Lensless X-Ray Spectromicroscopy on the Nanoscale

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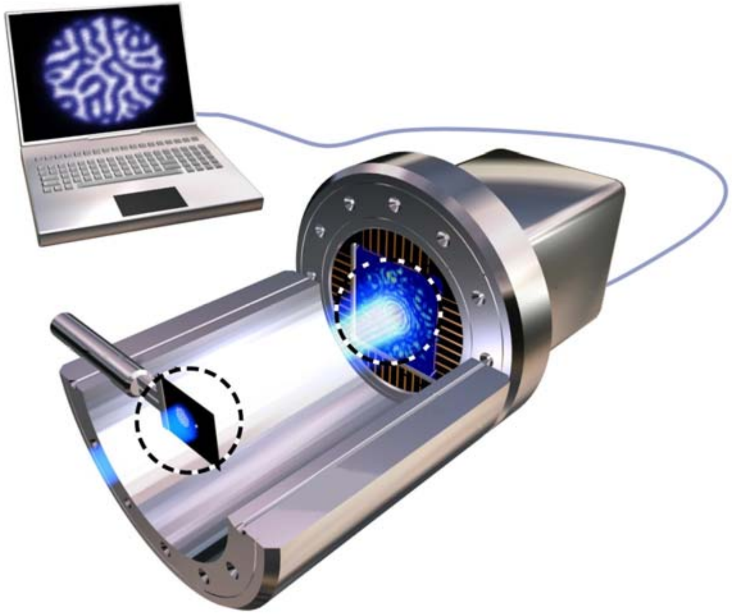


Bill Schlotter
Andreas Scherz
Jo Stöhr

Stefan Eisebitt
Olav Hellwig
Wolfgang Eberhardt

Outline

- **Fourier Transform Holography**

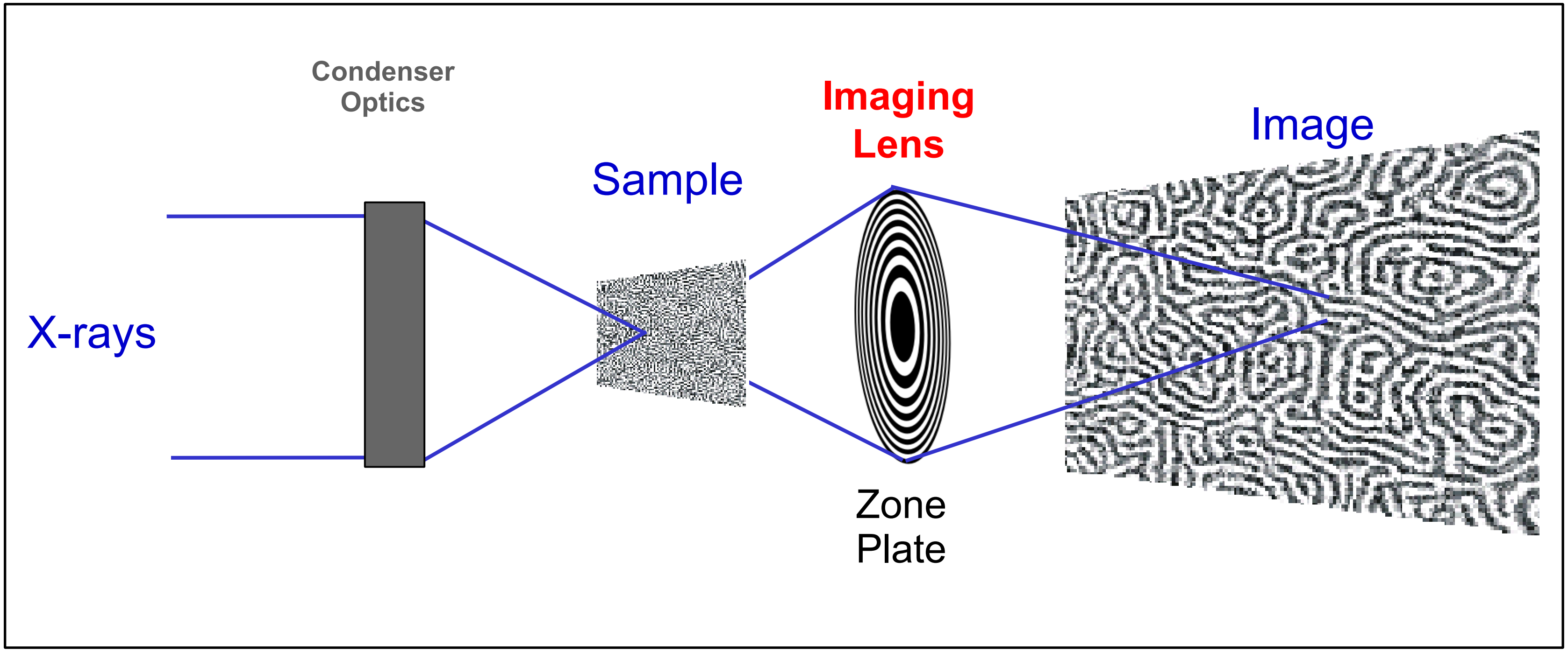
  An ideal microscopy technique for an ERL

