Sharp Images of Magnetization Phenomena Without Lenses

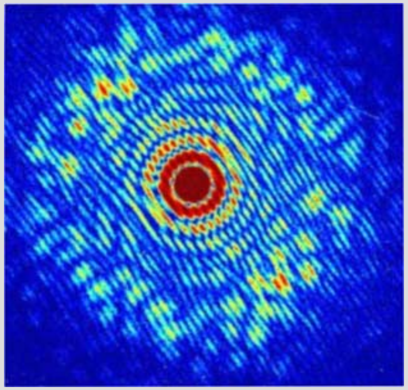
Jan Lüning

Laboratoire de Chimie Physique – Matière et Rayonnement
Université Pierre et Marie Curie (Paris)

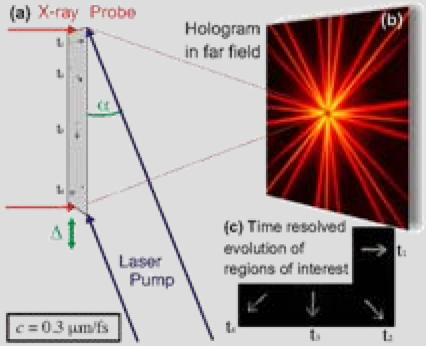
Synchrotron SOLEIL

Outline

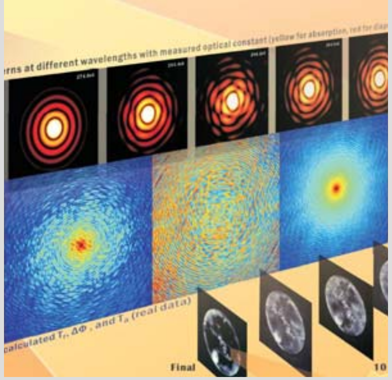
Fourier Transform Holography



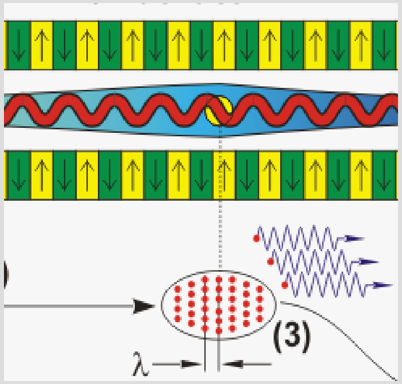
Unique Properties of FT Holography



New & Recent Developments



Free Electron X-ray Lasers



Collaborators

SSRL

Andreas Scherz
S. Roy
Jo Stöhr

DESY

Bill Schlotter
(U. Hamburg)

Ramon Rick
Diling Zhu
Mark Burkhardt
Benny Wu

BESSY

Christian Günther
Bastian Pfau
Stefan Eisebitt (TU Berlin)
Wolfgang Eberhardt

HITACHI
Inspire the Next

Olav Hellwig

Ian McNulty

LCP MR

W. Ma
B. Vodungbo
S. Chiuzbajan

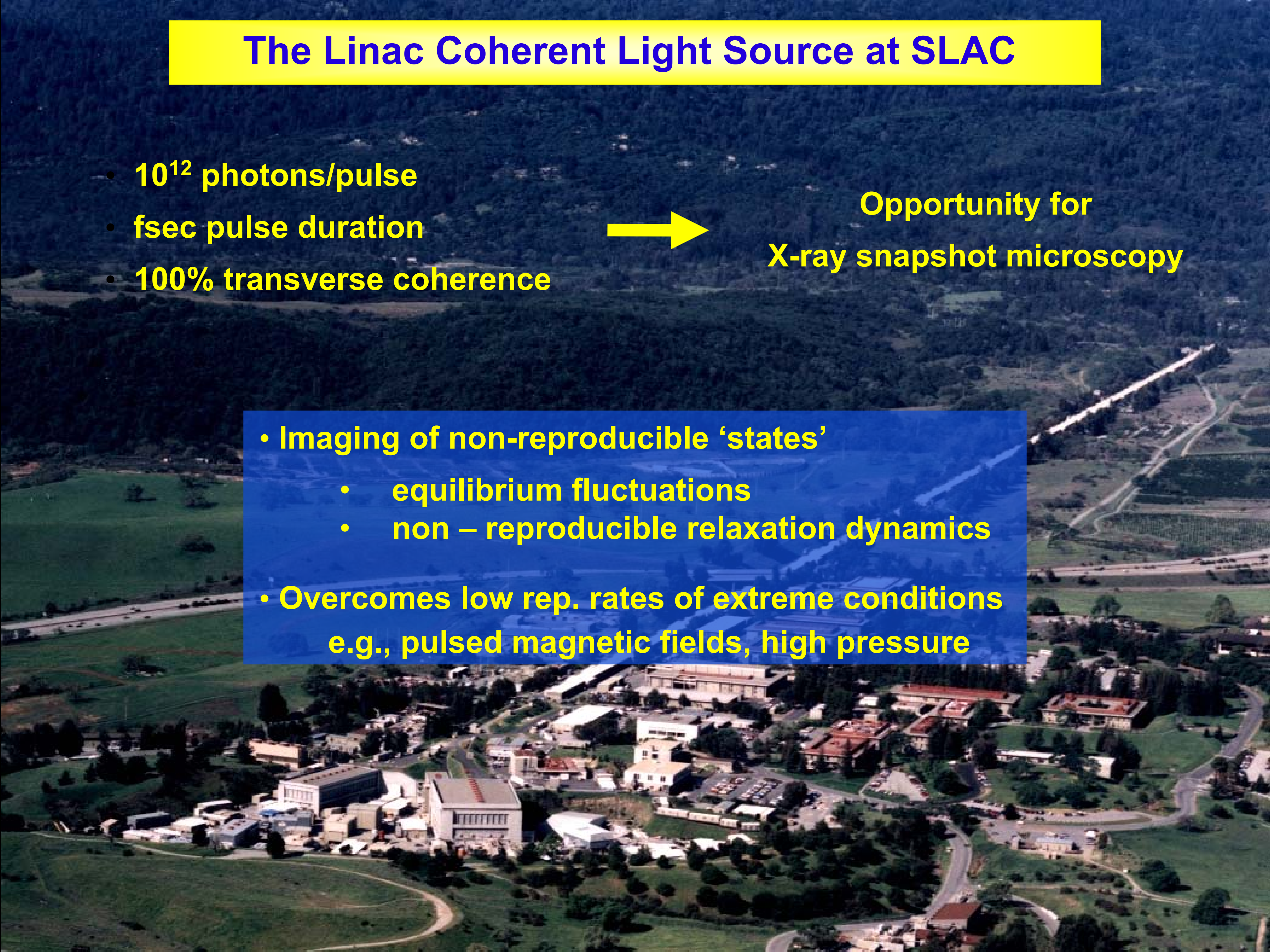
SOLEIL
SYNCHROTRON

Horia Popescu
Nicolas Jaouen
Maurizio Sacchi

CSNSM

F. Fortuna

The Linac Coherent Light Source at SLAC



- 10^{12} photons/pulse
- fsec pulse duration
- 100% transverse coherence

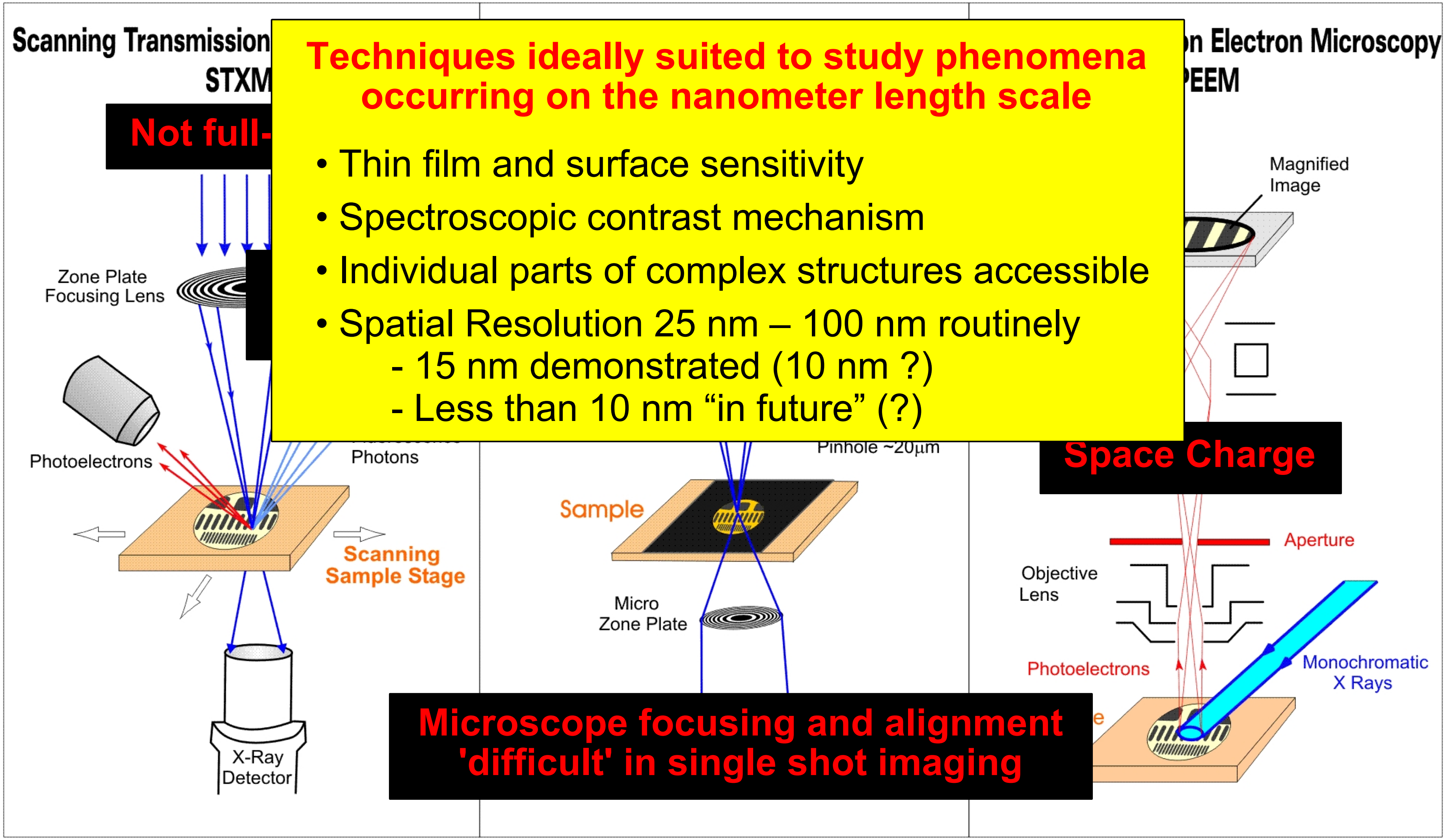
→

Opportunity for
X-ray snapshot microscopy

- Imaging of non-reproducible ‘states’
  - equilibrium fluctuations
  - non – reproducible relaxation dynamics
- Overcomes low rep. rates of extreme conditions
  e.g., pulsed magnetic fields, high pressure

X-ray spectromicroscopy techniques

Quantitative imaging with sensitivity to
elemental and chemical distribution and charge/spin ordering



Not full-

Techniques ideally suited to study phenomena occurring on the nanometer length scale

- Thin film and surface sensitivity
- Spectroscopic contrast mechanism
- Individual parts of complex structures accessible
- Spatial Resolution 25 nm – 100 nm routinely
    - 15 nm demonstrated (10 nm ?)
    - Less than 10 nm “in future” (?)

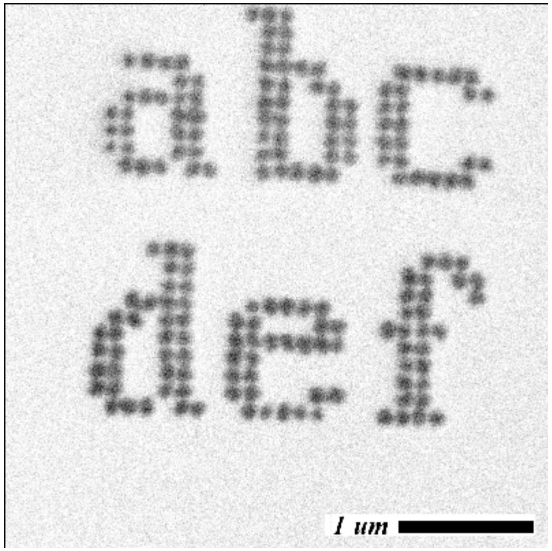
Space Charge

Microscope focusing and alignment 'difficult' in single shot imaging

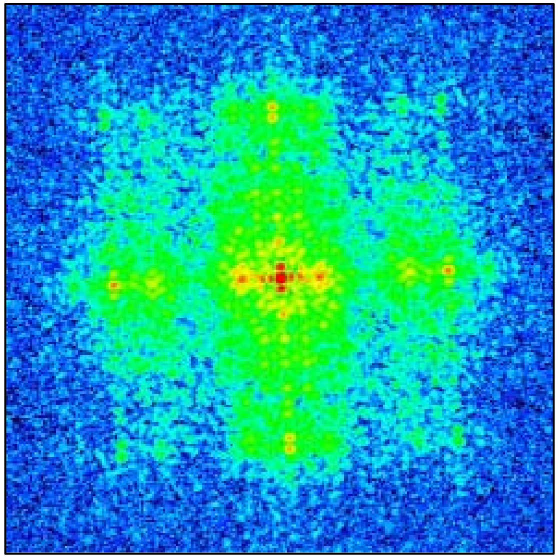
Demonstration of X-ray microscopy by iterative phase retrieval

J. Miao, P. Charalambous, J. Kirz, D. Sayre, Nature **400**, 342 (1999)

Real object (SEM picture)



Speckle

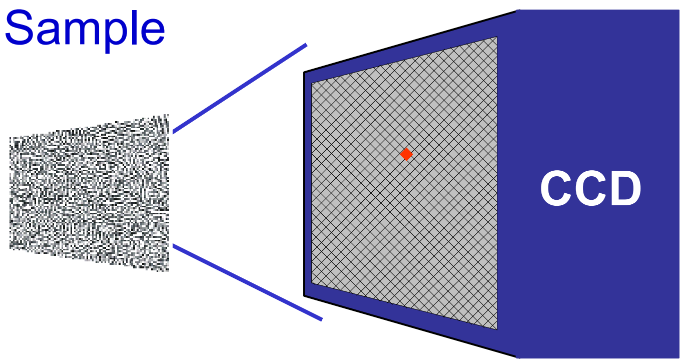


Reconstructed object

75 nm resolution using 730 eV (1.7 nm) X-rays

- Spatial resolution of 8 nm (2D) & 50 nm (3D), J. Miao et al., Phys. Rev. Lett. **89**, 088303 (2002)
- Reconstruction of a yeast cell (30 nm), Pierre Thibault et al., Acta Cryst. **A62**, 248 (2006)

Lens-less microscopy – Fourier space imaging



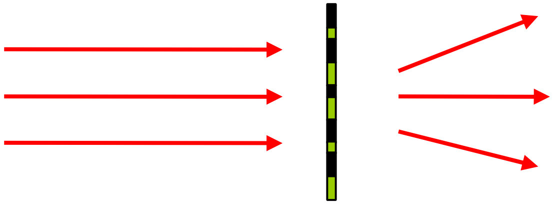
$$D_{p,q} = \sum_{i,j} S_{i,j} \cdot e^{-i \cdot 2\pi \frac{d([i,j] \rightarrow [p,q])}{\lambda}}$$

Idea:

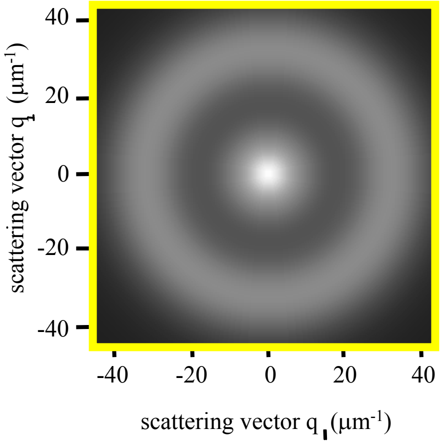
Replace lens with a two-dimensional detector to record scattered radiation in Fourier space

Potential for wavelength limited resolution: Highest detected momentum transfer (Fourier component) defines spatial resolution.

