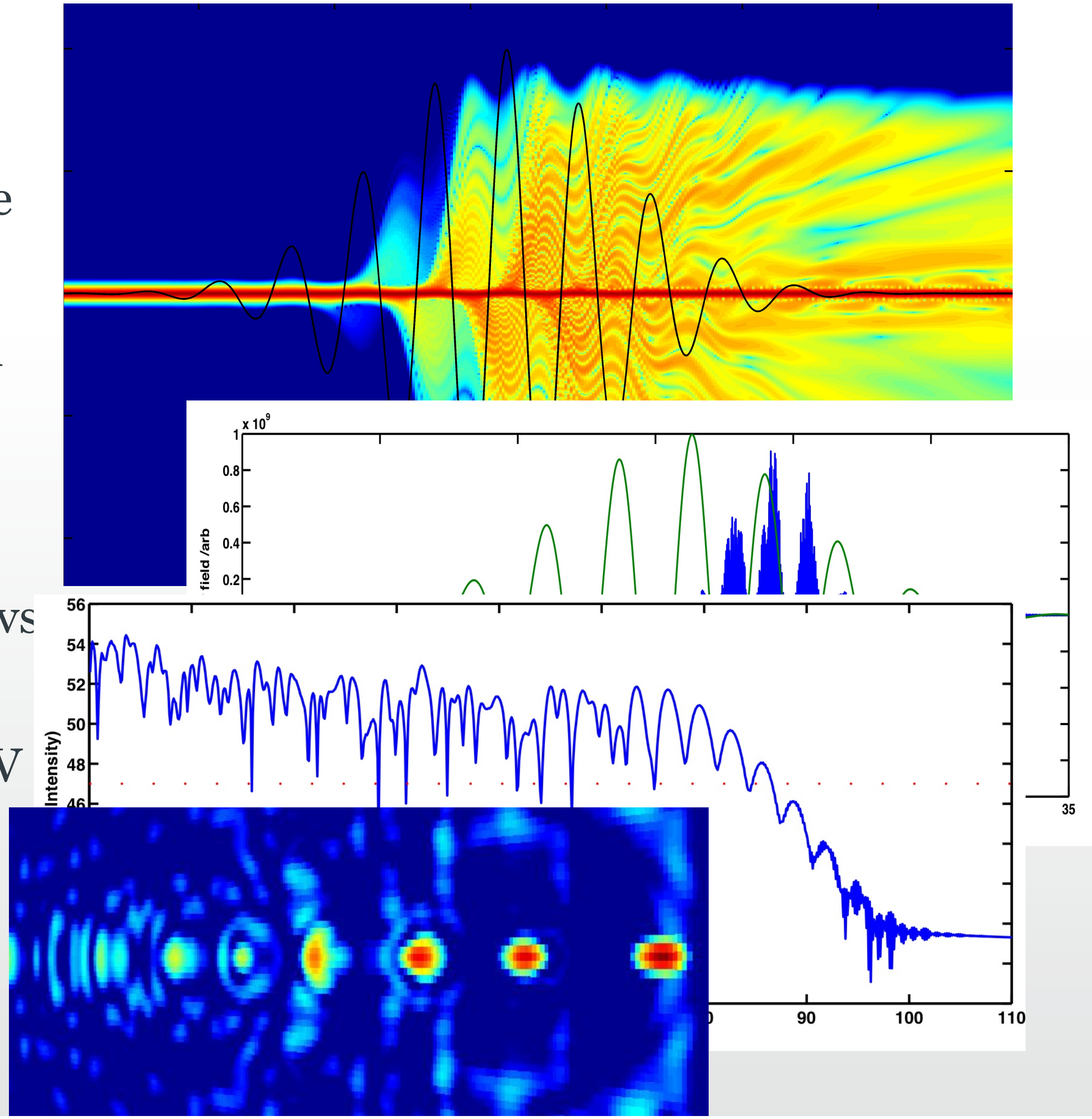
HHG– single atom process

- Electron wavefunction is partially dragged out of atom core
- On returning, it *interferes* with the part that was left
  - Oscillating electron motion in time
  - Emission from oscillating dipole
- Calculation: electron acceleration vs time
  - Fourier transform to give XUV spectrum
  - Propagate to far field to give observed spectrum



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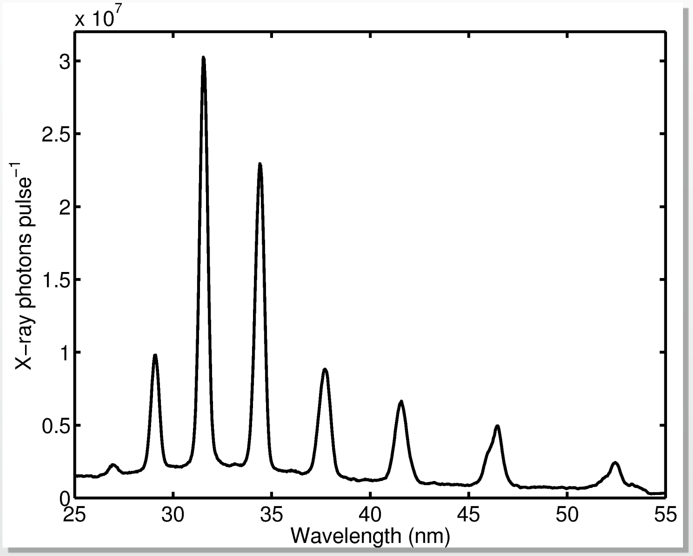
HHG sources at Southampton

- Pump laser: Ti:sapphire – 800 nm
  - 38 fs pulses, 3 mJ pulse energy, 1 kHz rep rate
  - mid 10^{14} W/cm^2 when loosely focused
- Geometrical phasematching via capillary waveguide or Guoy shift in gas cell (both sources used experimentally)
- XUV source output parameters:
  - Wavelength 18-40 nm
  - Efficiency ~10^{-6} – 1W input, 1 μW out
  - Low divergence (<1 mrad), high spatial coherence
  - >10^7 ph/pulse/harmonic – brightness similar to synchrotron
  - M^2 ~ 2 before focusing
  - Pulse envelope length ~10 fs



Producing an efficient XUV source

Biggest issue in engineering a useful HHG source: **Phase matching**

- Geometrical effects used to compensate for index mismatch
  - Jets – Gouy shift offsets single atom phase factors, gas index
  - Capillaries – waveguide phase velocity offsets against gas index

Both typically assume **linear propagation –** limits pump laser peak intensity

Significant issues

- How to get good conversion efficiency without requiring linear propagation?
- How to increase interaction lengths above that allowed by the Gouy shift
- How to control the spectrum of the output radiation?