- **Soft X-Ray ERL Beam Line**

  Unique capabilities for soft matter & magnetism

- **Magnetization Dynamics**

Lens-based microscopy – Real space imaging

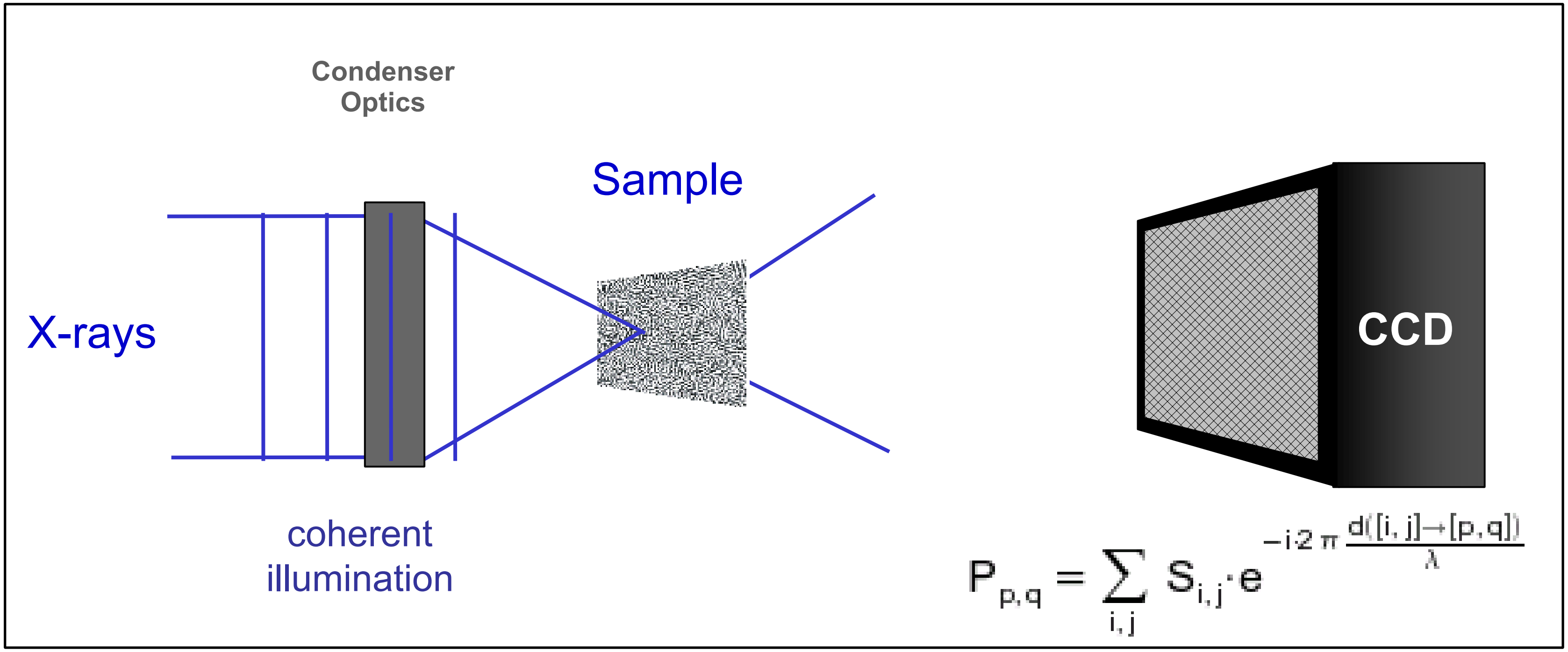


- Lens quality determines spatial resolution of microscope
- Zone plate efficiency problematic below 10 nm spatial resolution

ERL specific:

- Full field TXM requires incoherent source
- STXM benefits from coherence, but ‘slow’ as a scanning technique

Lens-less microscopy – Fourier space imaging



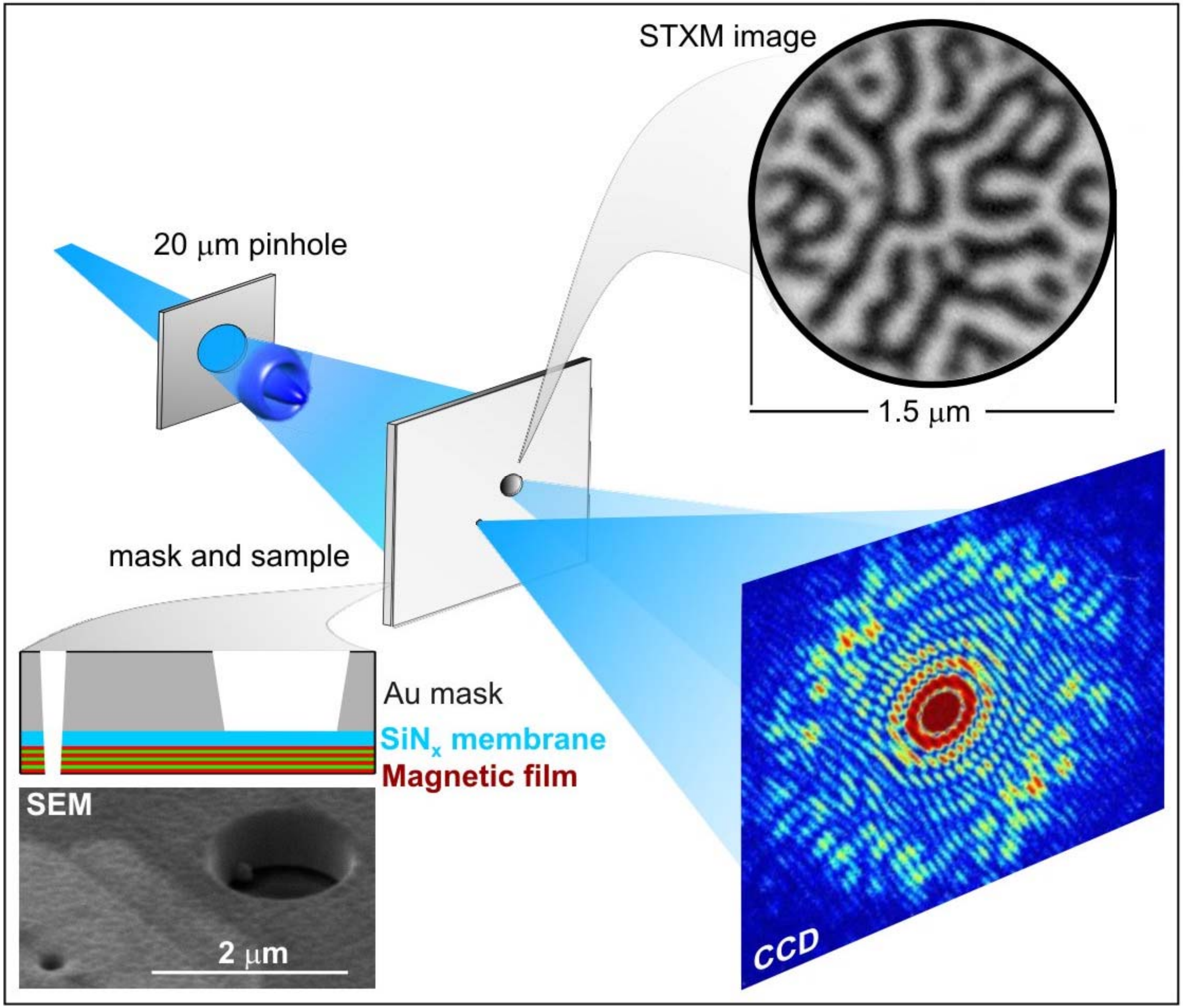
$$P_{p,q} = \sum_{i,j} S_{i,j} \cdot e^{-i \cdot 2\pi \frac{d([i,j] \rightarrow [p,q])}{\lambda}}$$