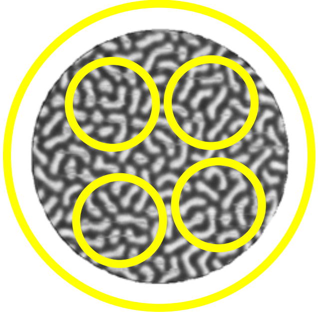
Incoherent versus coherent small angle X-ray scattering



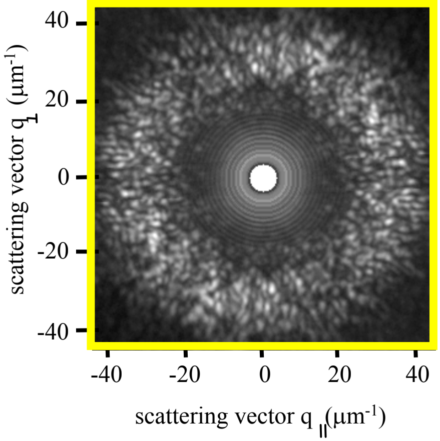
SAXS



Sample without long range order



Speckle



- Coherence length smaller than illuminated area.
- Incoherent superposition of ‘local’ speckle images.

Information about sample statistics

- Coherence length larger than illuminated area.
- Scattering from all ‘objects’ adds up coherently.

Sample specific information preserved

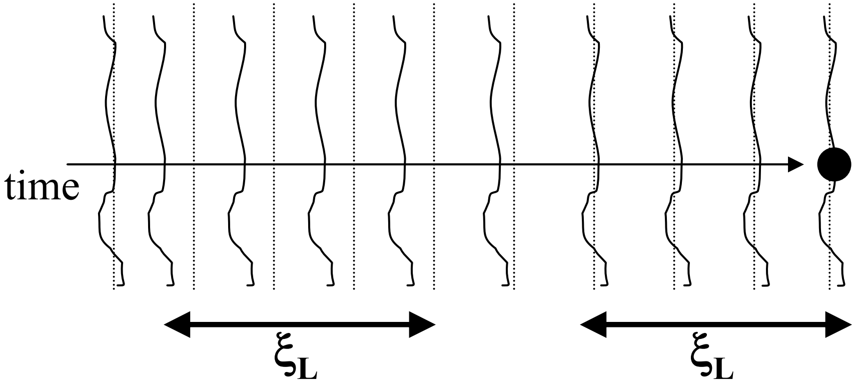
Coherence Lengths

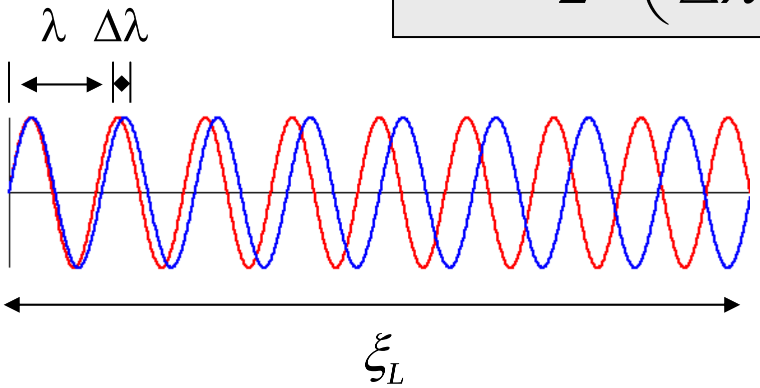
Coherence required to observe interference effects: Amplification and Extinction

Temporal / Longitudinal coherence

$$\xi_L = \frac{\lambda}{2}\cdot\left(\frac{\lambda}{\Delta\lambda}\right)$$

Michelson Interferometer:
How long can path difference be?

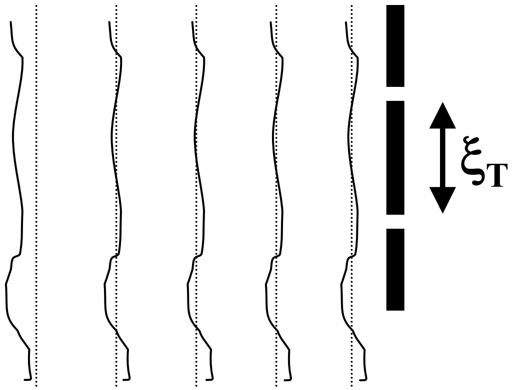


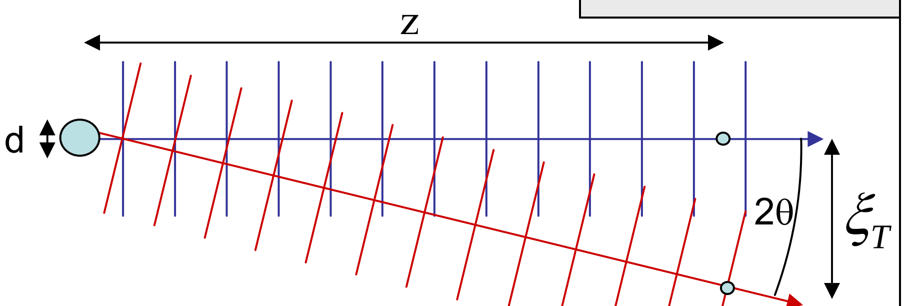


Transverse coherence

$$\xi_T = \frac{z\,\lambda}{2\,\pi\,d}$$

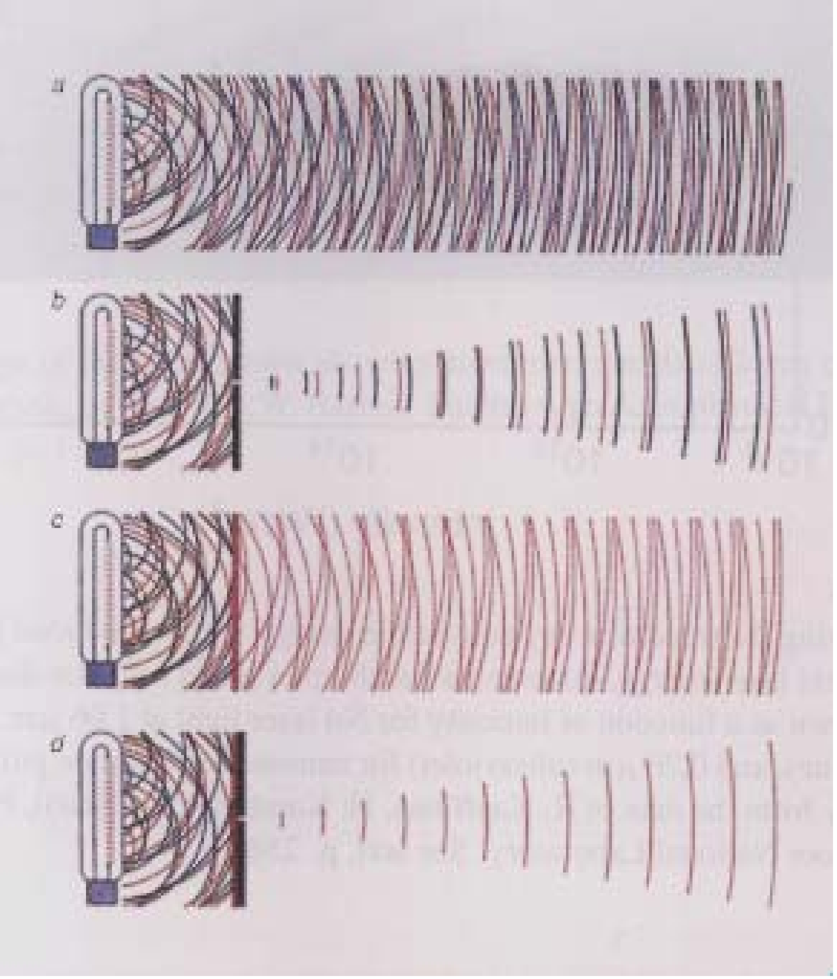
Young’s double slit:
How far apart can slits be?





$$d\cdot\theta = \lambda/2\pi$$

How to make a Photon Beam Coherent?



A. Schawlow, Sci. Am. **219**, 120 (Sept 1968)

Synchrotron sources are polychromatic and incoherent/chaotic

Pinhole:
spatial filtering → lateral coherence

Monochromator:
spectral filtering → longitudinal coherence

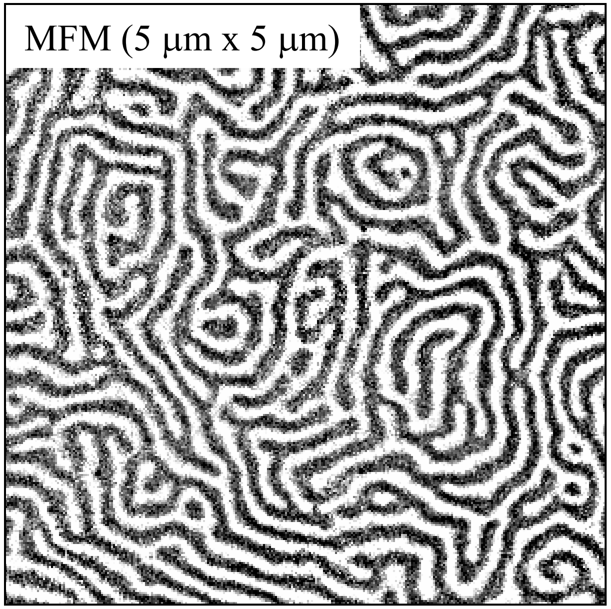
Spatially and spectrally filtered beam
is coherent

Coherence ~ Brilliance $\cdot \lambda^3$

$\Rightarrow$ Undulator source allows to extract a 'sufficient' coherent flux

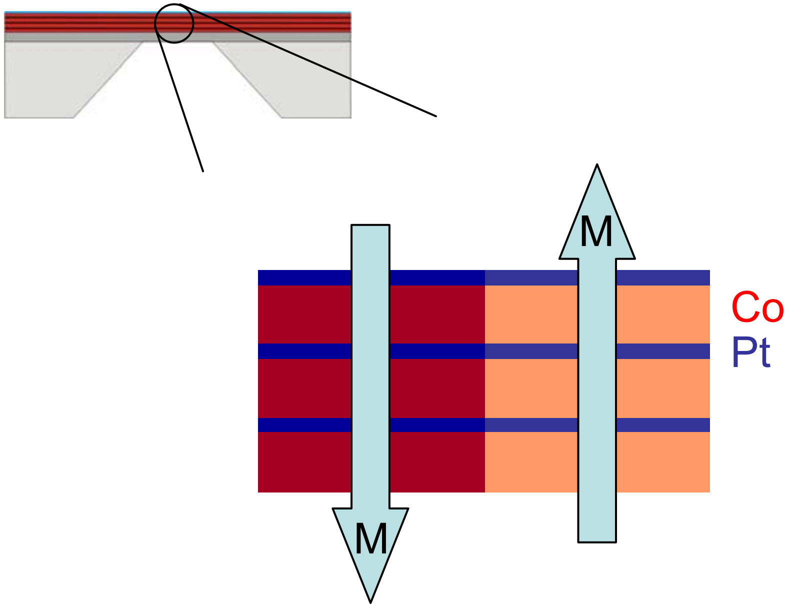
Magnetic Fruit-Fly – the worm domain sample

Top view



Magnetic test medium for
resonant coherent
x-ray scattering

Side view

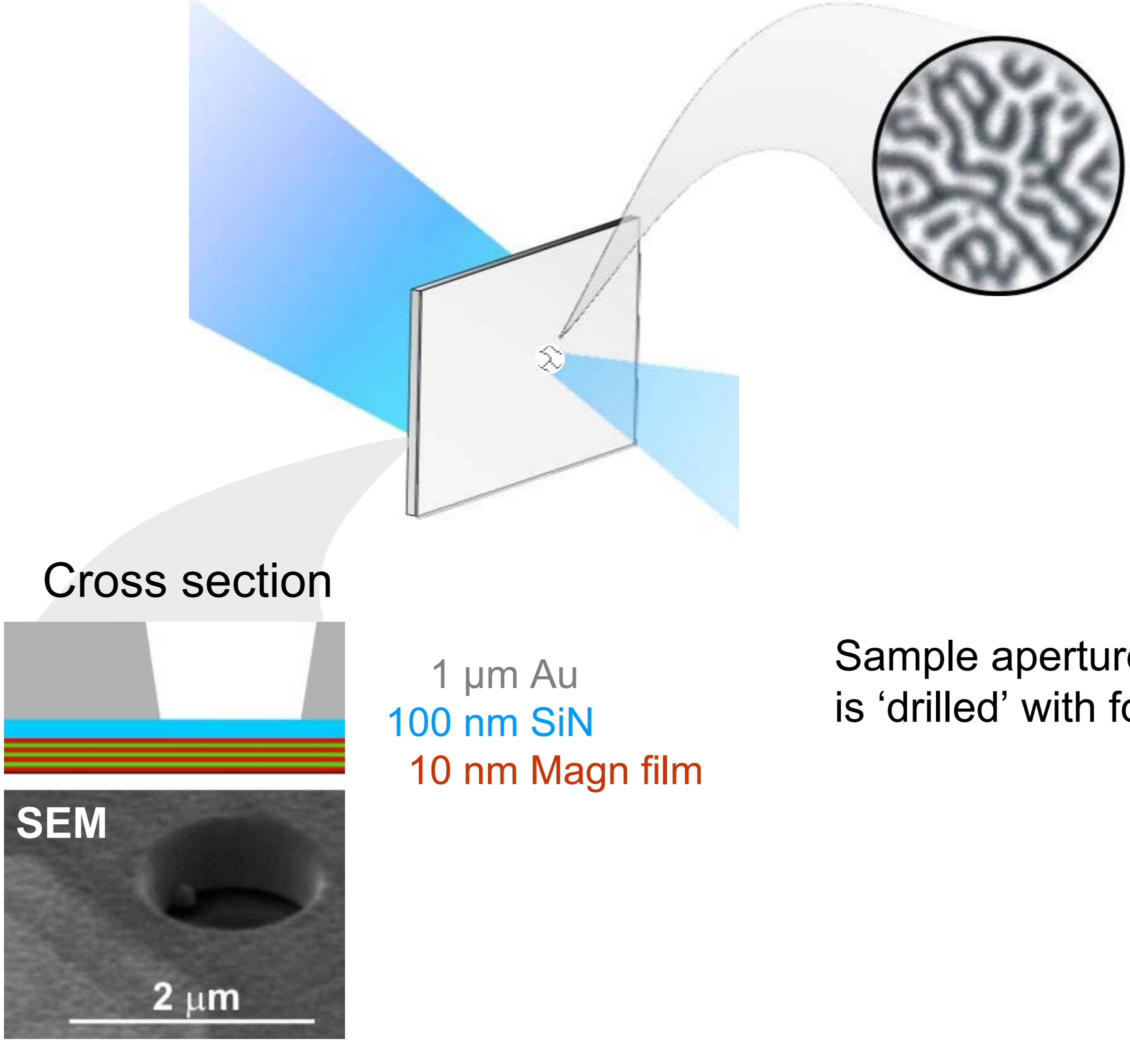


Sample: E. Fullerton, O. Hellwig
J.-U. Thiele, IBM/Hitachi Almaden

SiN_x(160 nm) / Pt (20 nm) /
[Co (3 nm) / Pt (0.7 nm)]$_{50}$ /
Pt (2 nm)