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XUV generation in gas-filled capillaries: nonlinear propagation models

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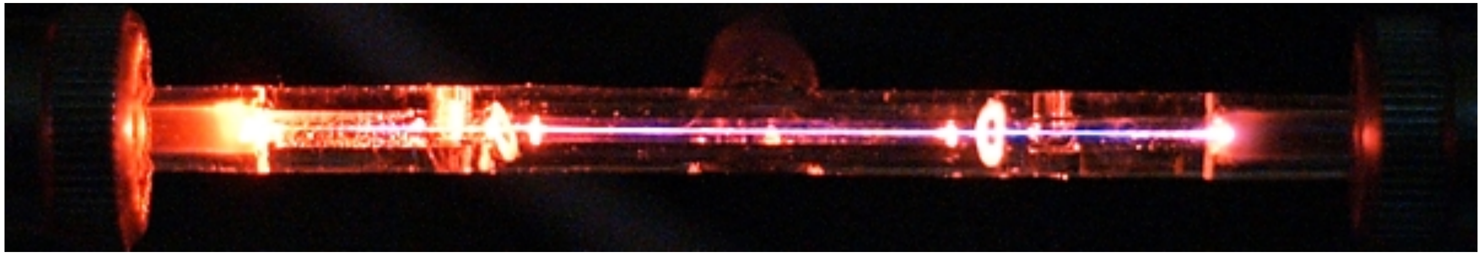
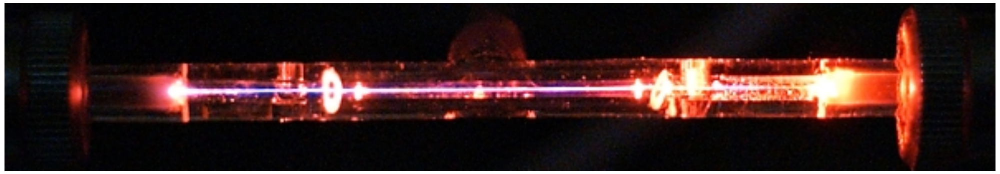
XUV generation in capillary waveguides



- Capillary can extend interaction lengths considerably
- Phase matching by using waveguide mode index of capillary
  - Reasonable model for low intensities
- XUV generation produces plasma in significant percentages
  - Effective nonlinearity couples modes
  - no “cut-off” in the classic fibre sense - all modes can propagate, unlike fibre case
- Need **nonlinear propagation model** to predict XUV generation



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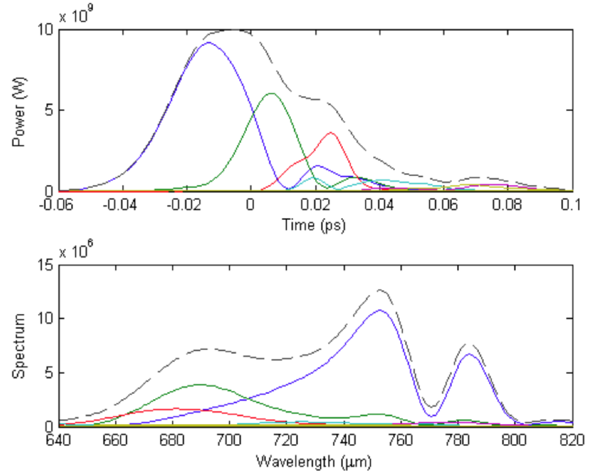
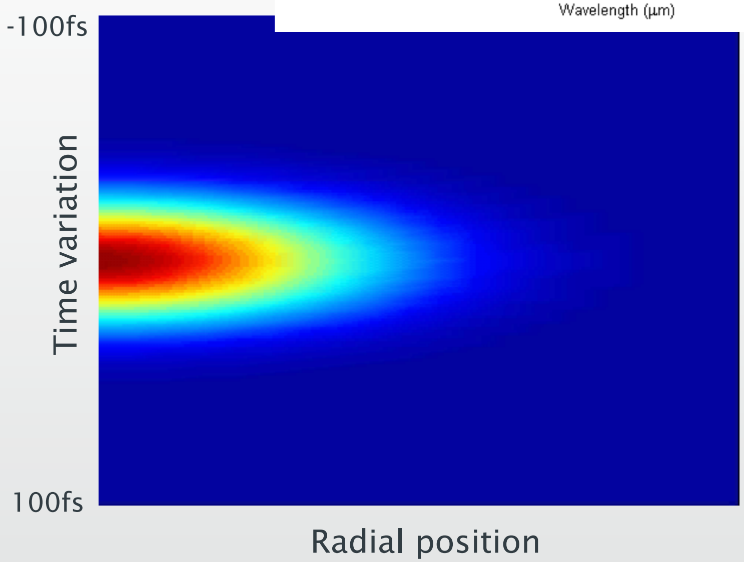
Modelling nonlinear propagation in gas-filled capillaries

- Model: split-step propagation with nonlinear mode mixing:
  - Adapted from fibre models: *(Poletti & Horak, JOSA B* ***25****, p. 1645 (2008))*
    - Nonlinearities from plasma generation as well as gas $\chi^{(3)}$ etc.
    - Considers 20-30 radially-symmetric modes
  - Significant pulse distortion, spatial and spectrally
  - Significant compression