Idea:

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

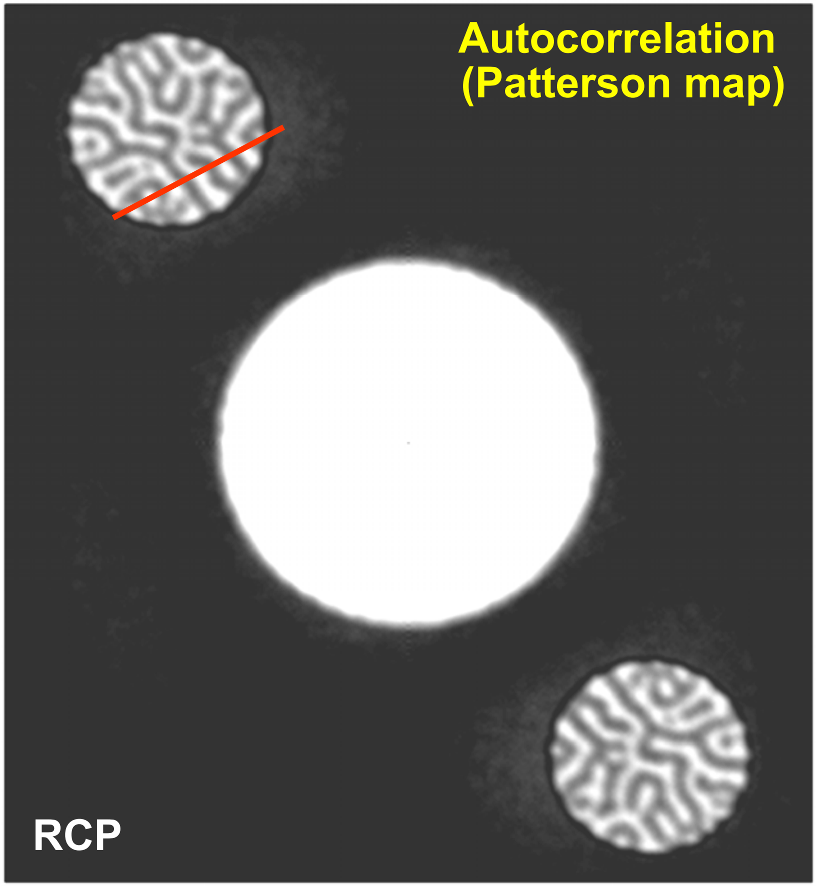
Potential for wavelength limited resolution: Highest detected momentum transfer (Fourier component) defines spatial resolution.

Fourier Transform x-ray spectro-holography

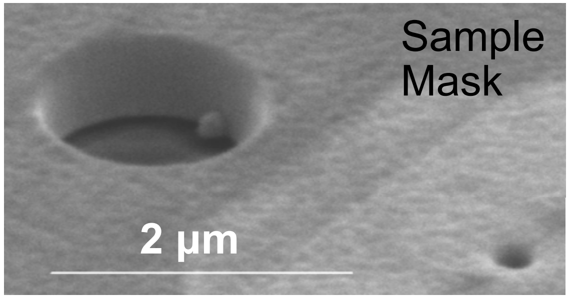


Digital image reconstruction

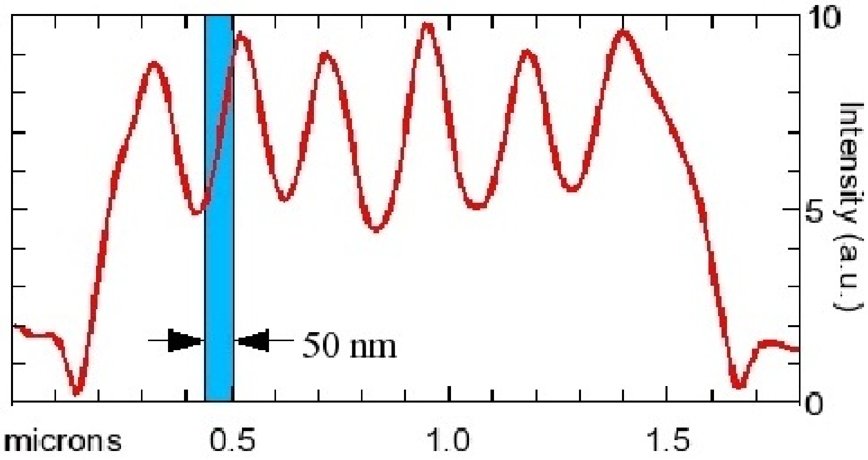
Single Fourier transformation of scattering intensities yields the auto-correlation of sample, which contains image of sample due to the off-axis geometry in FT holography. (correlation theorem)