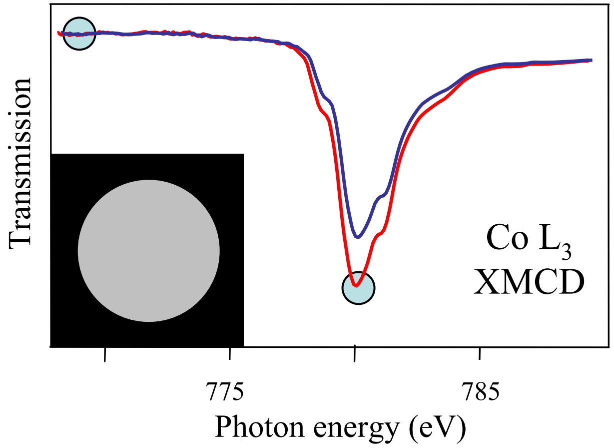
perpendicular magnetization

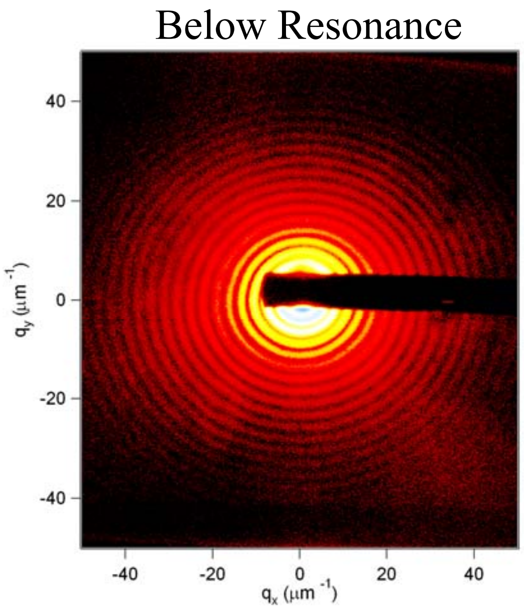
Experimental geometry

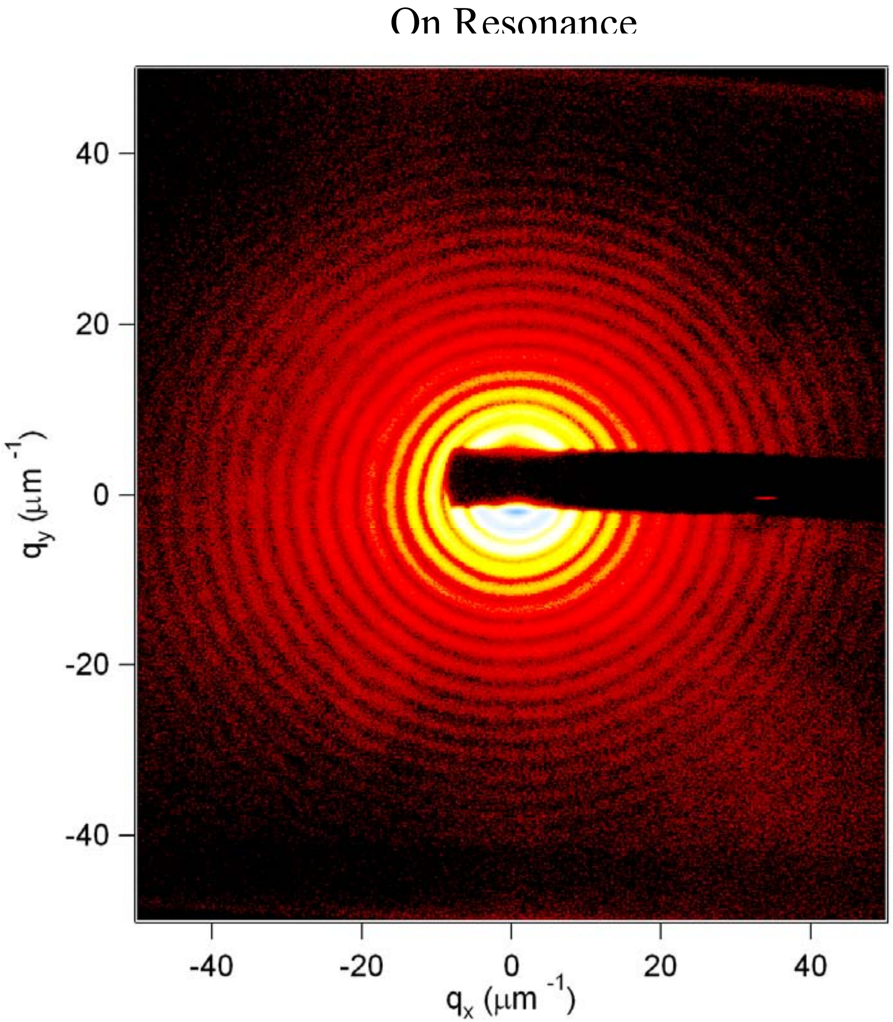


Sample aperture in X-ray opaque Au film is ‘drilled’ with focused ion beam

Magnetic Scattering Contrast



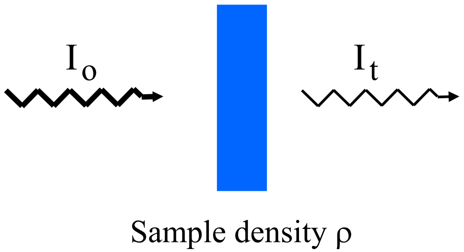




λ = 1.59 nm, 2.5 mm ∅ Pinhole
fully coherent illumination: visibility = 1, M = 1

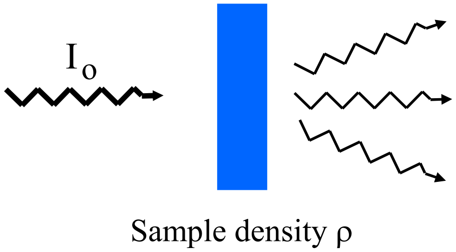
X-ray Magnetic Circular Dichroism in Absorption / Scattering

Absorption



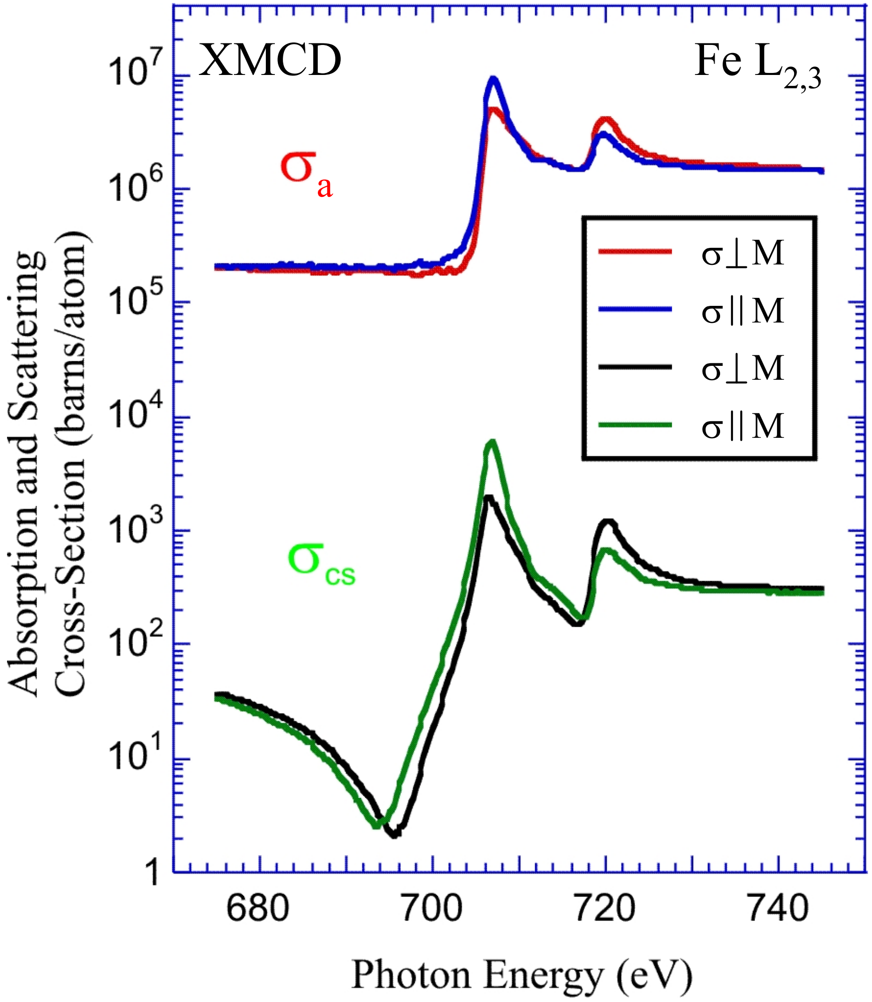
$$I_t = I_o e^{-\sigma_a \rho t}$$

Small Angle Scattering



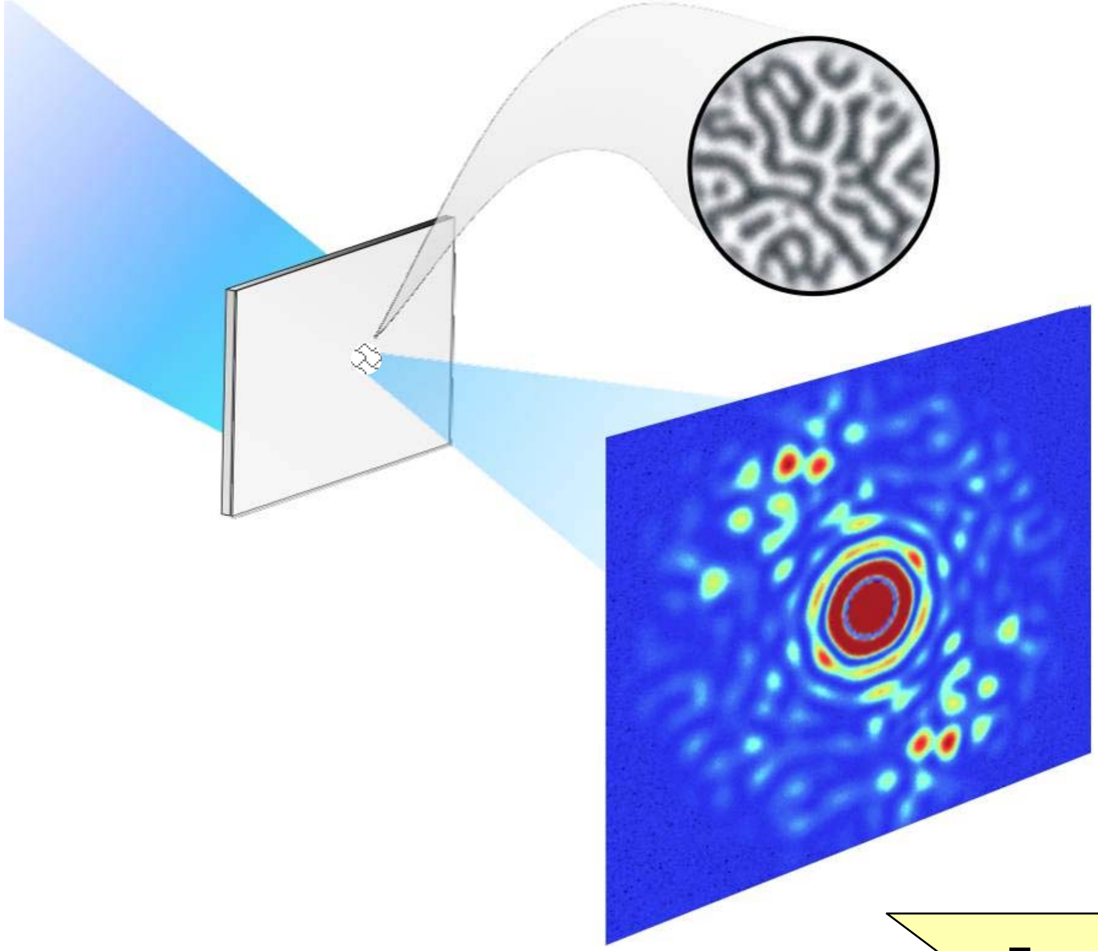
$$I_{cs} = I_o \sigma_{cs} / \sigma_a$$

$$\sigma_{cs} = c \mid f_1 + i f_2 \mid$$



Data from Jeff Kortright (LBNL)