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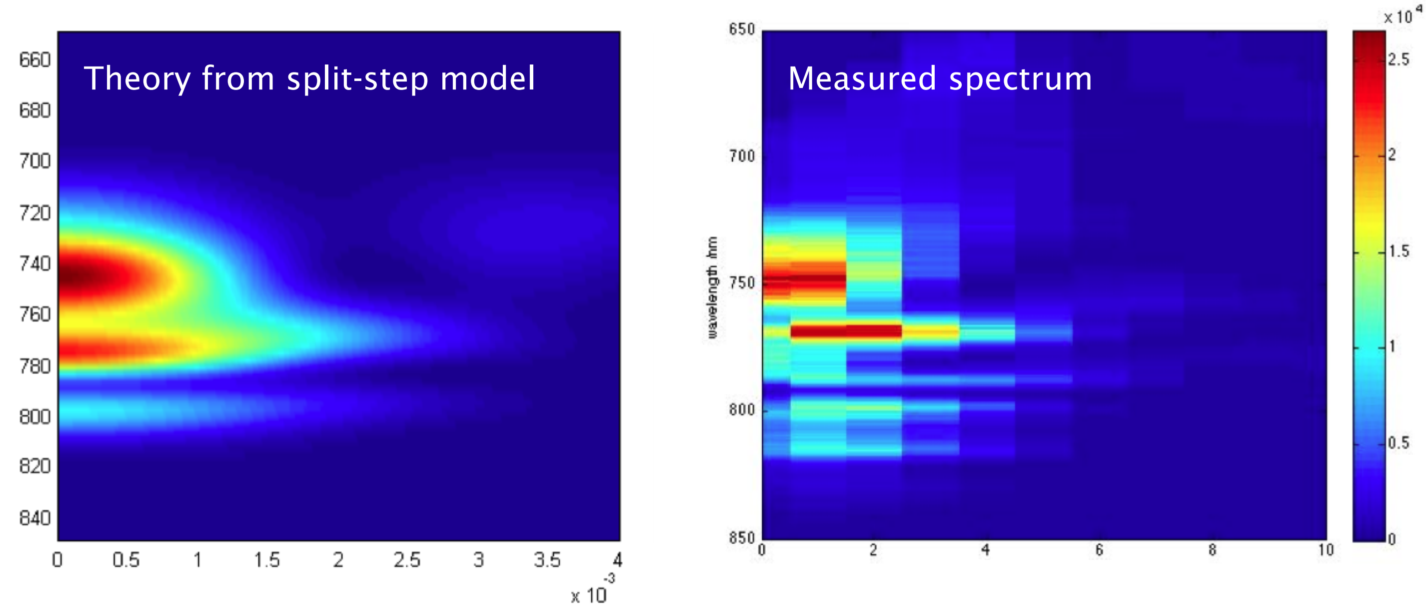
Comparison with experiment:

- Looking at radial distribution of pump wavelengths in the far field of 7 cm capillary output:



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Scattering and imaging at 29 nm

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Motivation - applications of imaging

- Water window X-rays for biological imaging: 280 - 530 eV
  - resolution spans length scale from SEM/TEM to optical – 10nm to 1µm
  - contrast from difference in carbon and water absorption
- Presently being done at synchrotron sources – could be done with already-demonstrated HHG sources.

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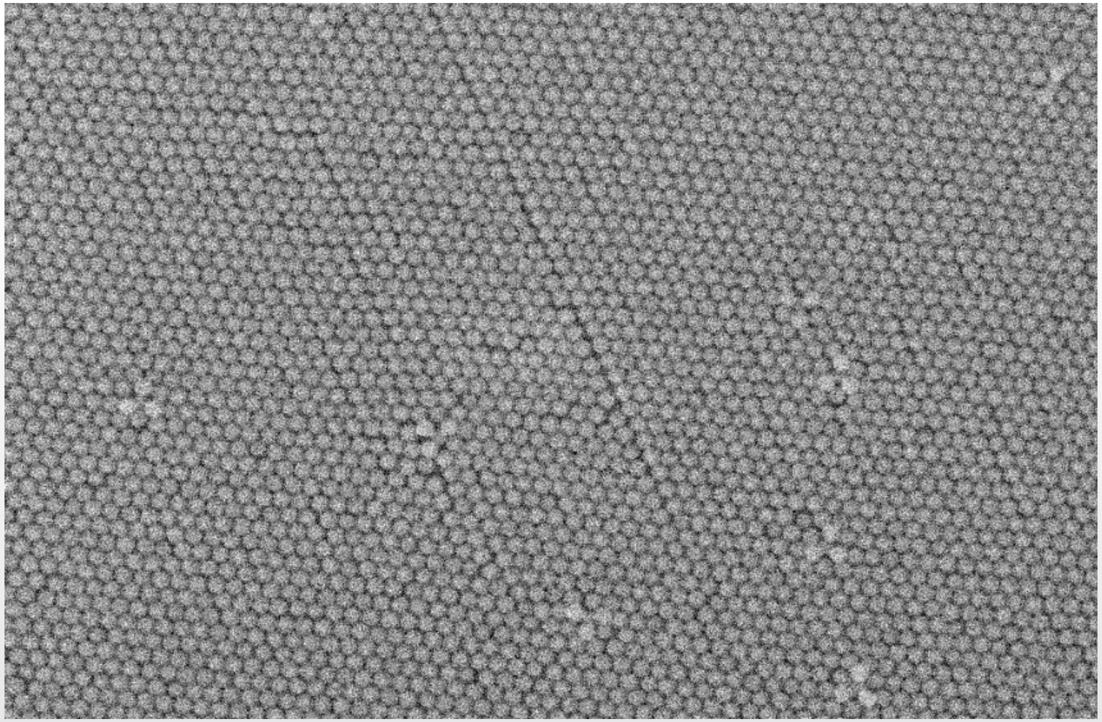
Diffraction at 29nm: structural effects & refractive index determination

- Samples: self-assembled 200nm diameter sphere arrays



- Diameter 196nm, size variation < 5%
- Single layer of spheres on 50 nm SiN membrane
- Ordering good, but not perfect.
- Uses: photonic/plasmonic crystal templates

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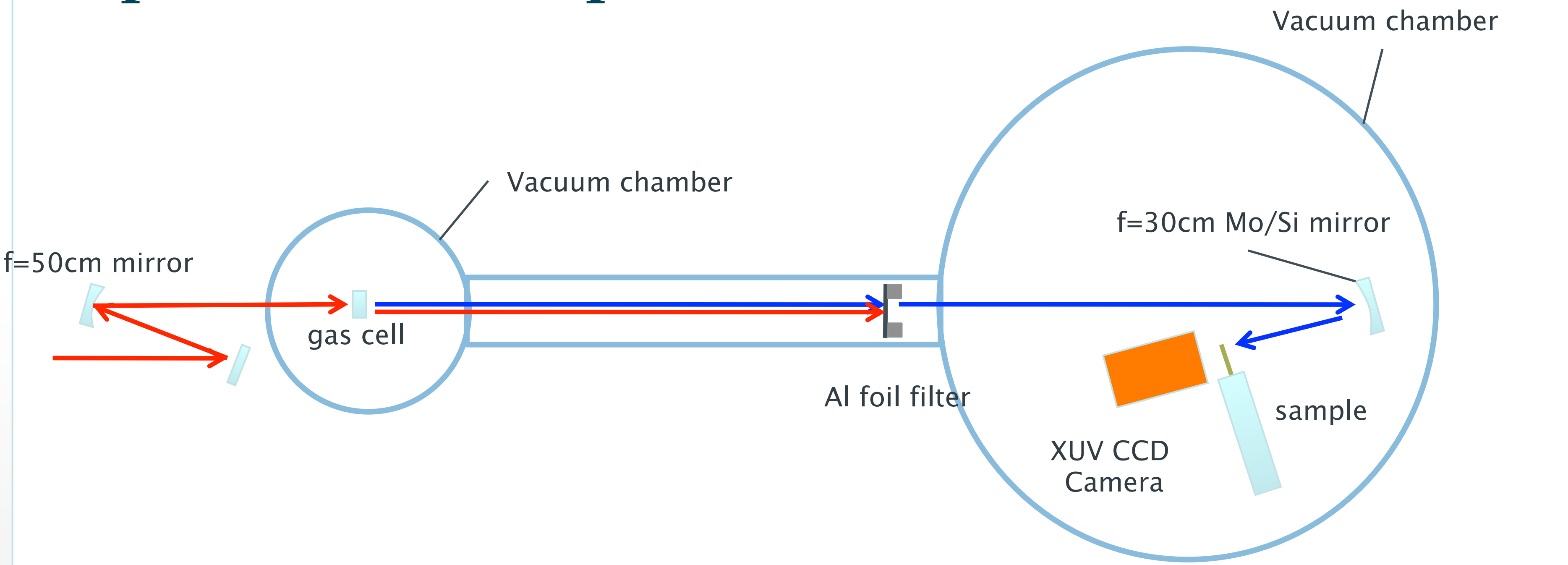
Experimental setup