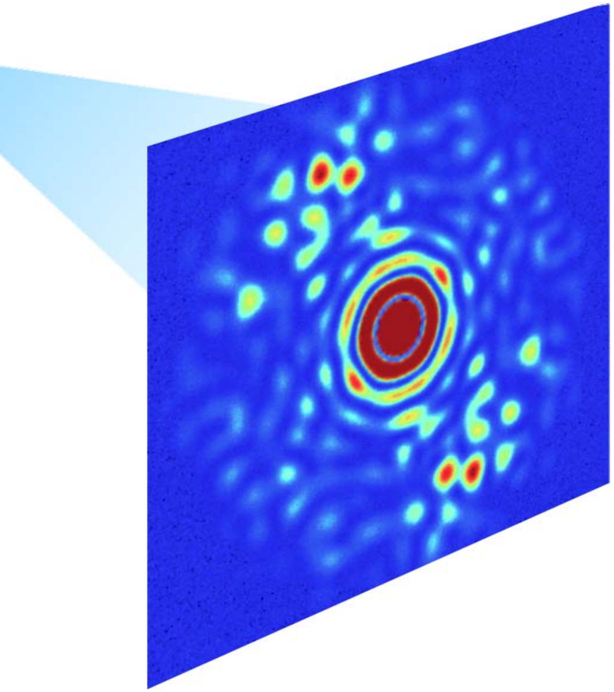
Intensity in image center, which contains self-correlation of apertures, is truncated.



10% - 90% intensity rise over about 50 nm



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)

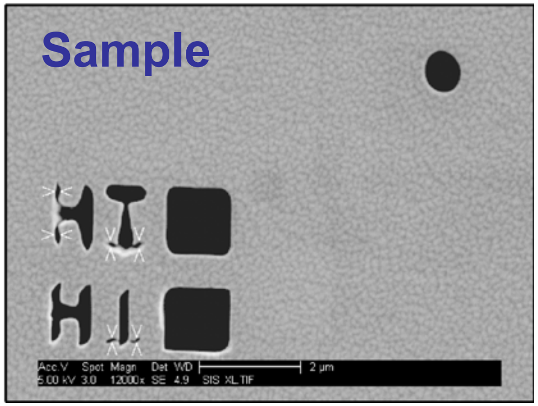
- Phase information is encoded in detectable intensity fluctuations
- True imaging technique
- Reference size determines spatial resolution

2) Iterative Phase Retrieval (Sayers 1952)

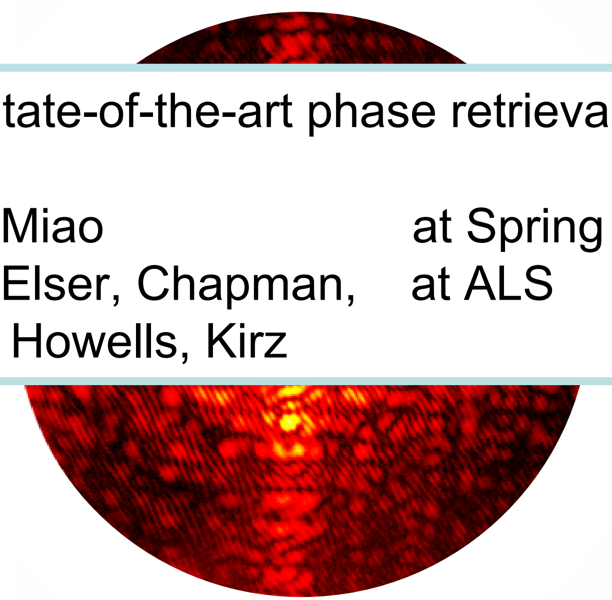
- Use iterative algorithm to retrieve scattering phases from additional scattering intensities
- Detected momentum transfer defines spatial resolution
- Surround sample with ‘known’ support and measure additional scattering intensities (‘oversampling’)

Holography and phase retrieval

from coherent x-ray scattering *alone*

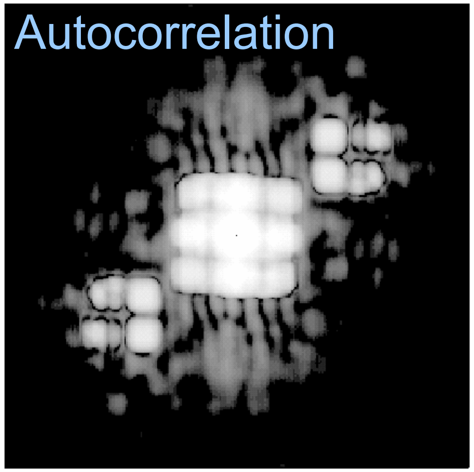


Coherent Scattering

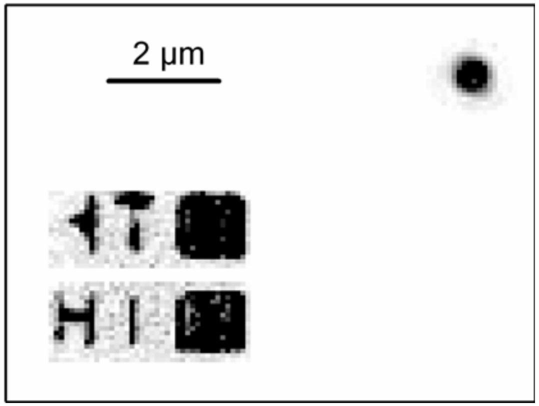


State-of-the-art phase retrieval

- Miao at Spring 8
- Elser, Chapman, Howells, Kirz at ALS



Phase Reconstruction



Appl. Phys. Lett., **84**, 3373 (2004)