Phase problem in X-ray scattering

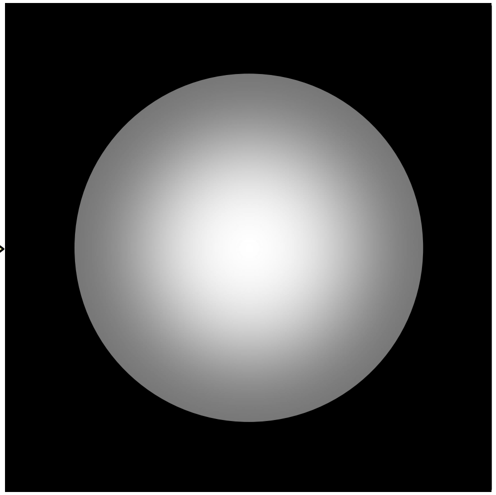


Scattering amplitude is complex, but only intensities are detected

$$I_{p,q} = \left| M_{p,q} \cdot e^{-i\phi_{p,q}} \right|^2$$

Fourier Transform

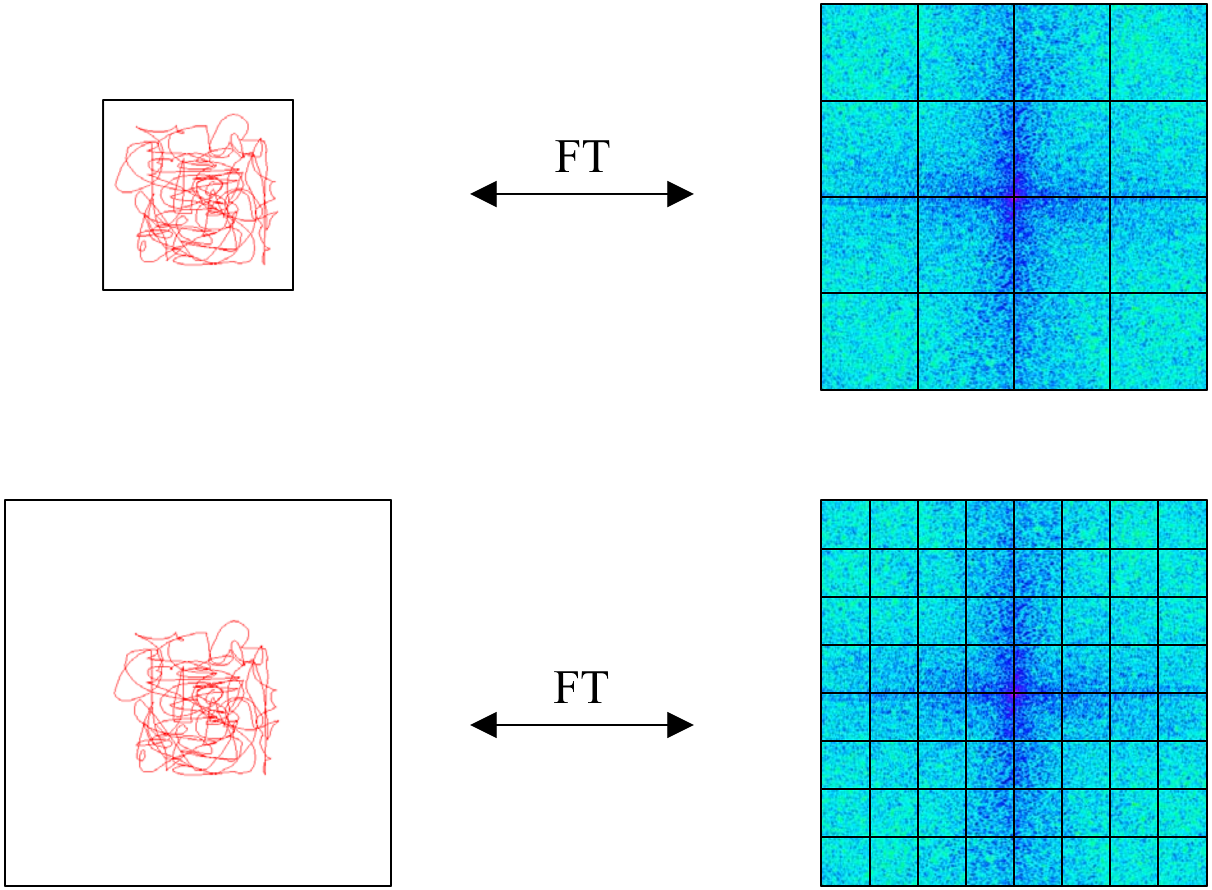
Auto-correlation



Convolution theorem applied to diffraction

$$(a \otimes a) = FT^{-1}\{FT(a) \cdot FT(a)\}$$

Oversampling ‘compensates’ lost phase information



N x N

FT

N x N

> √2N x √2N

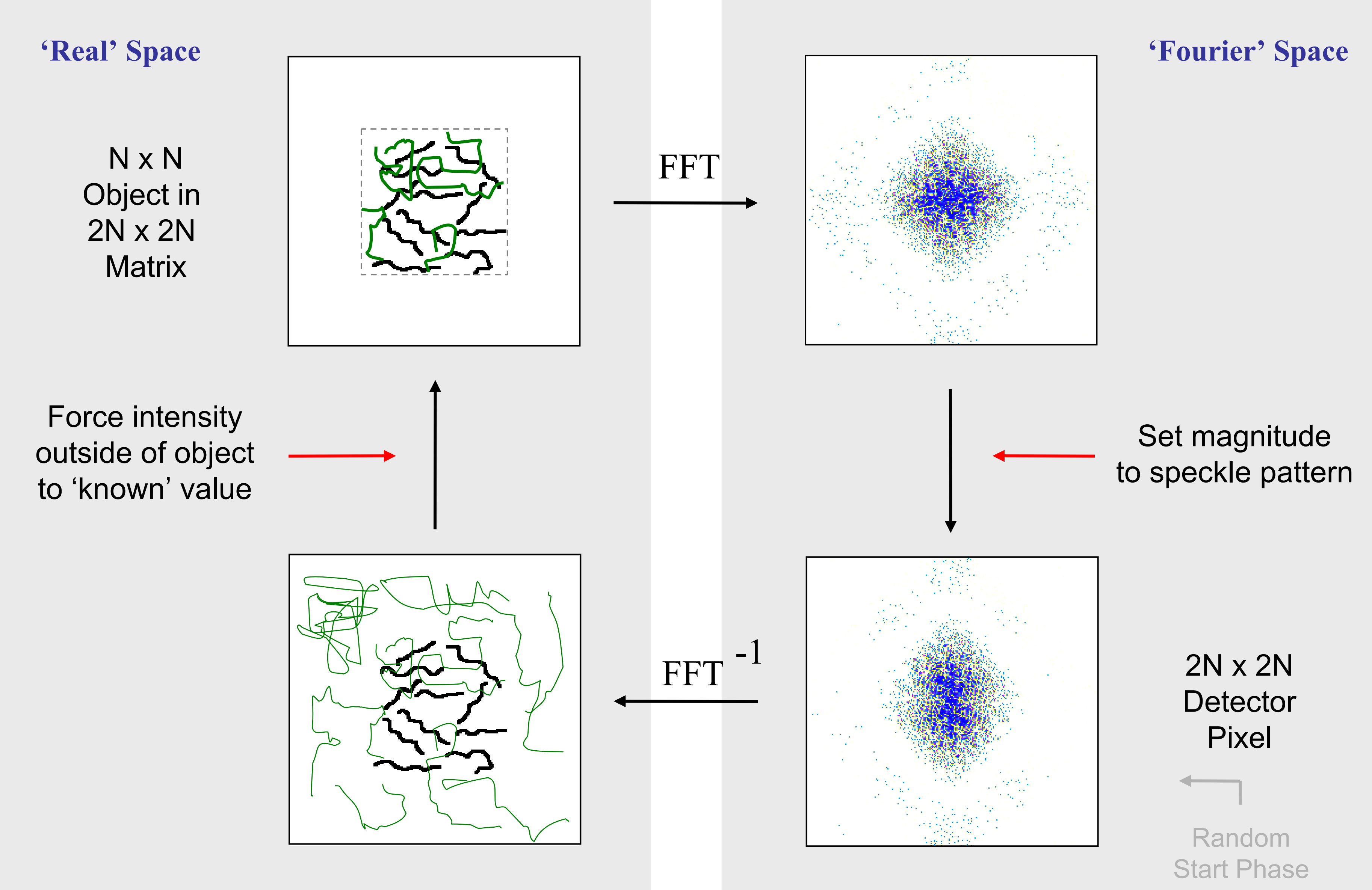
FT

> √2N x √2N

Phase reconstruction through iterative algorithm

Iterative algorithm for phase reconstruction

Algorithm idea and method developed by Sayre, Gerchberg & Saxton, Bates, Fienup, …



Algorithm explained for example by J. Miao et al, Phys. Rev. B 67, 174104 (2003)

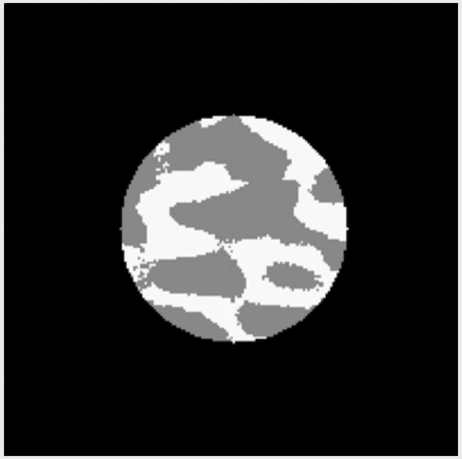
How important are the scattering phases?

$A = M \cdot e^{i\Phi}$ Experiments are intensity measurements, phase information is lost

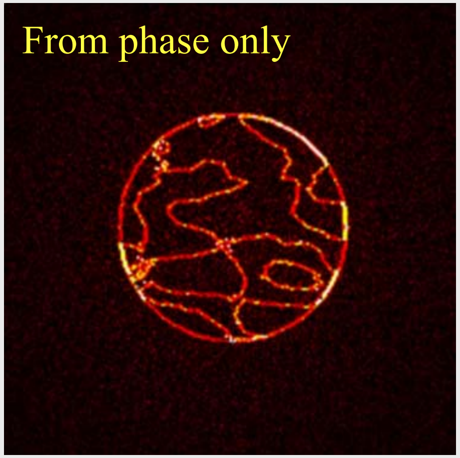
Real Space

Fourier Space

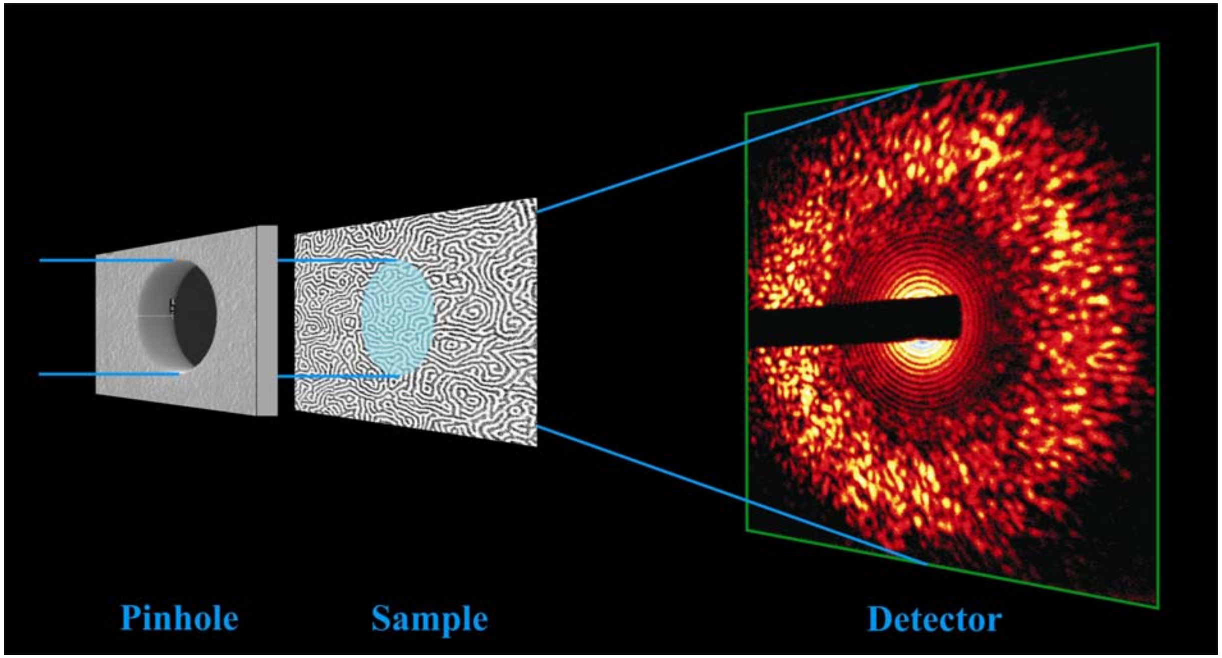
Magn.



Phase



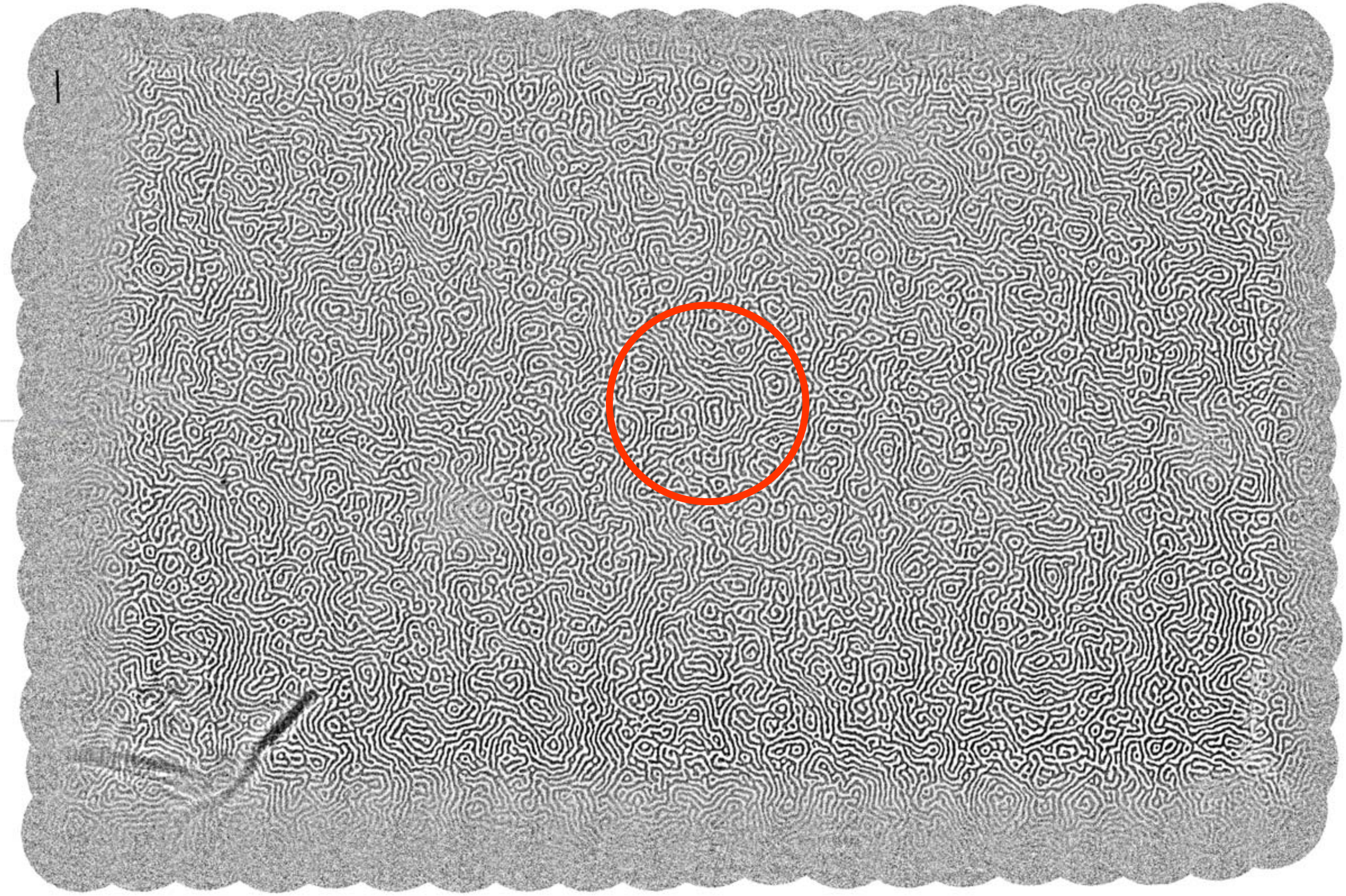
Reconstruction of Real Space Image



Phase problem can be solved by “oversampling” speckle image

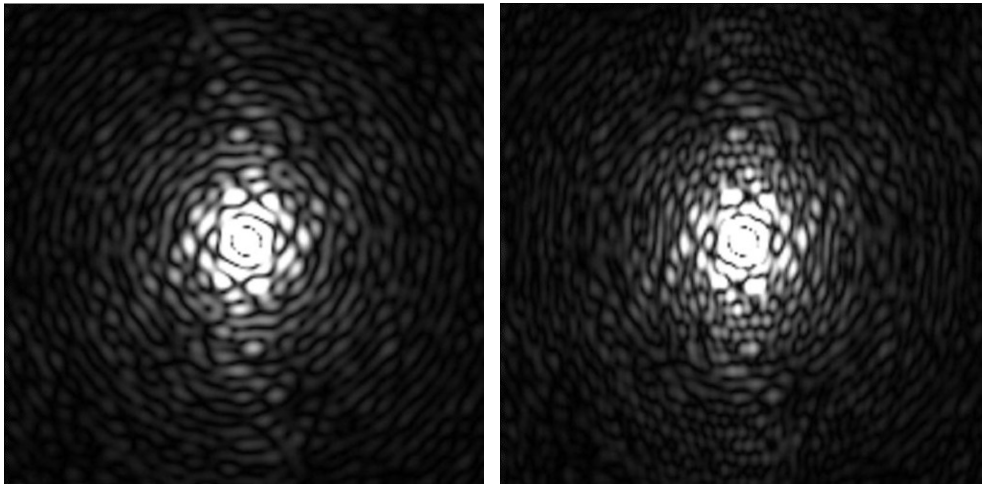
Which worms did we ‘image’?

Stitched TXM image of entire sample membrane
(P. Fischer, ALS)



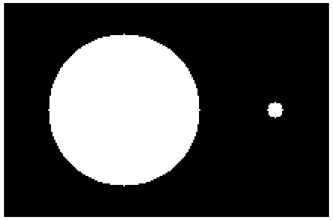
How to obtain ‘*more*’ information?