XUV source: HHG in Ar-filled capillary or cell, peaked at ~29 nm
XUV mirror: Spherical Mo/Si multilayer (IOF Jena)
Detector: ANDOR XUV CCD, 17mm from sample

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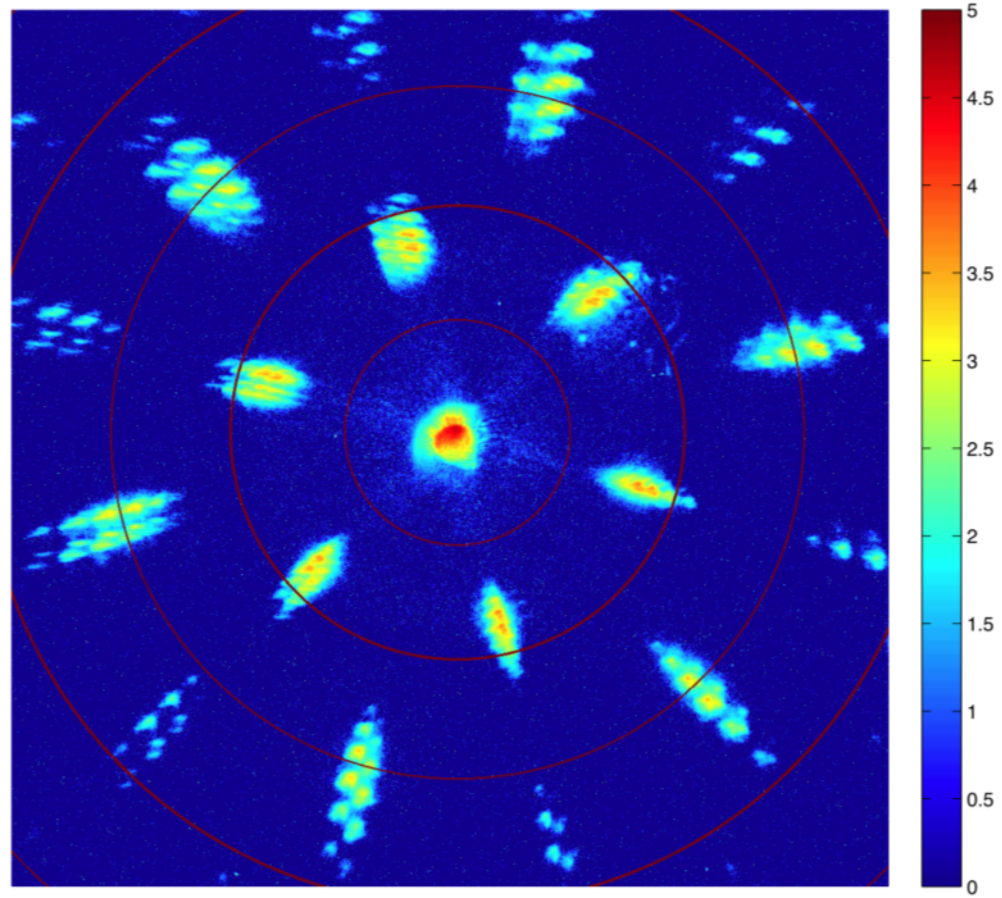
Scattering from ordered sphere regions

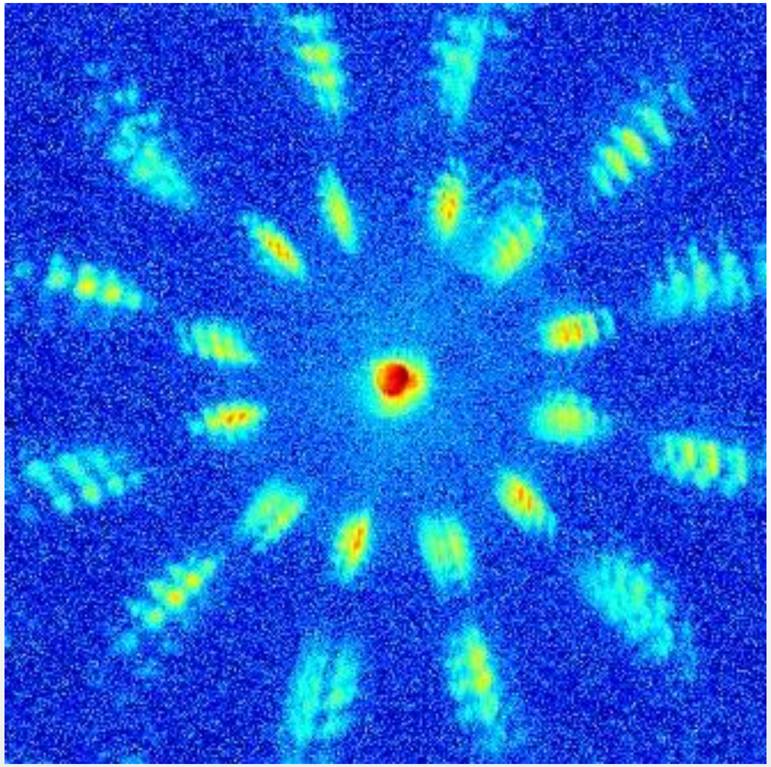


Red rings are 100 mrad angle contours
Intensity scale is logarithmic.