Key properties of Fourier transform X-ray holography

- ***True imaging technique***

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

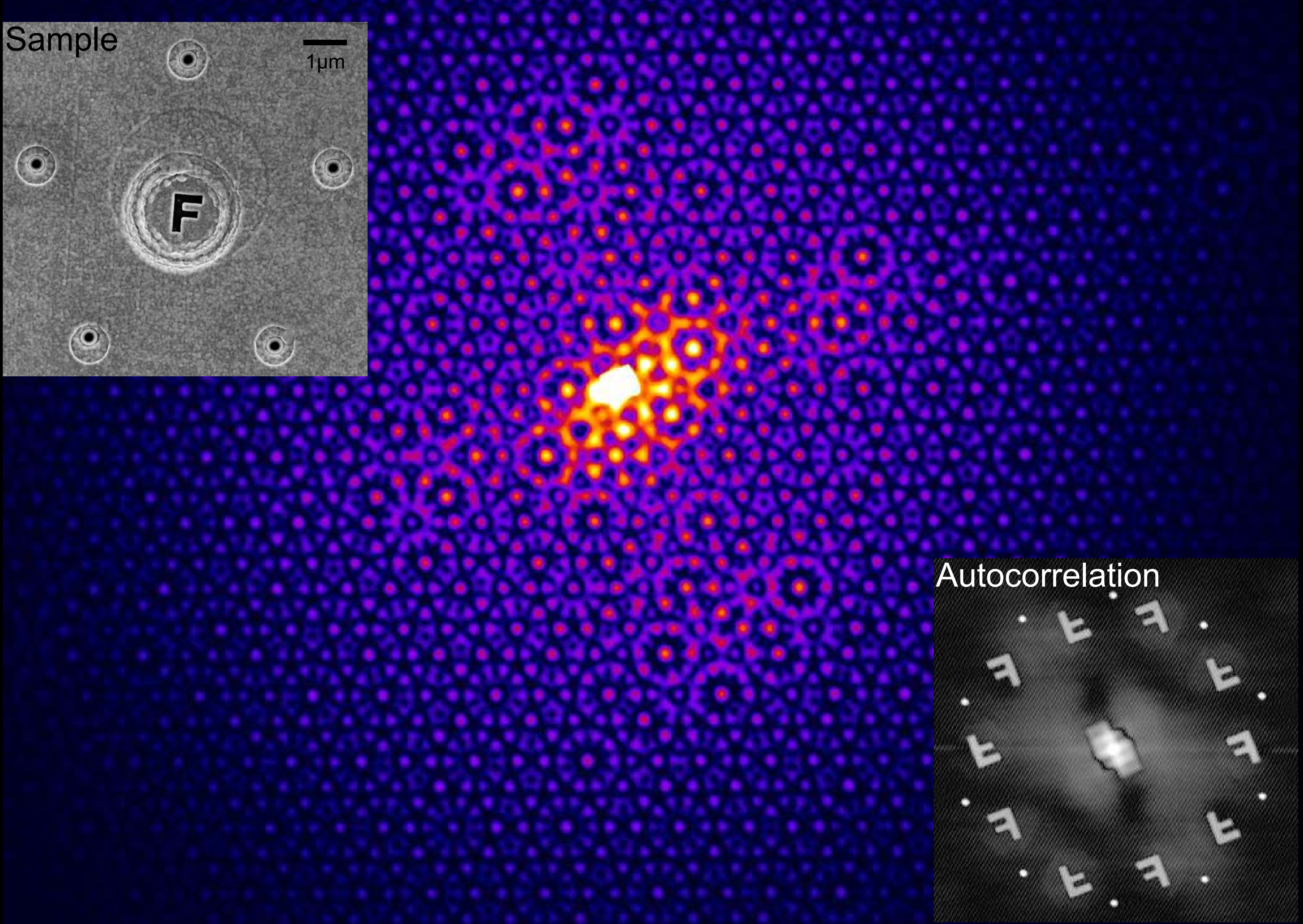
- ***Nanometer resolution with micron stability***
  Setup is basically insensitive to vibrations
  or thermal drifts

- ***Wide applicability***
  - Sample on/in/behind object aperture
  - Rapid sample change, since no alignment
  - No space constraints around sample
  - UHV to ambient pressure (to be shown)

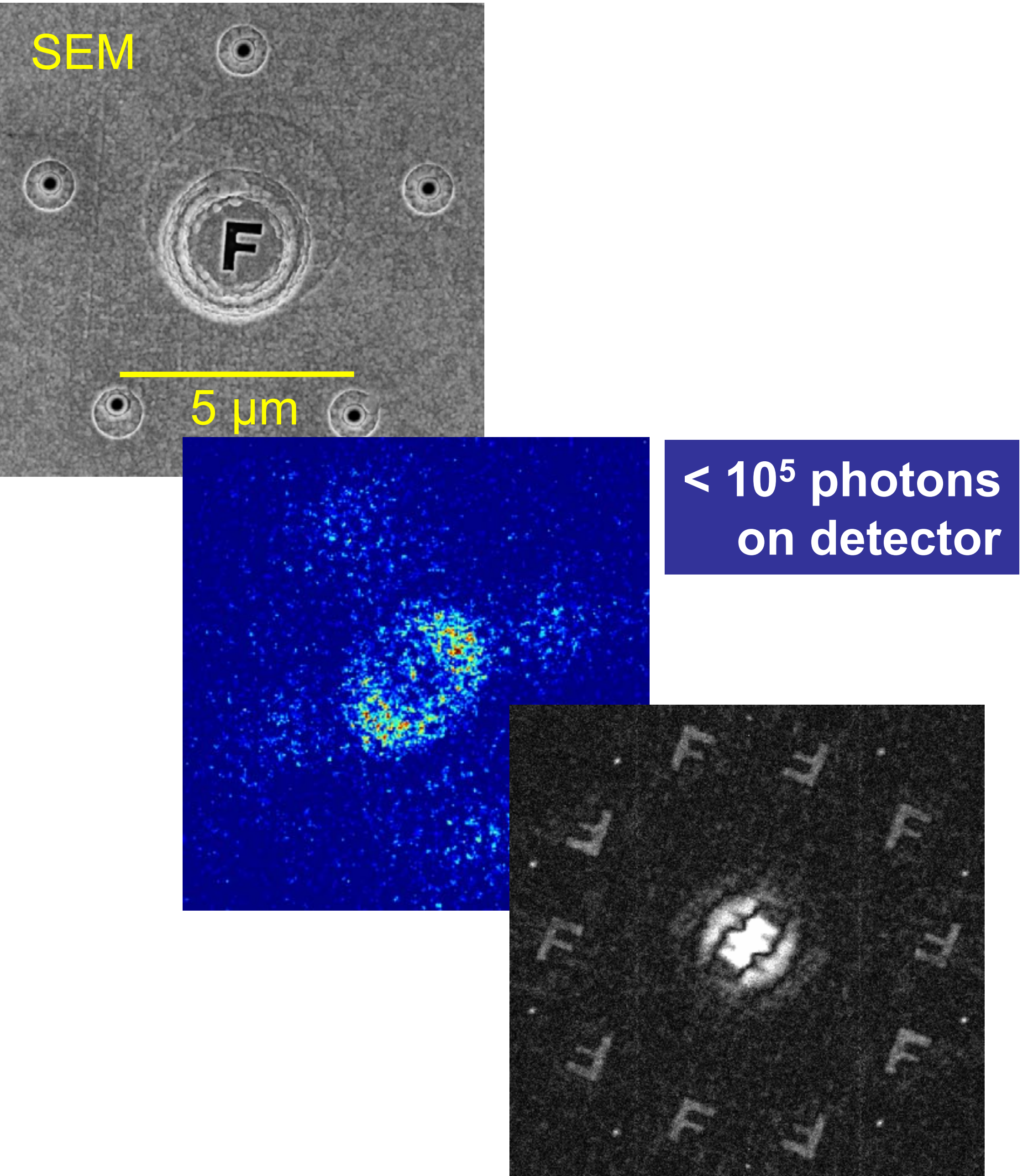
- ***Reflection geometry***
  Thin film and surface sensitivity