Reference Beam / Fourier Transform Holography



No reference hole

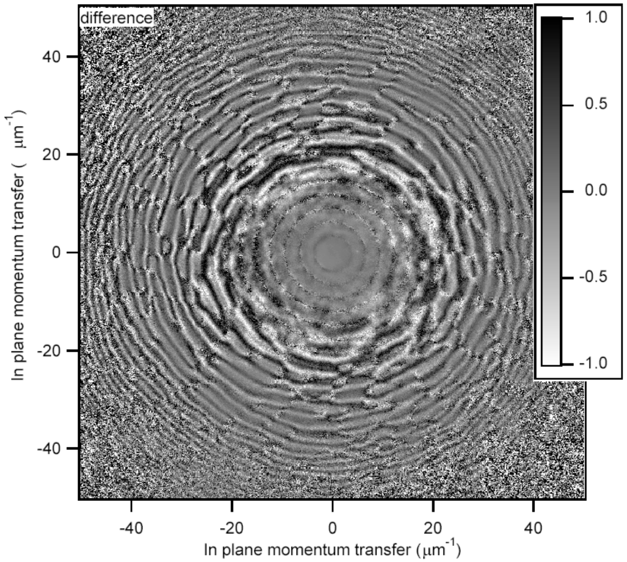
With reference hole

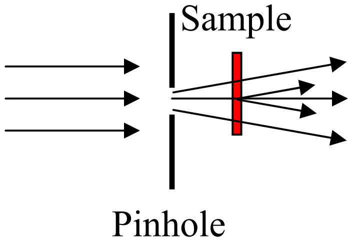


H. He et al., PRB **67**, 174114 (2003)

In-line Holography

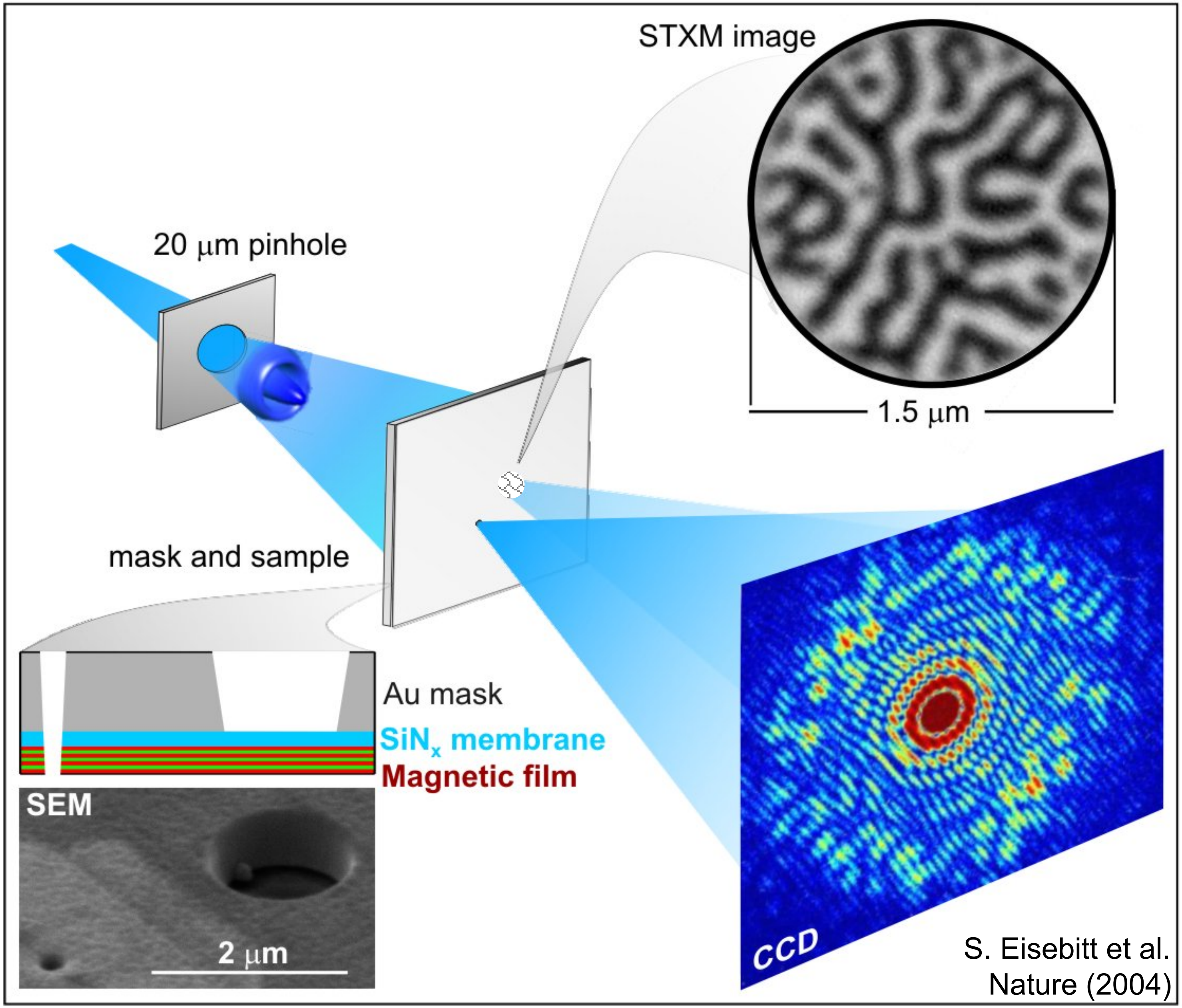
RCP – LCP “Dichroism”





S. Eisebitt et al., PRB **68**, 104419 (2003)

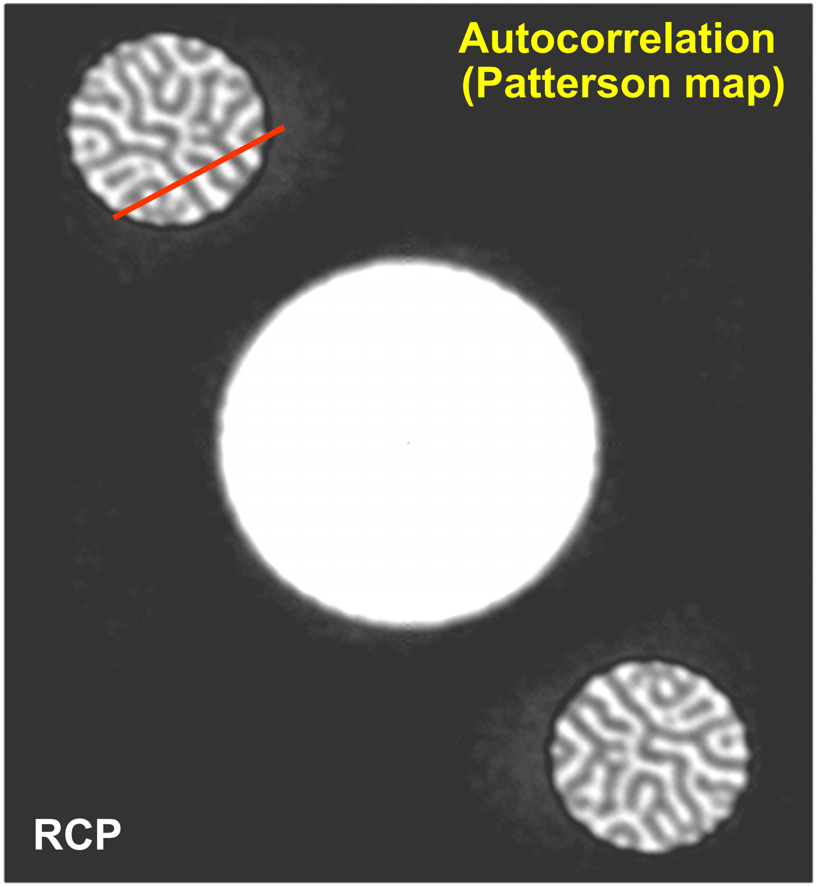
Fourier transform X-ray spectro-holography



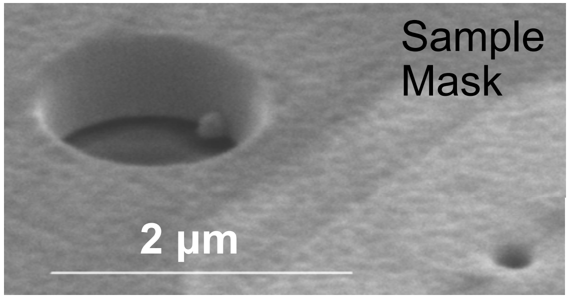
S. Eisebitt et al.
Nature (2004)

Digital image reconstruction

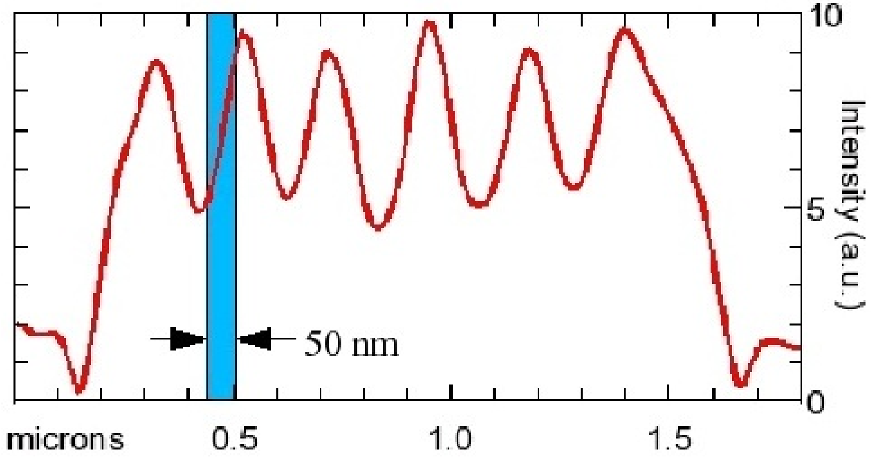
Single Fourier transformation of scattering intensities yields the auto-correlation of sample, which contains image of sample due to the off-axis geometry in FT holography. (convolution theorem)



Intensity in image center, which contains self-correlation of apertures, is truncated.



10% - 90% intensity rise over about 50 nm



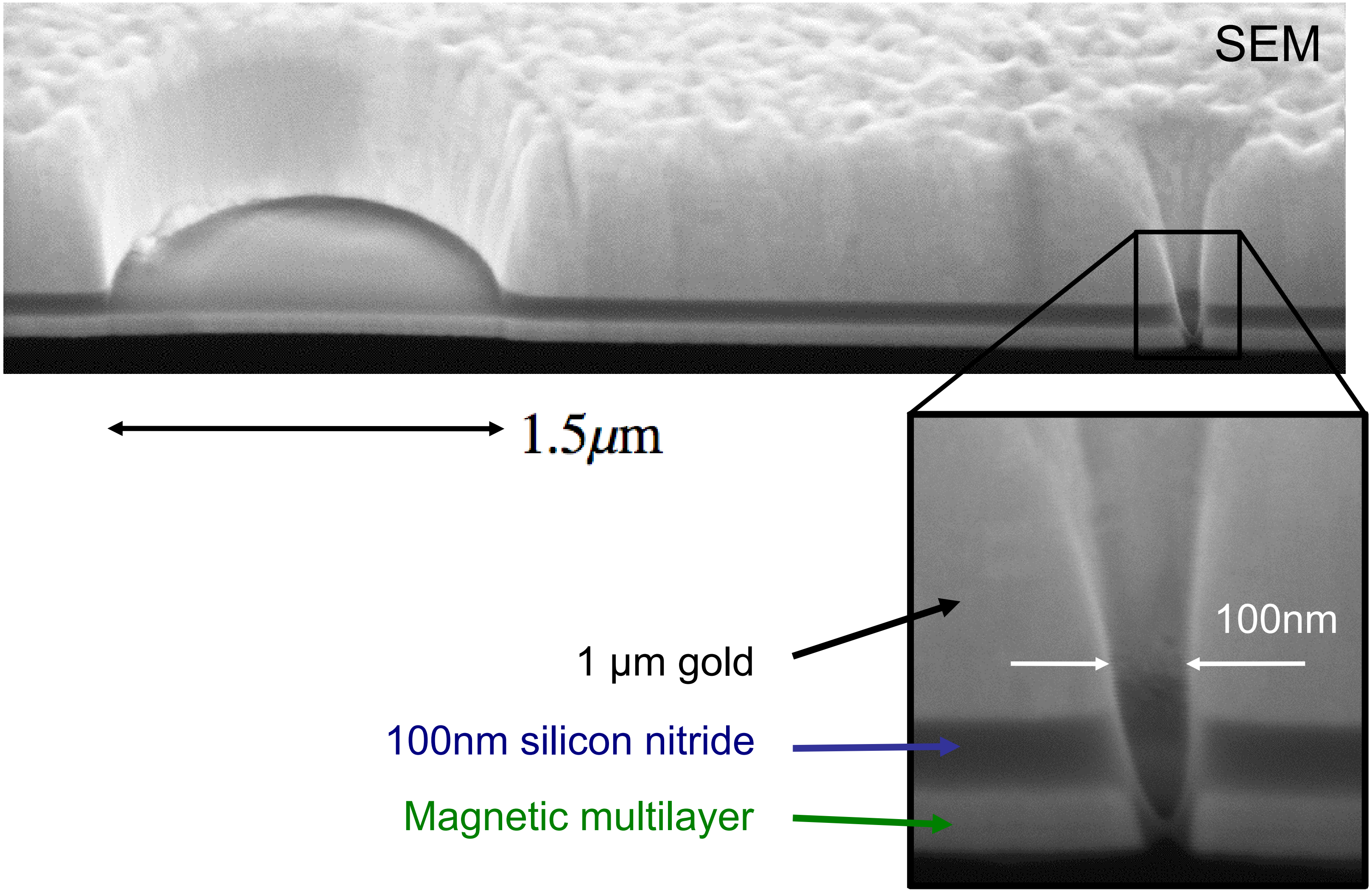
Key properties of Fourier transform X-ray holography

- ***True imaging technique***
- ***Wavelength limited spatial resolution***

Spatial resolution in FT hologram given by effective size of reference beam source.

Integrated mask sample structure

Patterned with focused ion beam



1 µm gold

100nm silicon nitride

Magnetic multilayer

Key properties of Fourier transform X-ray holography

- ***True imaging technique***

- ***Wavelength limited spatial resolution***
  Deconvolution and phase retrieval algorithm

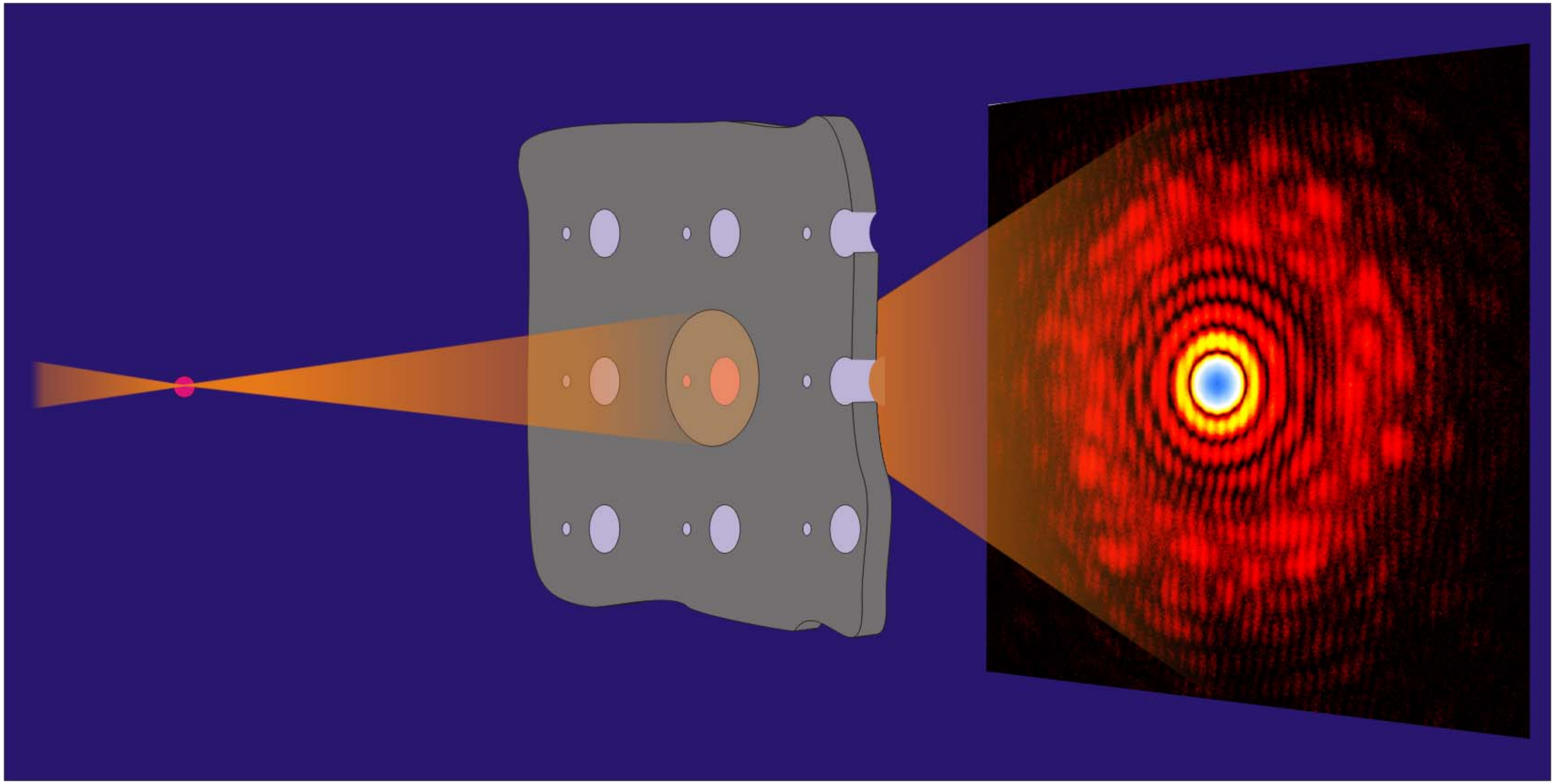
- ***Simple and rather 'cheap' setup***

- ***Nanometer resolution with micron stability***
  Setup is basically insensitive to vibrations
  or thermal drifts

- ***Ideally suited for in-situ studies***
  No space constraints around sample

- ***Ideally suited for sample arrays***
  No alignment or focusing required

- ***Wide applicability***
  Samples can be grown or placed in aperture or
  on back of mask or placed separately behind it.
  Reflection geometry may be possible.