XUV transmission diffraction from 196nm sphere array, ~10μm XUV focal spot on sample

•Radially: multiple wavelengths give multiple spots
•Tangentially: structural information
•Other distortions arising from XUV phase front distortion

Scattering from multiple grains



Scattering patterns are very sensitive to grain boundaries

This image shows the ~20μm XUV spot positioned over two grains, with ~30°between the lattice orientations

XUV beam positions with single crystal diffraction patterns are common across samples

Modelling scattering using Mie theory:

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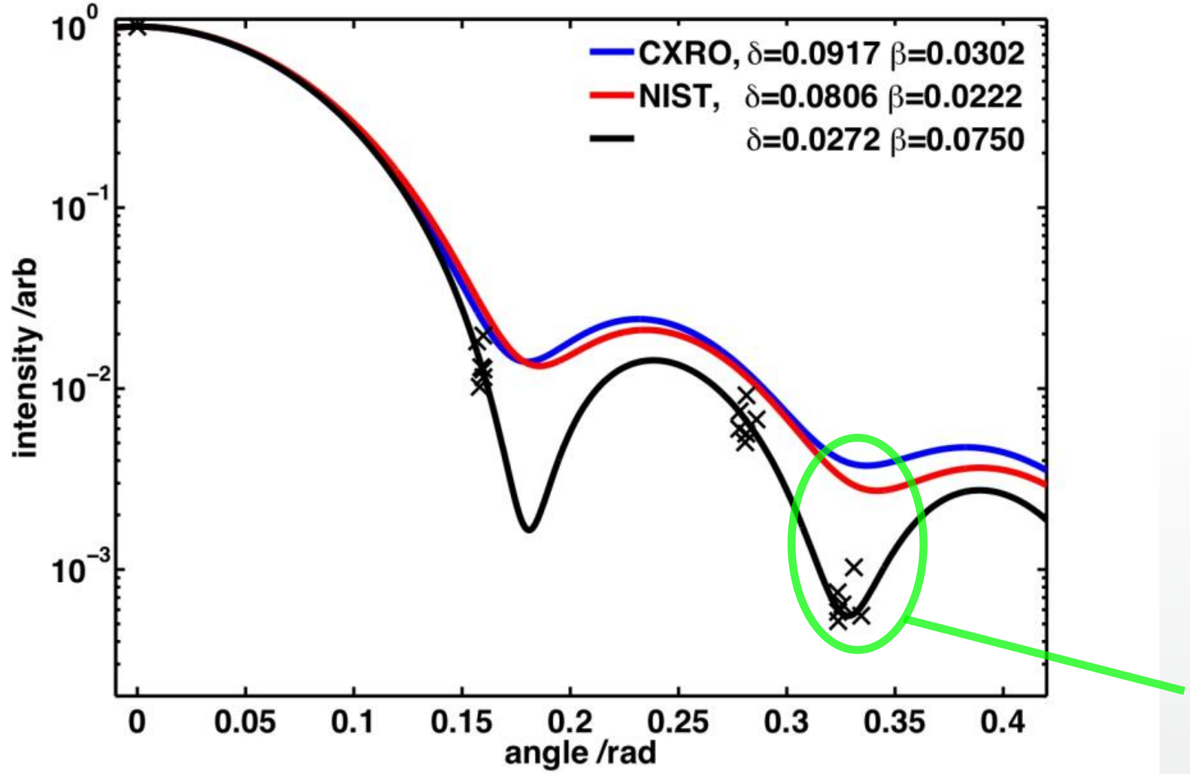

Scattering form factor calculated using Mie theory

Refractive index in XUV is usually written as $(1-\delta) + i\,\beta$.

Variation of real or imaginary parts affects different scattering orders *differently*, so it is possible to extract *both* from multiple scattering order data

- Red & blue lines are predicted scattering using NIST and CXRO database values of index
- Solid line is for values given in legend

Index variation with wavelength

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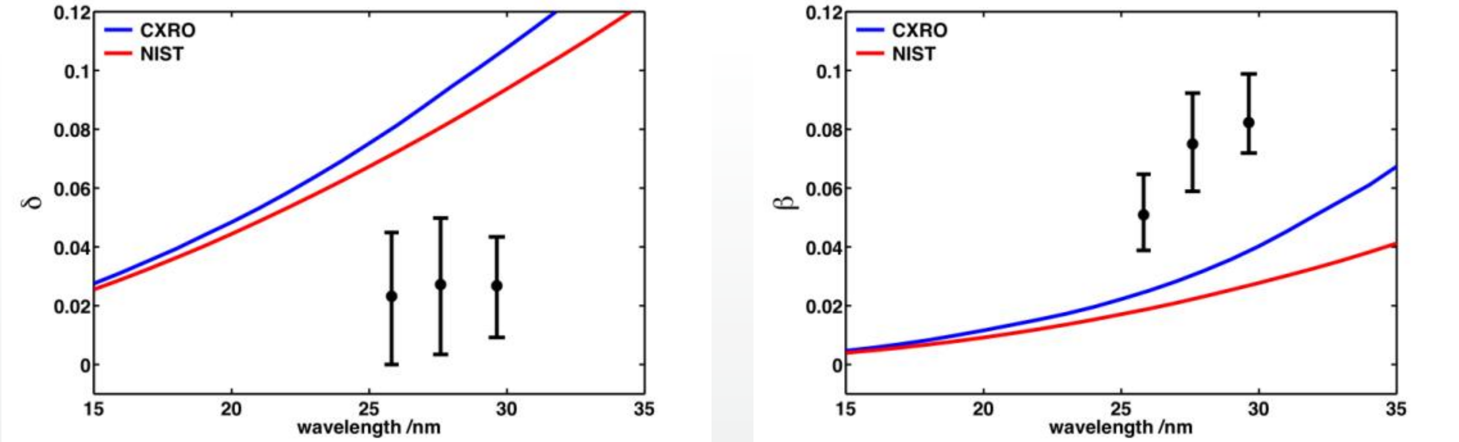
Analysis of the data can provide a value for the complex index to compare to theoretical values calculated from oscillator strengths.