- ***Inverted structure***
  Sample and reference on transparent support

Multiple reference FT holography

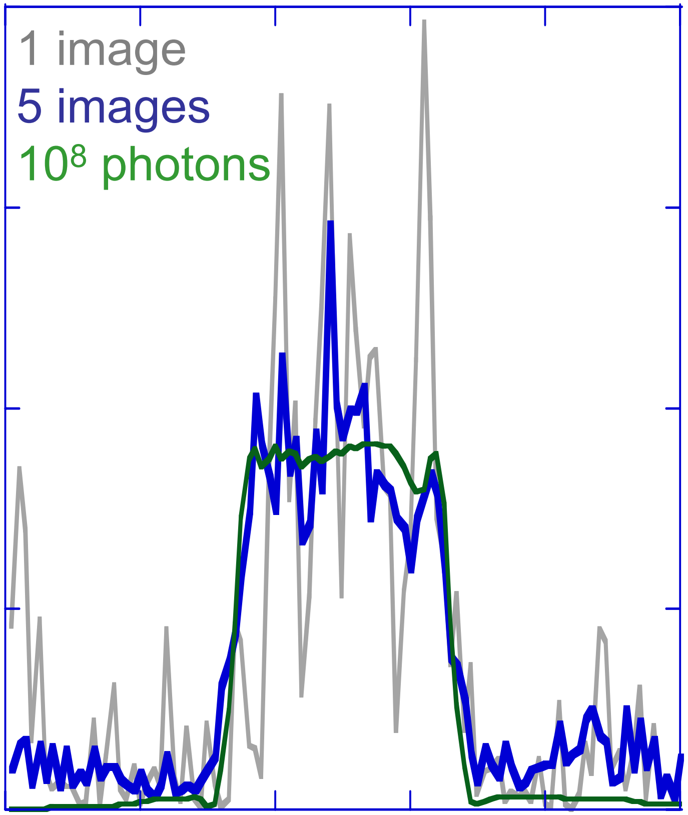


Multiple reference Fourier transform holography

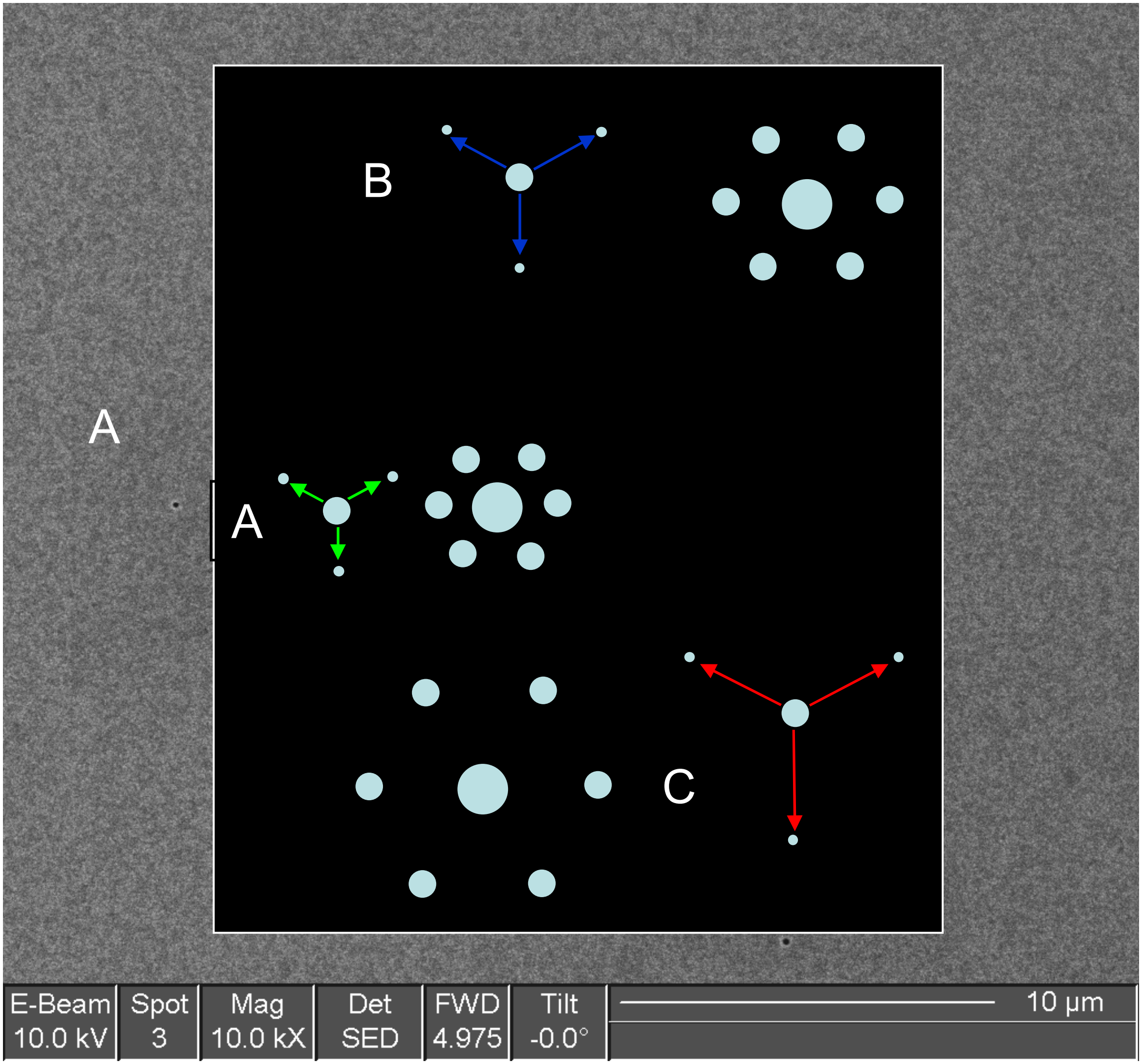


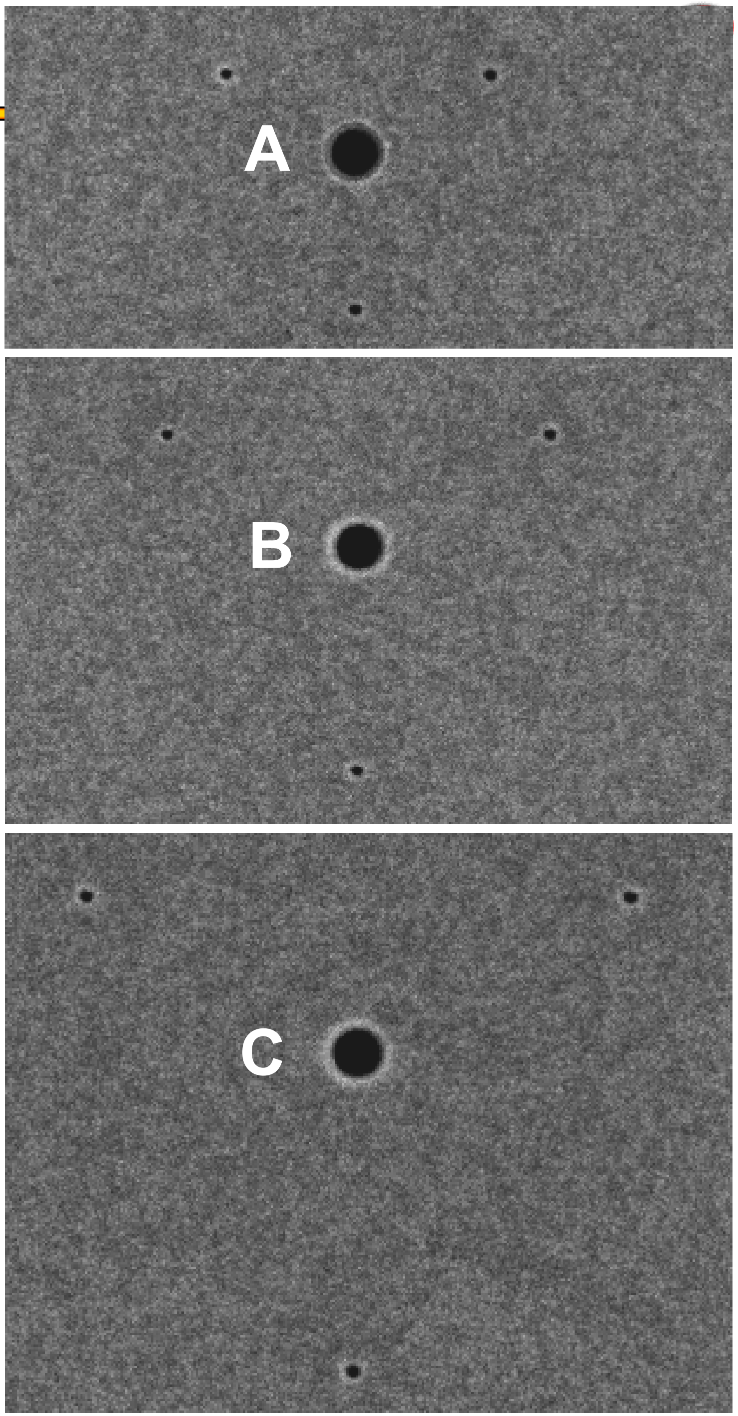