Key properties of Fourier transform X-ray holography

General properties of
coherent diffraction microscopy

- ***Wavelength limited spatial resolution***

- ***Simple and rather 'cheap' setup***

- ***Nanometer resolution with micron stability***
  Setup is basically insensitive to vibrations or thermal drifts

- ***Ideally suited for in-situ studies***
  No space constraints around sample

- ***Ideally suited for sample arrays***
  No alignment or focusing required

- ***Wide applicability***
  Samples can be grown or placed in aperture or on back of mask or placed separately behind it.

- ***Reflection geometry***
  may be possible

Exploring FT holography

Obtaining a sample’s autocorrelation yields for microscopy ‘unusual’ capabilities. *SIMULTANEOUSLY:*

- real and imaginary part of the refractive index are recorded
  → resonant phase contrast imaging (A. Scherz et al., PRB 07)

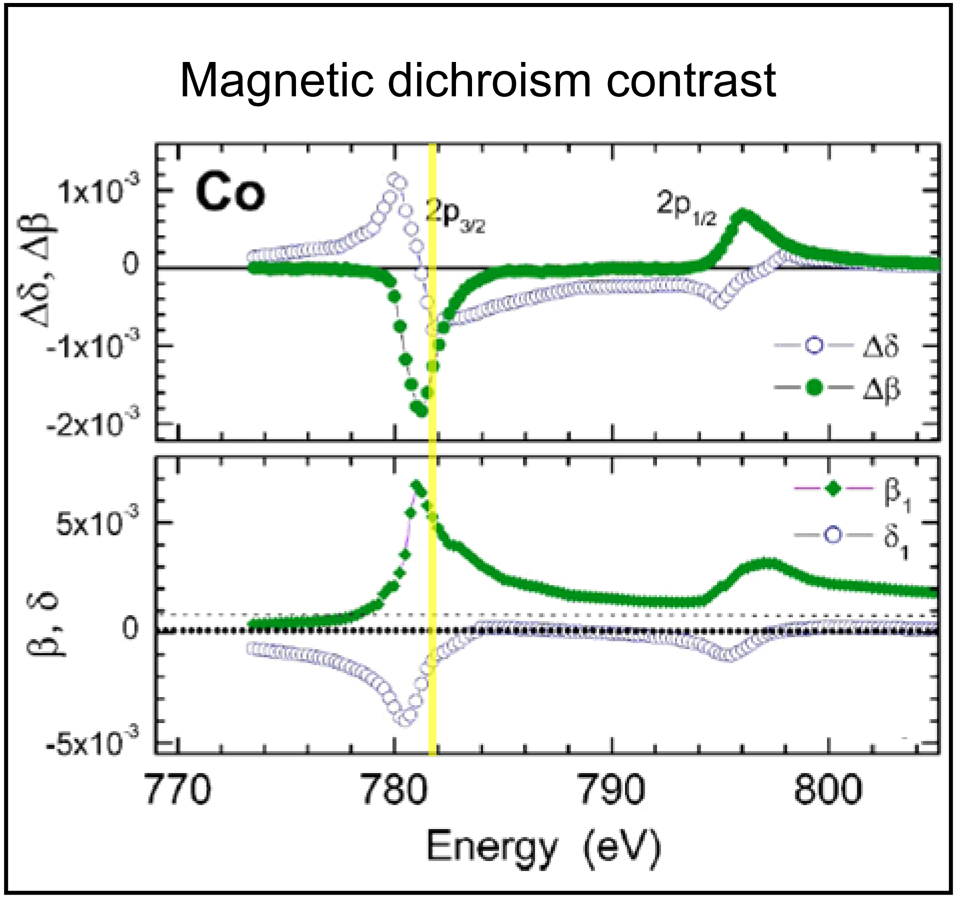
- multiple images can be recorded without increasing the radiation dose absorbed by the sample
  → increased image quality (B. Schlotter et al., APL 06)

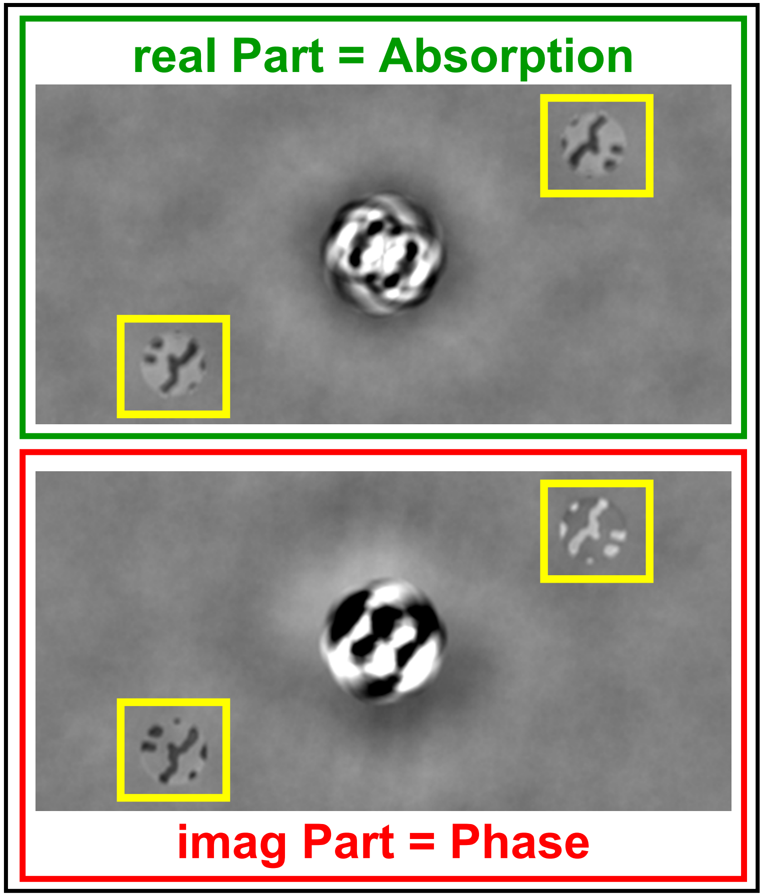
- spatially distributed sample areas can be imaged with high resolution
  → imaging with femtosecond time resolution (B. Schlotter et al., OL 07)

Absorption and phase contrast imaging in FTH

$n = 1 - \delta + i\beta$

Refractive index is complex, hence, the autocorrelation is complex!


Magnetic dichroism contrast


real Part = Absorption

imag Part = Phase

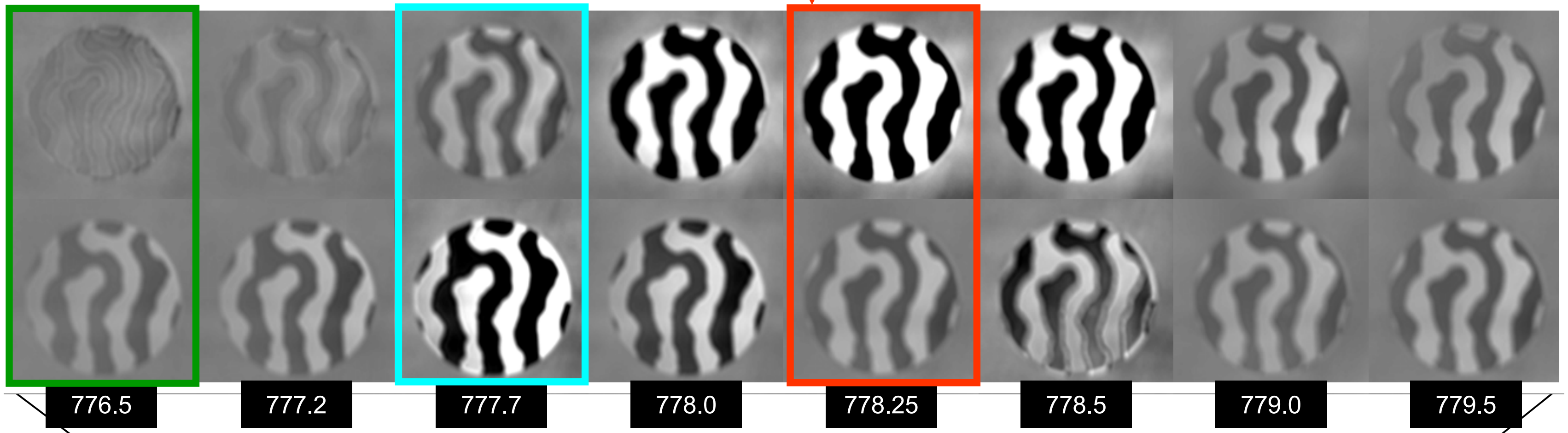
Data of β & δ from C. Mertens (BESSY)