Plot below shows theoretical data for δ and β (dotted lines) and our data points from XUV Mie scattering



Mills et al, Appl. Phys. Lett. **93**, 231103 (2008)

XUV diffraction gives a more accurate value for the dielectric constant than extrapolation techniques

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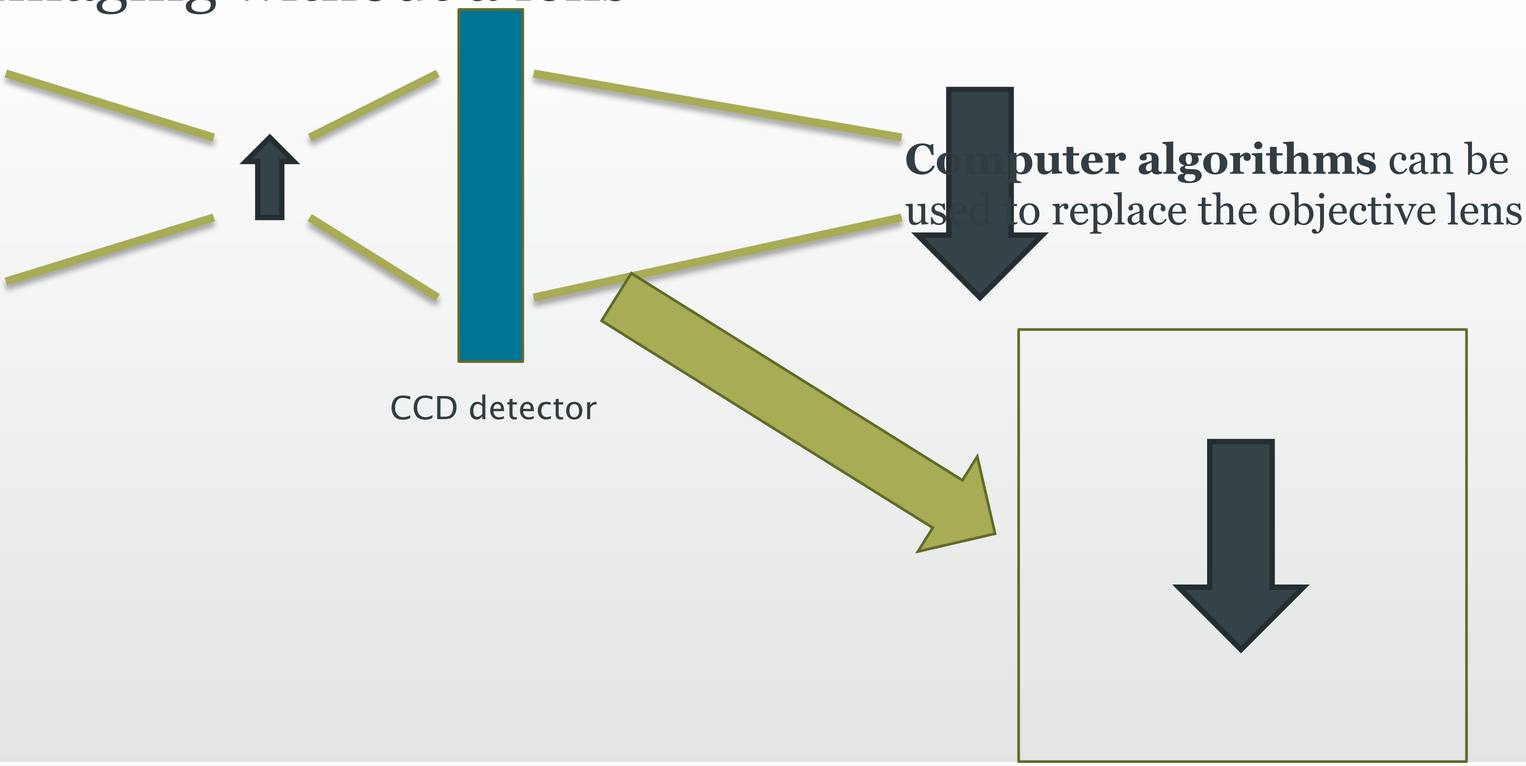
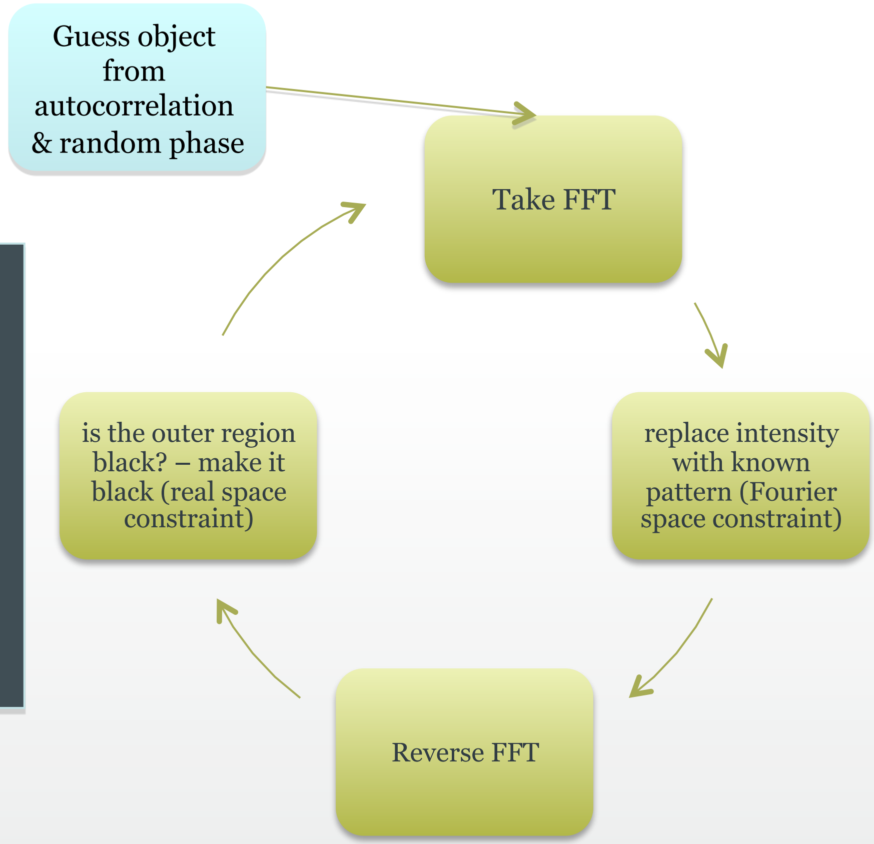
Lensless imaging, or Coherent Diffractive imaging (CDI)

- Microscopy in XUV is hard because of lack of optics
  - Possible using zone plates, or multilayer mirror objectives
- Imaging without a lens

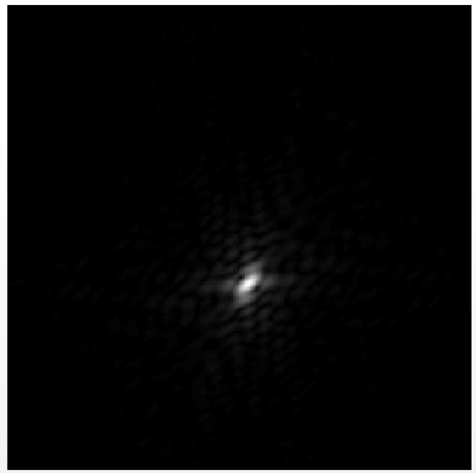


Example of phase retrieval algorithm:

Real space



Fourier space



Development:

- W. Gerchberg & W.O. Saxton, Optik 35, p.237 (1972)
- J. Fienup, Optics Letters 3, p.27 (1978)

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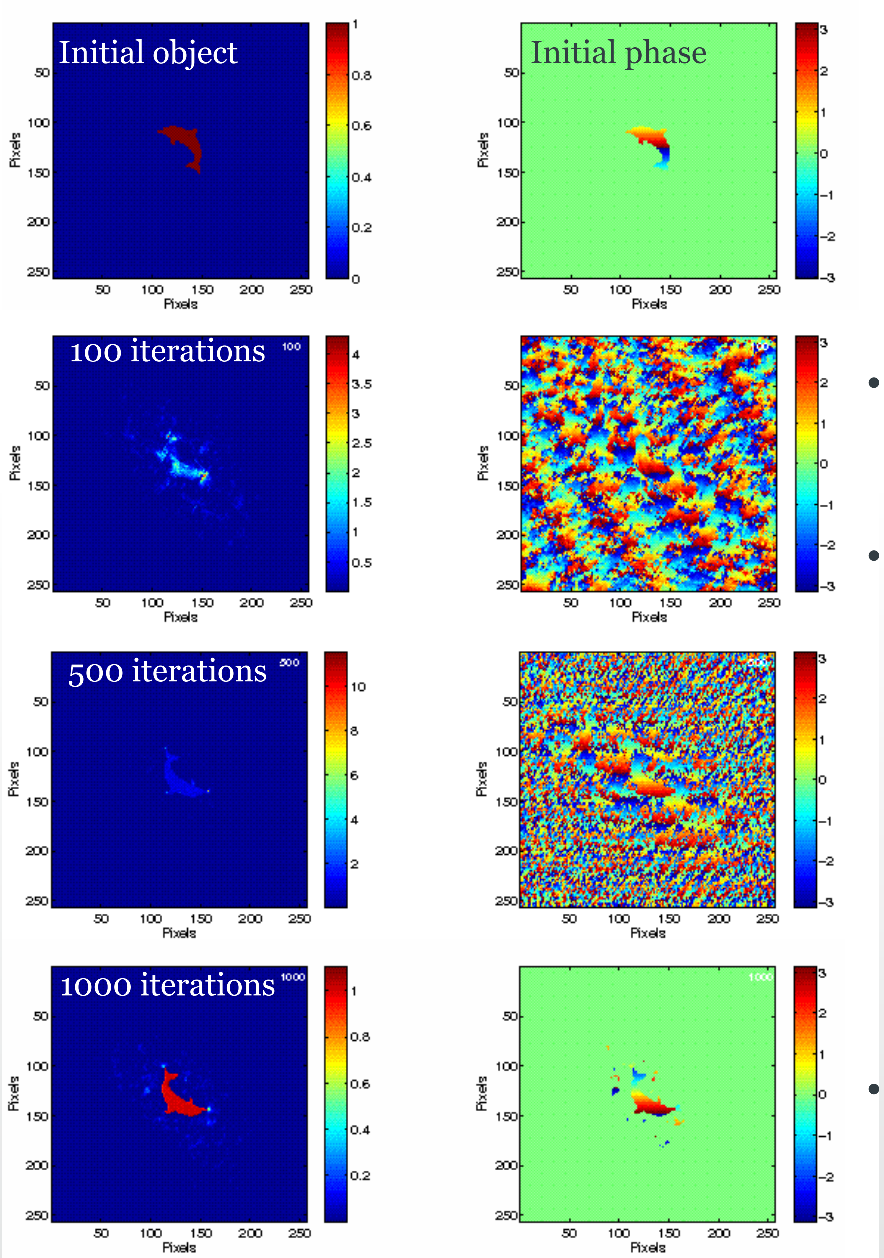
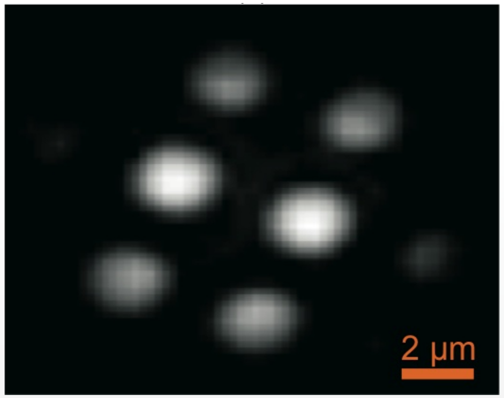
Phase retrieval in simulations



- Intensity and phase are both recovered well in most cases
- Tricks to ensure you don’t get stuck in local minima
  - *HIO* – hybrid input/output
    - Controls how “hard” the constraints are applied
  - *Shrinkwrap*
    - Dynamically changes real space constraint
    - Improves reconstruction with noisy data
- Computing time is relatively short
  - Few minutes for small images

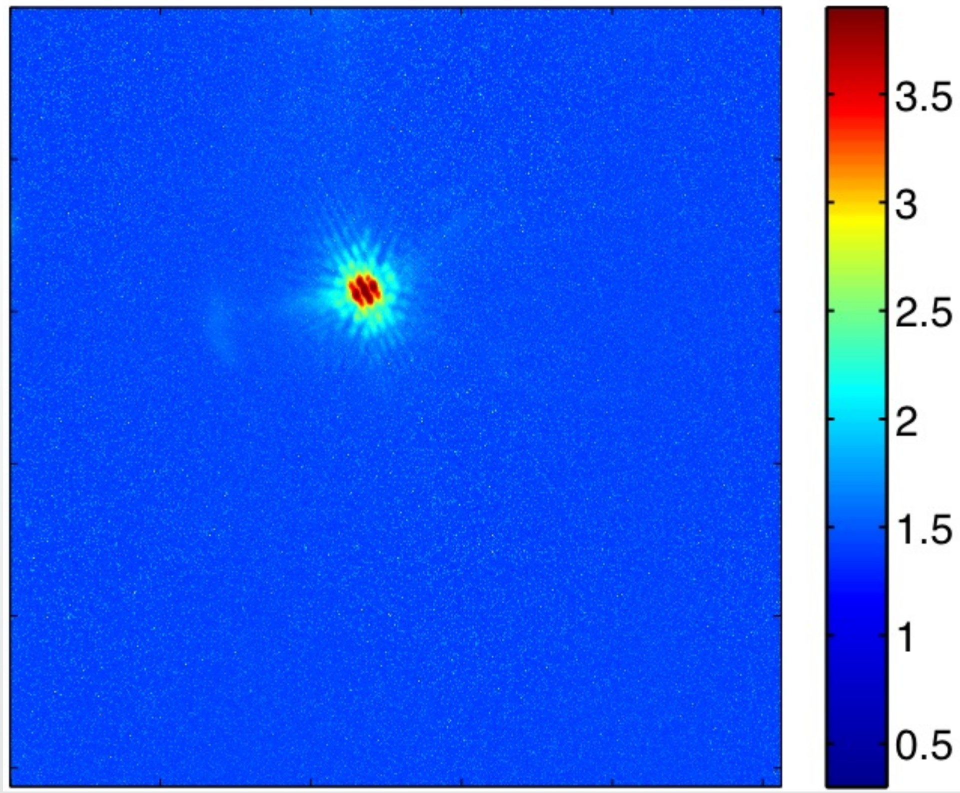
Experimental imaging at 29nm using CDI

- Object: 2µm diameter holes in Au-coated Si_3N_4 membrane, apertured by 5µm pinhole