< 10^5 photons on detector

Image signal improves with number of references



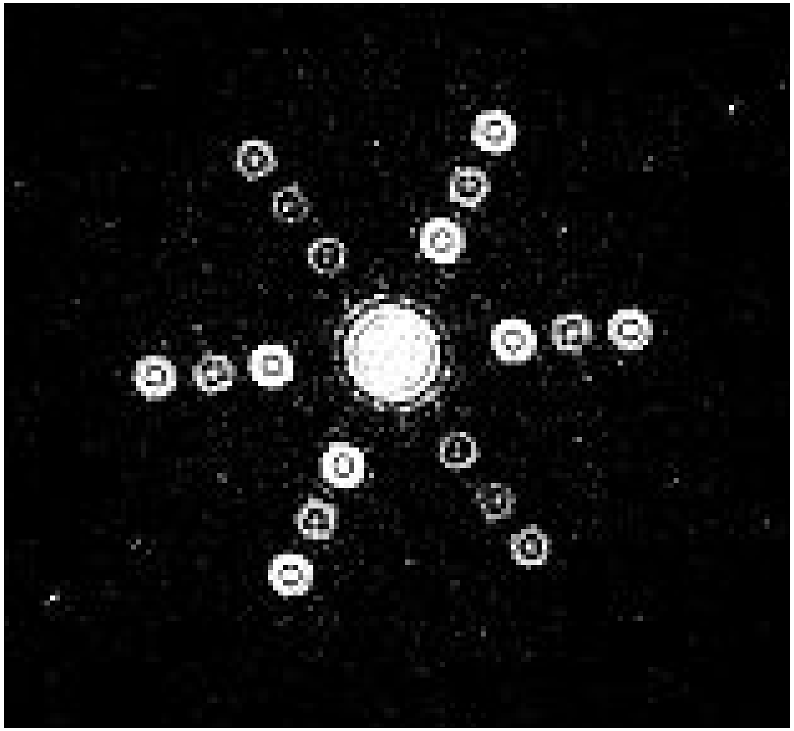
Sample and reference multiplexing



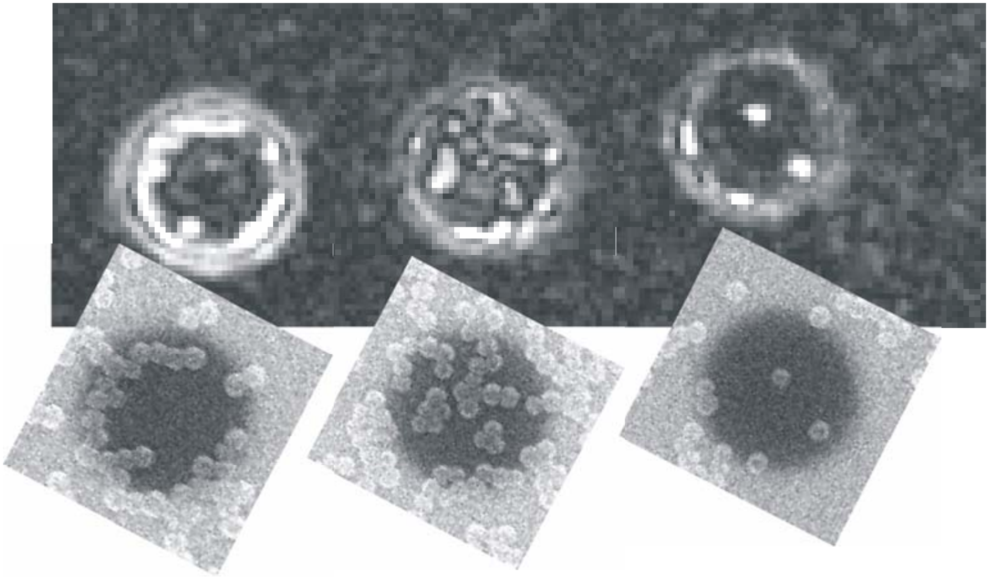


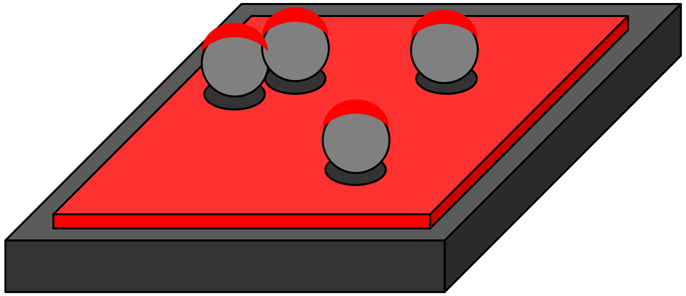
Patterned magnetic storage media