A. Scherz et al., PRB 07

Energy dependence of scattering contrast

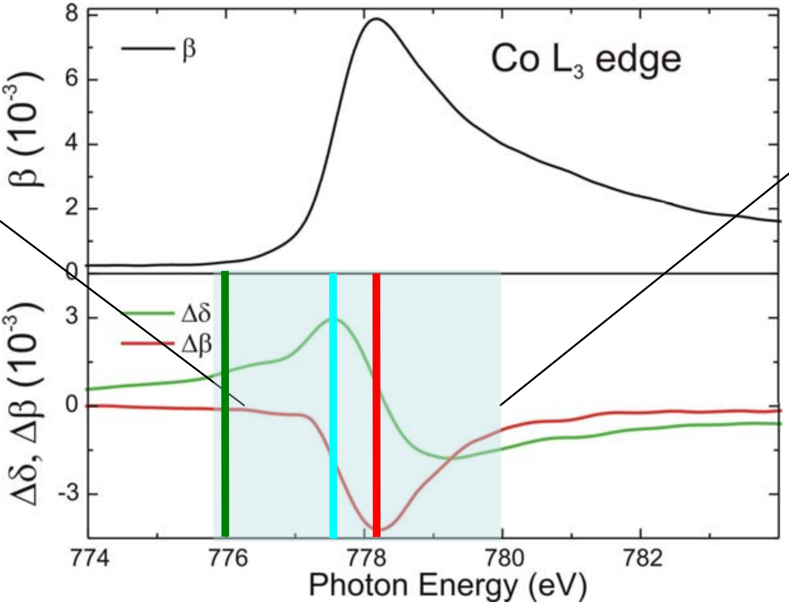
Solely absorption contrast

REAL (n) = Absorption



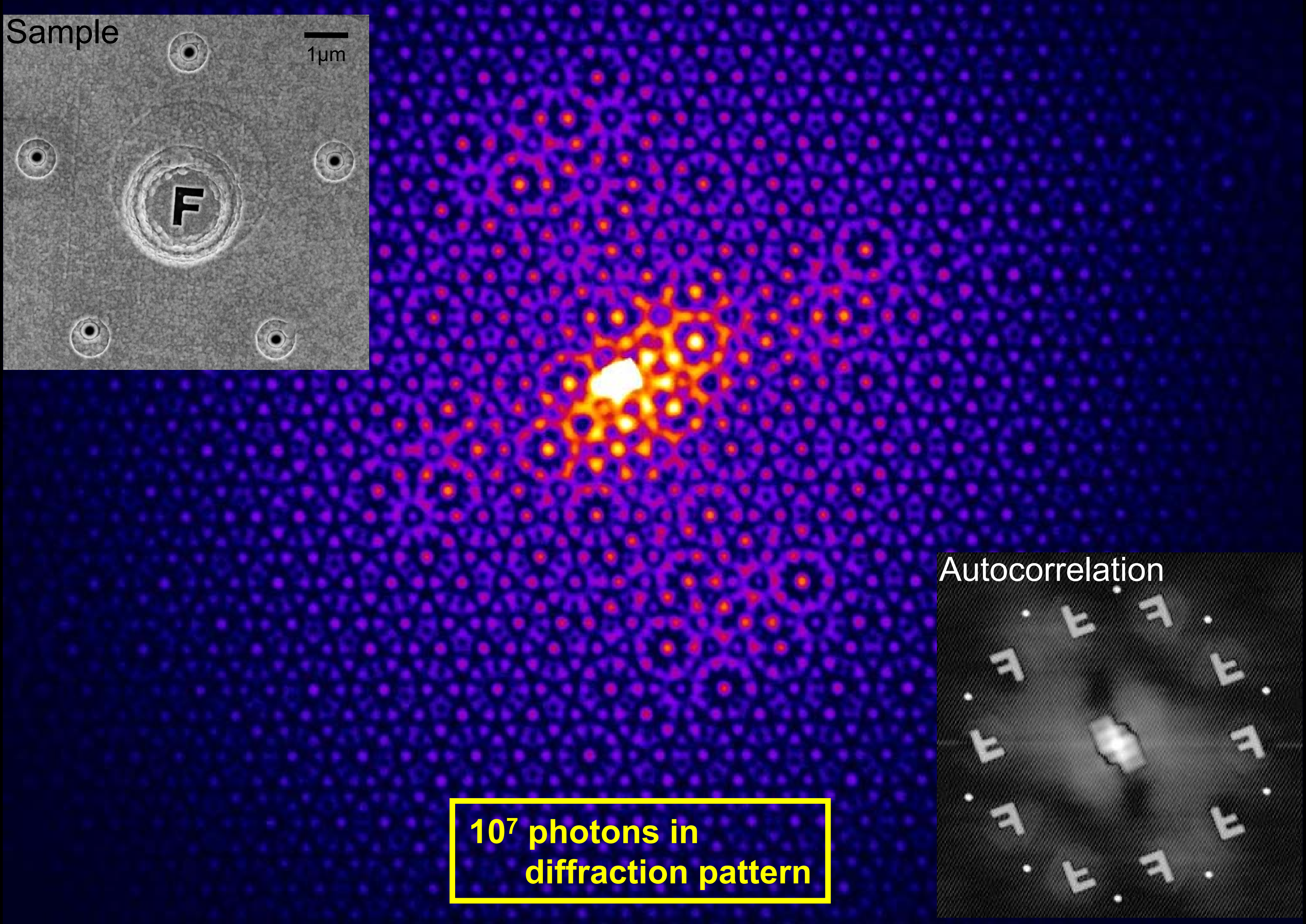
Solely phase contrast

Imag (n) = Phase



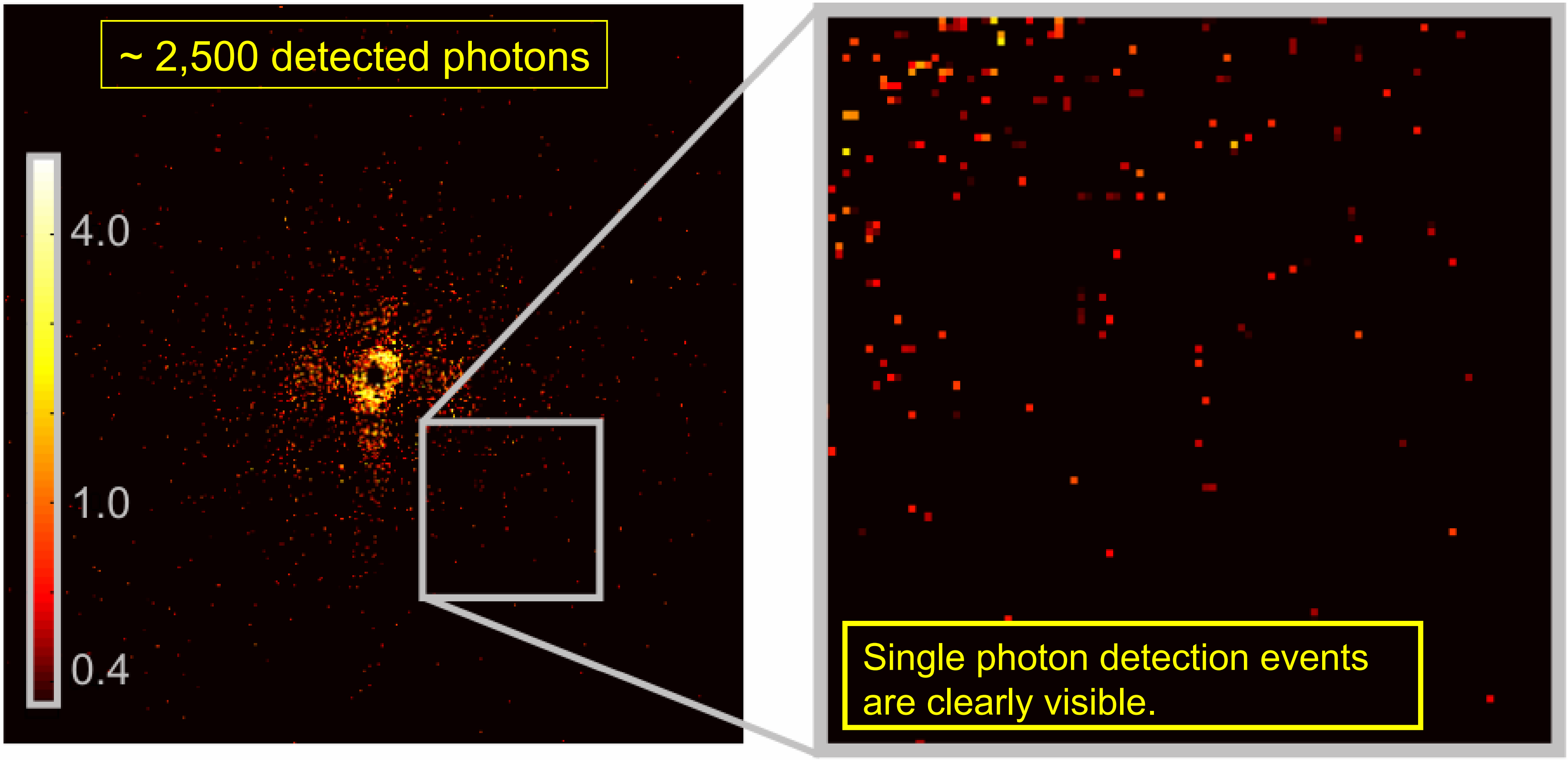
A. Scherz et al., PRB 07

Multiple reference FT holography



10^7 photons in diffraction pattern

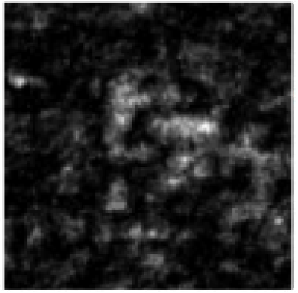
Weak Illumination Fourier Transform Holography



B. Schlotter et al., APL 06

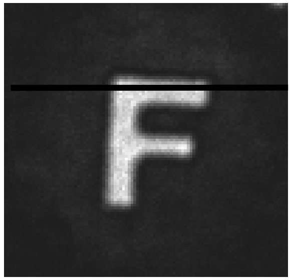
Weak Illumination Fourier Transform Holography

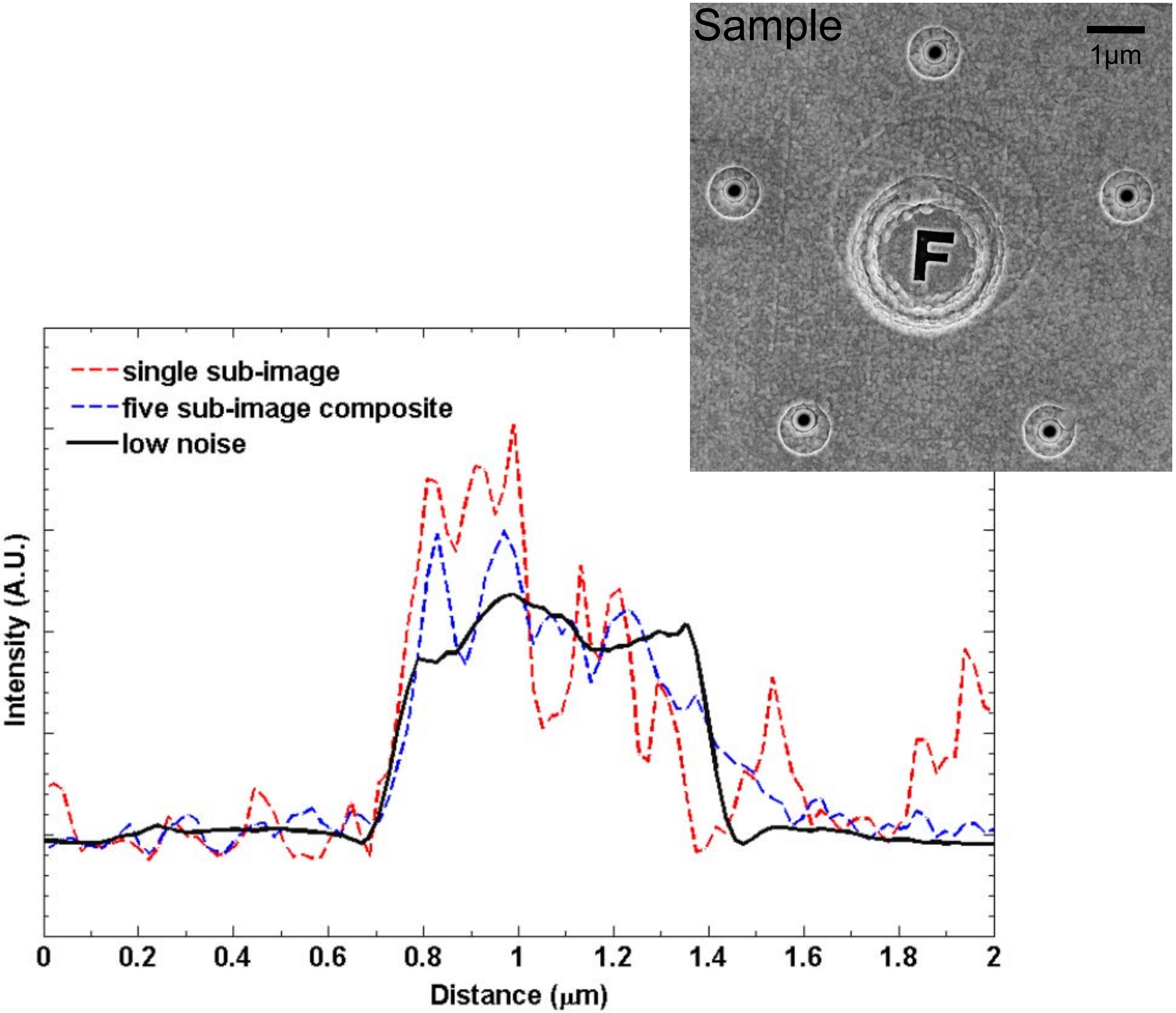
Single sub-image



5 sub-image composite

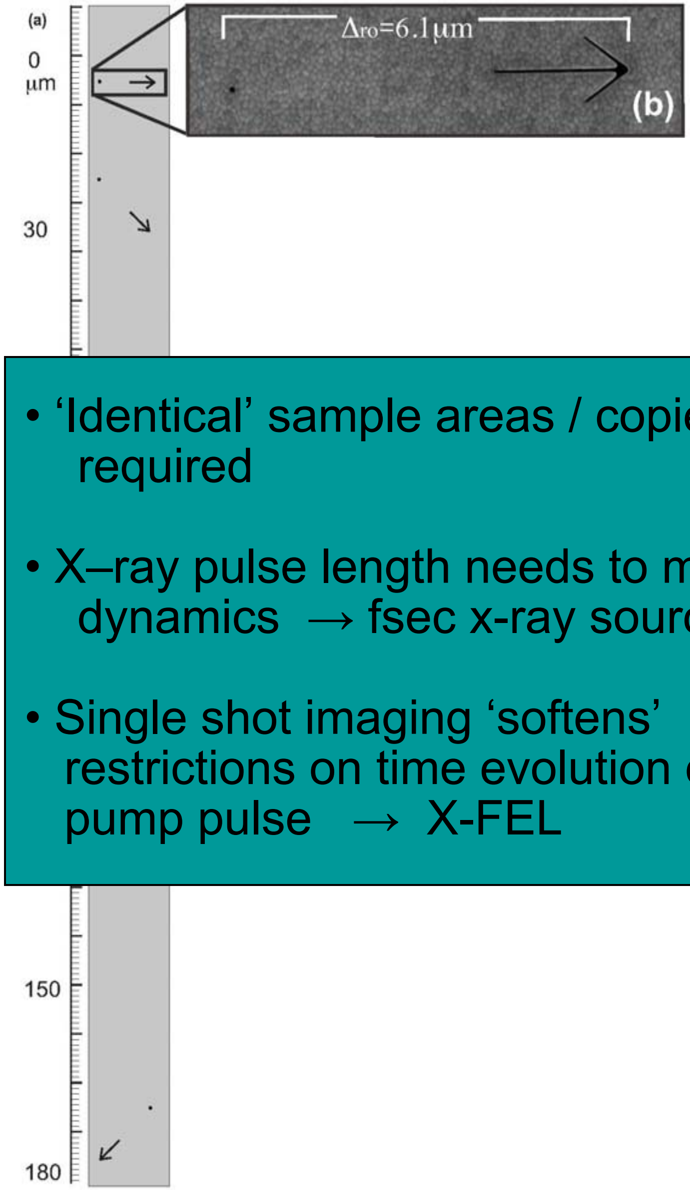
Low Noise



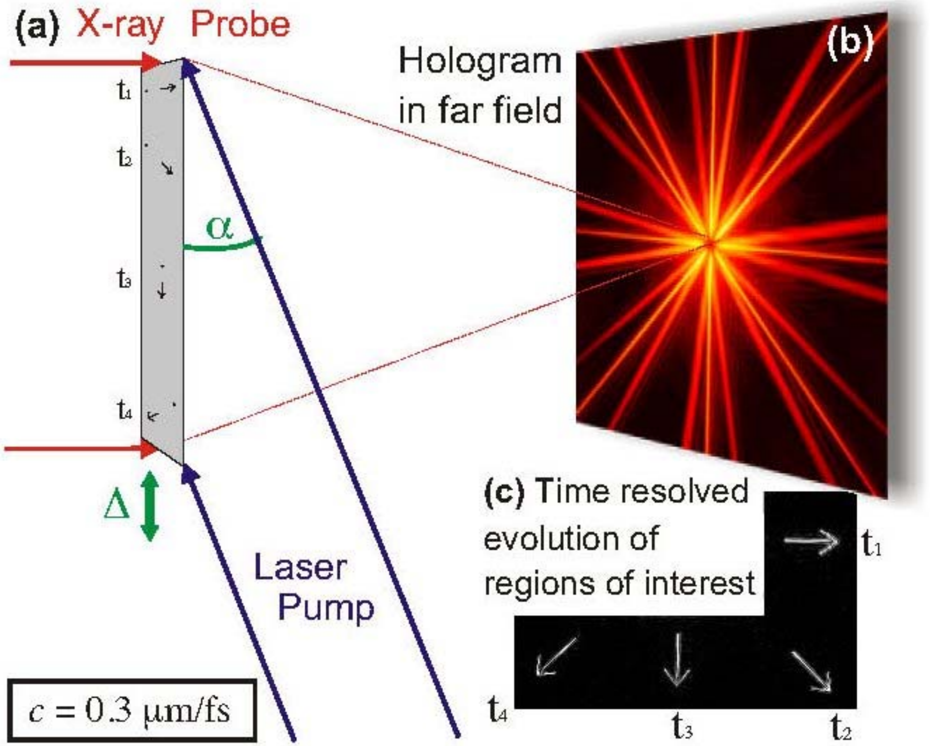


B. Schlotter et al., APL 06

Accessing dynamics by extending the FOV in FTH



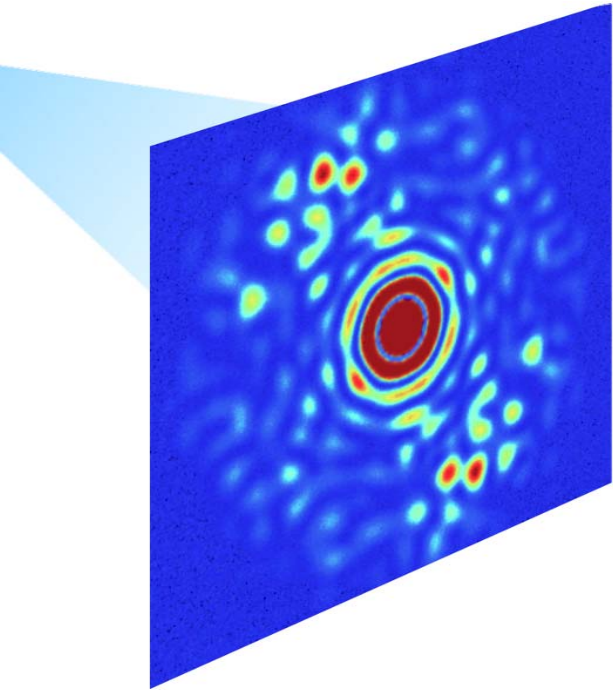
- ‘Identical’ sample areas / copies required
- X–ray pulse length needs to match dynamics → fsec x-ray sources
- Single shot imaging ‘softens’ restrictions on time evolution of pump pulse → X-FEL



Follow dynamics occurring on time scales ranging from ~ 1 fs to several ps

B. Schlotter et al., OL 07

Solving the phase problem



Phase problem in X-ray scattering:

Wave on detector is complex, but only intensity is measured, phase information is lost

Solutions:

1) X-ray Holography (Gabor 1948, Stroke 1965)

- Phase information is encoded in detectable intensity fluctuations
- True imaging technique

2) Iterative Phase Retrieval (Sayers 1952)

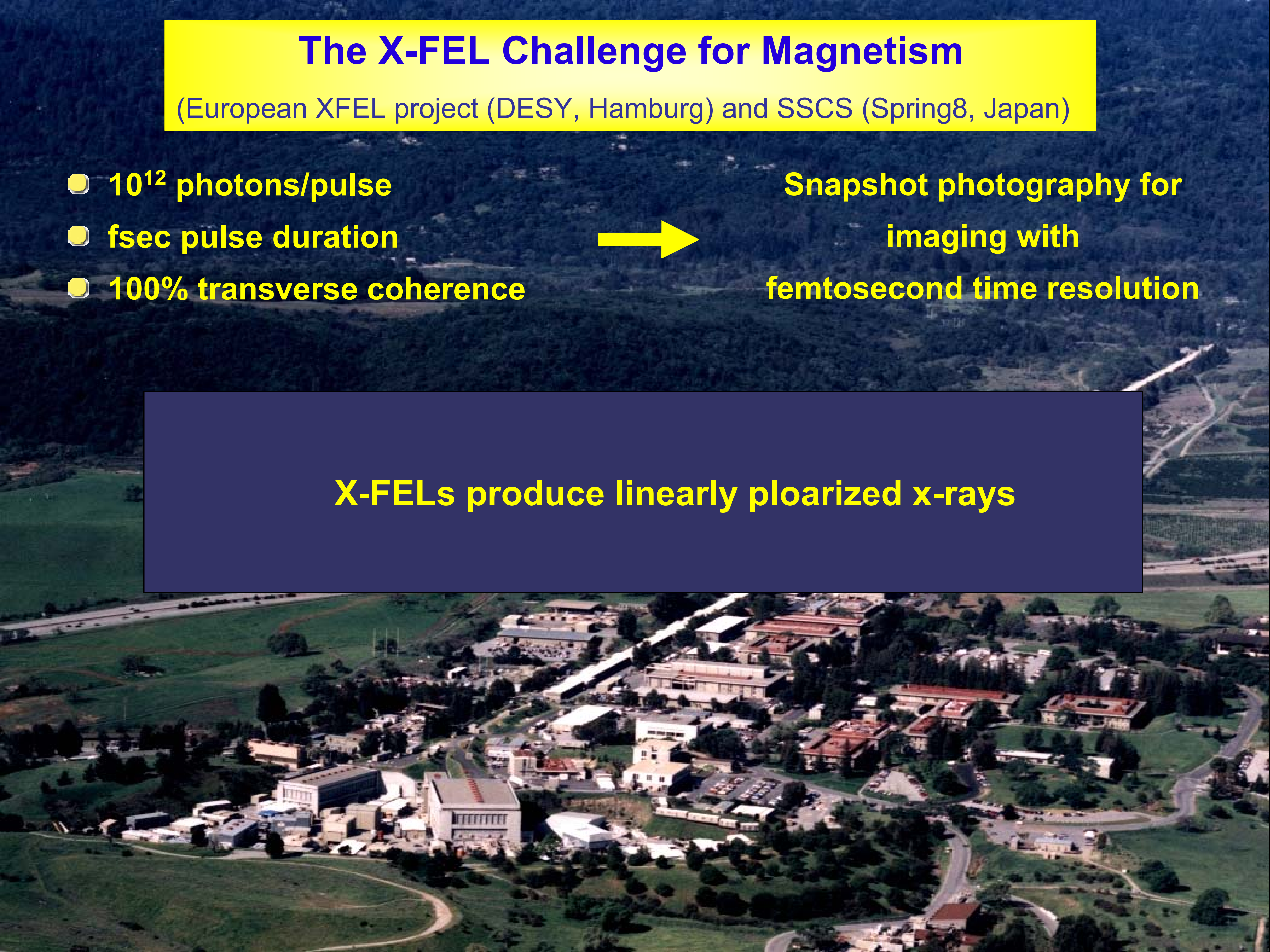
- Surround sample with ‘known’ support
- Measure additional scattering intensities (‘oversampling’)
- Use iterative algorithm to retrieve scattering phases from additional scattering intensities

3) other techniques

- Curved wave fronts
- Multiple wavelength Anomalous Diffraction (MAD)
- ?