Optical image is distorted by diffraction from 2µm holes – pinhole and membrane are not flush.

Log$_{10}$ of scattered intensity from pinhole/mask object



HIO/shrinkwrap: evolution of retrieved image

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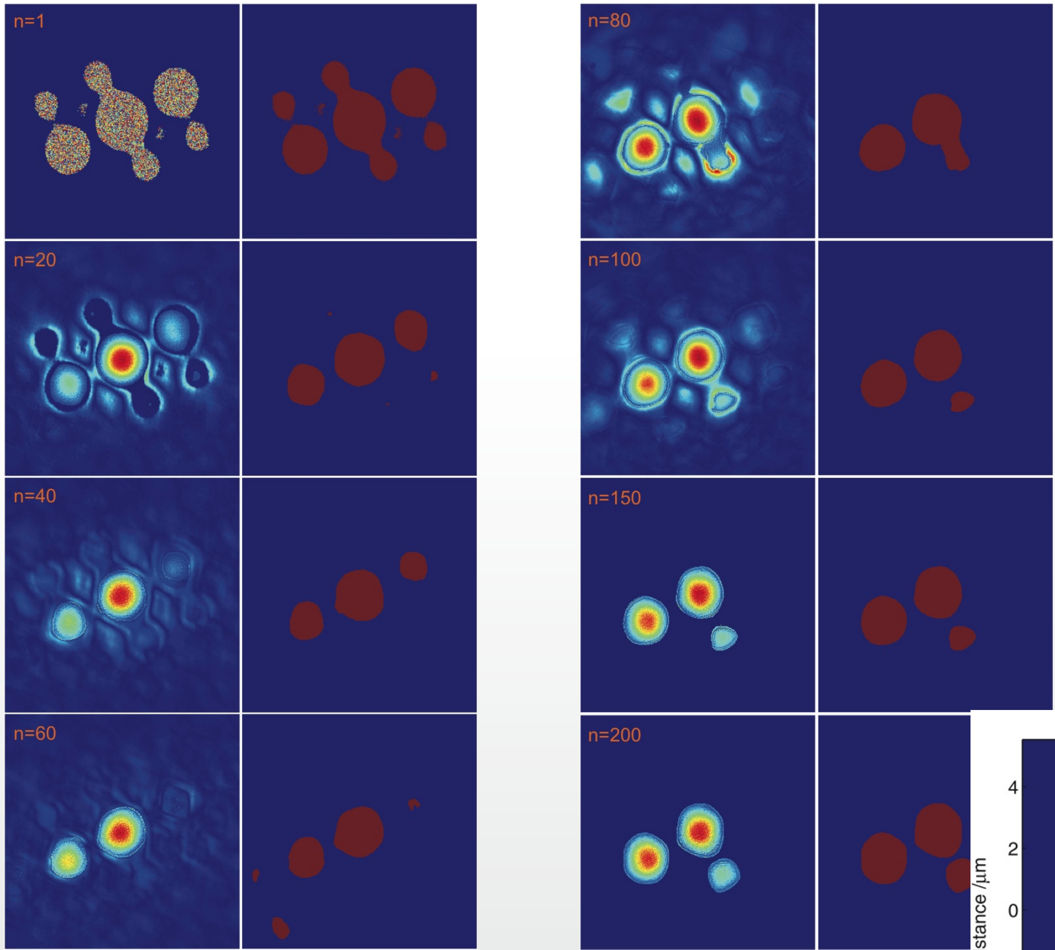
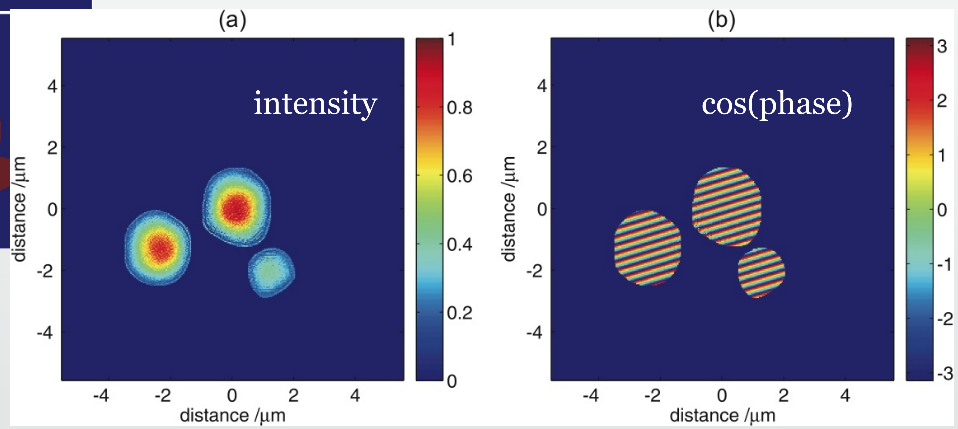


Evolution with number of iterations from 1-200

In each column, the left image is the retrieved intensity. The right image is the shrinkwrap mask

Final retrieved image:



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Summary & Future prospects

- High-field nonlinear optics can provide useful sources of ultrafast XUV/soft X-ray radiation
- Developing efficient sources requires detailed understanding of the generation process, and in particular of nonlinear propagation
- XUV/soft X-ray imaging with lab-based sources is a viable technique, and has great potential in many areas because of
    - Intrinsic high resolution & elemental contrast
    - Huge developments in phase retrieval algorithms and computer hardware
    - Relative ease of producing high energy ultrafast pulses
    - Availability of XUV/soft X-ray reflective optics for focusing

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People

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- Dr. Ben Mills, Dr. Ed Rogers, Dr. Matt Praeger, Dr Peter Horak, Prof Dave Hanna *(Optoelectronics Research Centre, Southampton)*
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