The X-FEL Challenge for Magnetism

(European XFEL project (DESY, Hamburg) and SSCS (Spring8, Japan)



- 10^{12} photons/pulse
- fsec pulse duration
- 100% transverse coherence

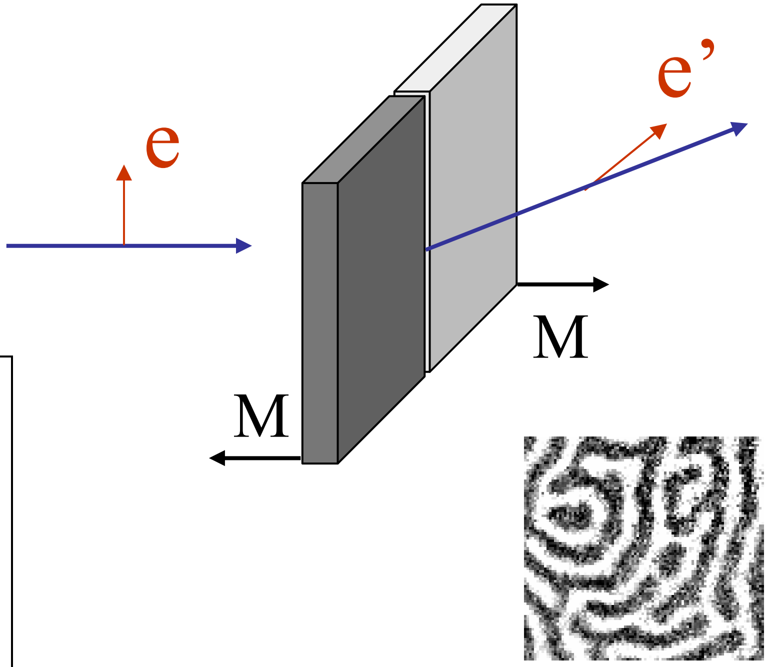
→ Snapshot photography for imaging with femtosecond time resolution

X-FELs produce linearly ploarized x-rays

Polarization and Resonant Magnetic Scattering

J. P. Hannon, G. T. Trammell, M. Blume, D. Gibbs, Phys. Rev. Lett 61, 1245 (1988)

$$I \propto \left|\sum_n \exp(i\mathbf{q}\cdot\mathbf{r}_n)\, f_n\right|$$

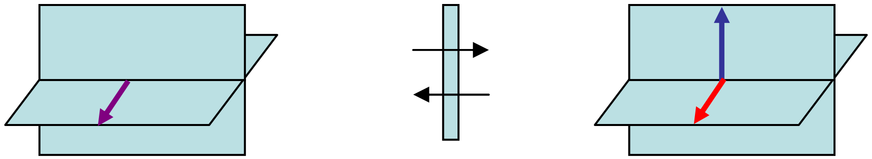


Resonant scattering amplitude (in this geometry):

charge

magnetic (XMCD)

$$f_n = (\mathbf{e}'\cdot\mathbf{e})F_n^c - i(\mathbf{e}' x\, \mathbf{e})\mathbf{M}_n\cdot F_n^{m_1}$$

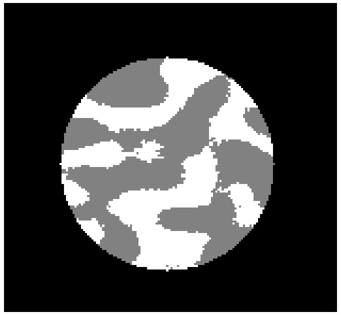


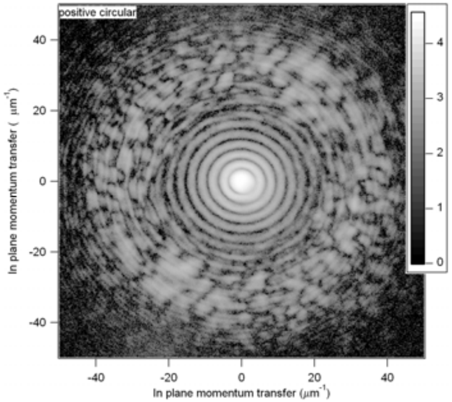
Circular polarization: Eigenstate

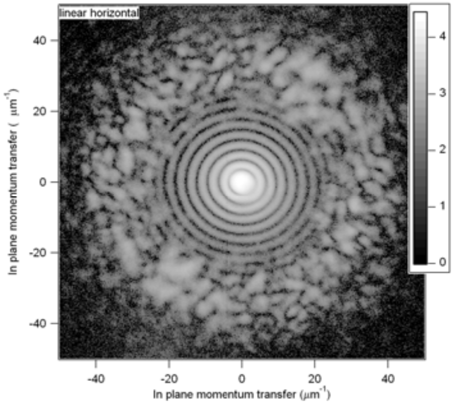
Linear Polarization: π→σ , σ→π

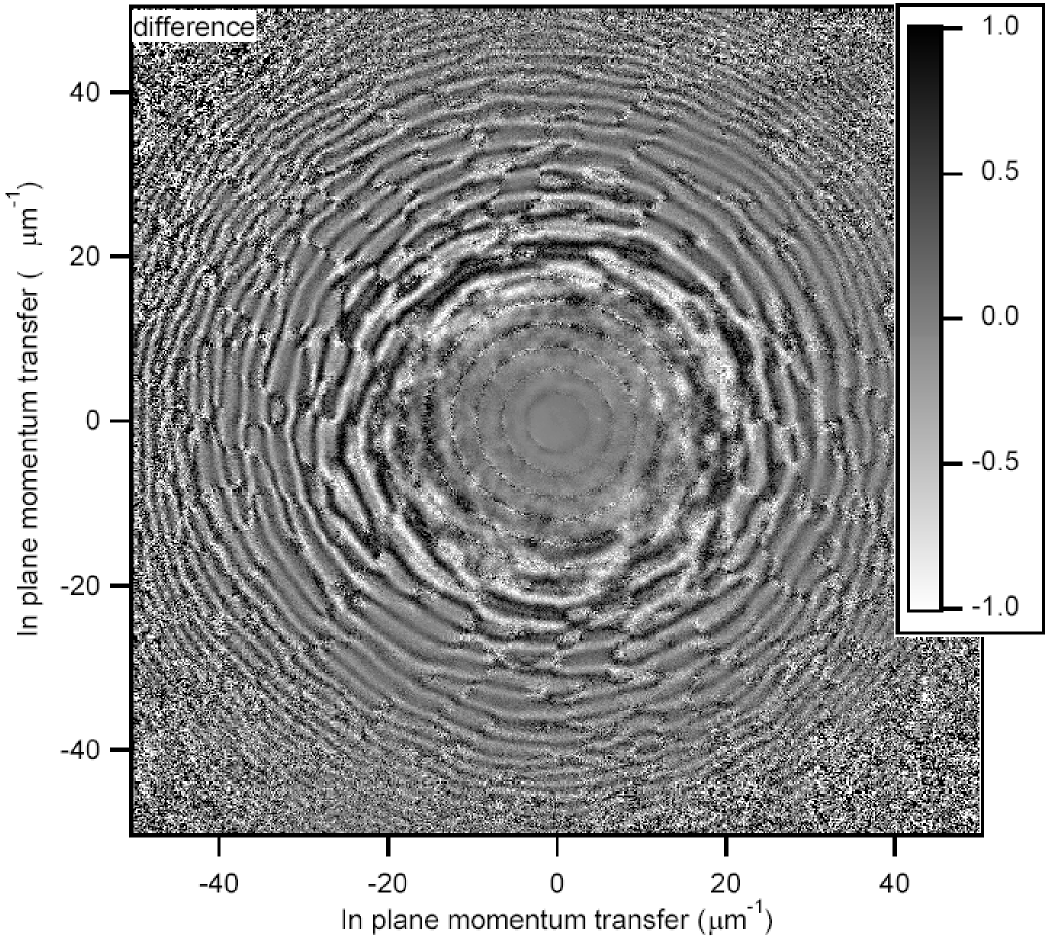
Interference of Charge and Magnetic Scattering

∅ Pinhole > ς mag. domains





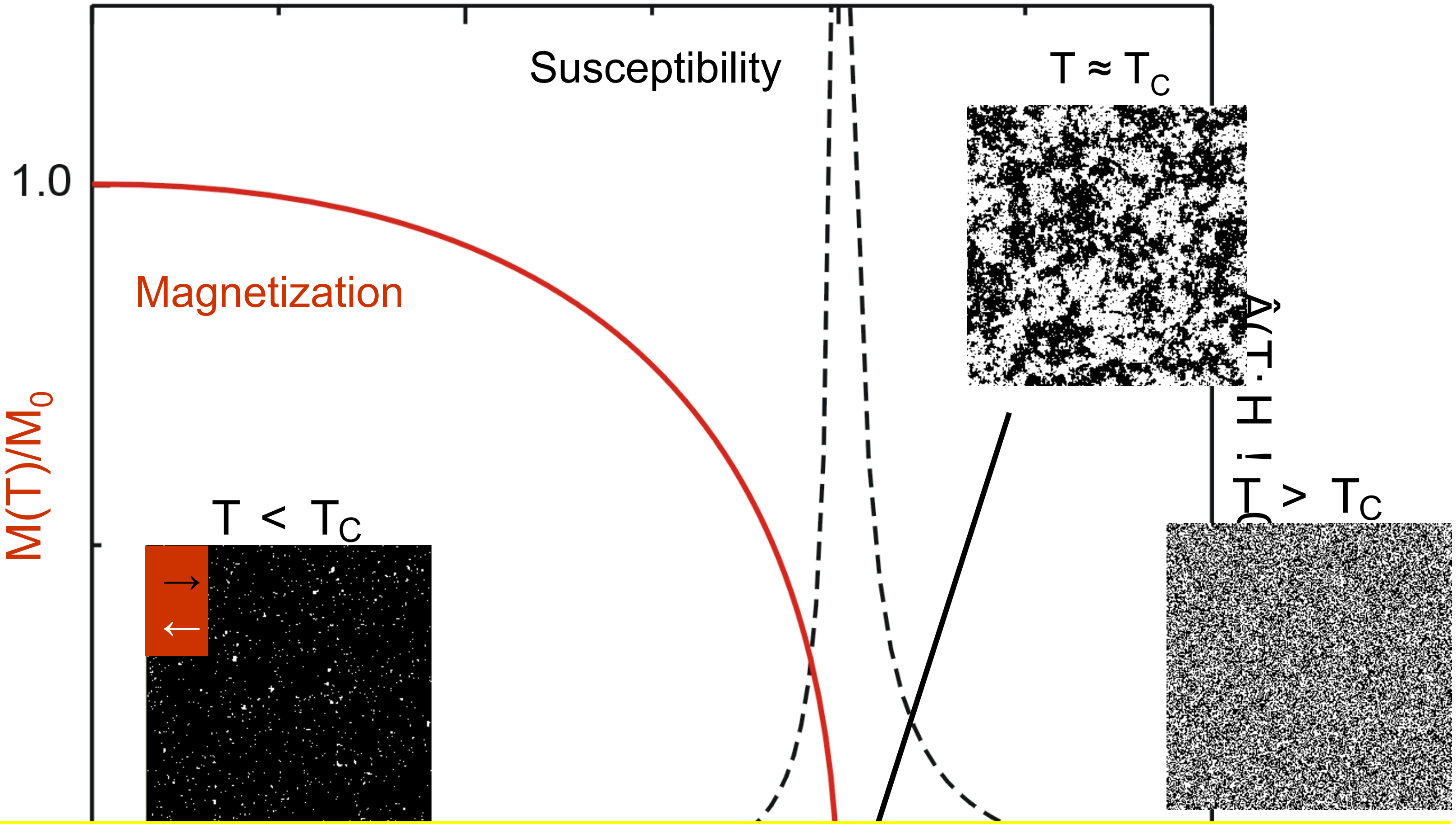




Interference = Positive-Circular — Linear

$$I_R - I_L = (Pin + Mag)^2 - (Pin^2 + Mag^2) = 2\,(Pin \cdot Mag)$$

Magnetic phase transitions

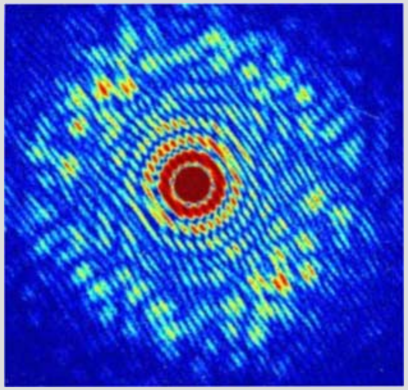


- Image of critical fluctuations is computer simulation of Ising model (Web page of Schwabl, TU Munich).
- Critical fluctuations in 3D are expected to be small and fast
- In 2D fluctuations expected to be larger

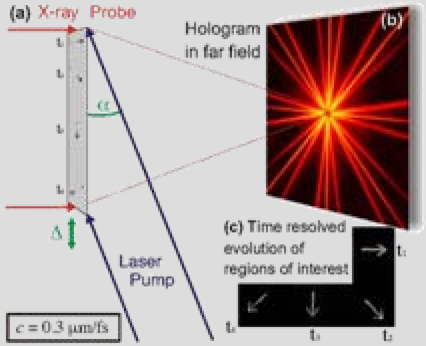
Conclusions

- Current status of lensless x-ray microscopy
  - holography
  - phase retrieval
- Novel & unique experimental opportunities
- Ideally suited for future coherent x-ray sources

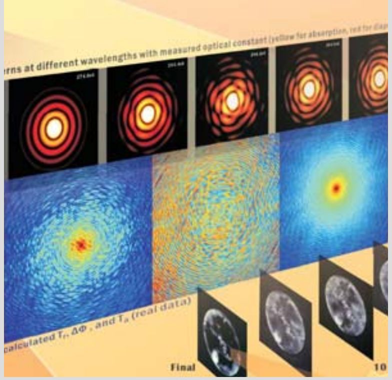
Fourier Transform Holography



Unique Properties of FT Holography



New & Recent Developments



Free Electron X-ray Lasers