Autocorrelation



9 nm Co/Pd ML on Ø 58 nm PS spheres



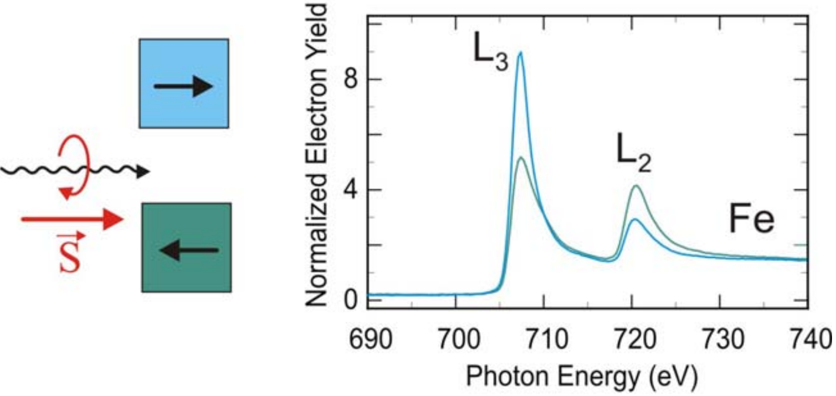


M. Albrecht et al., Nature Mater. 4 (2005) 203.
T. Ulbrich et al., PRL. 96 (2006) 077202

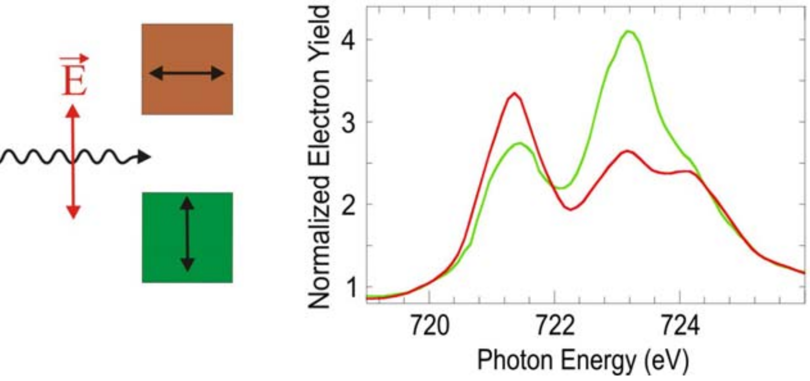
Universität Konstanz

Benefits of soft x-ray energy range

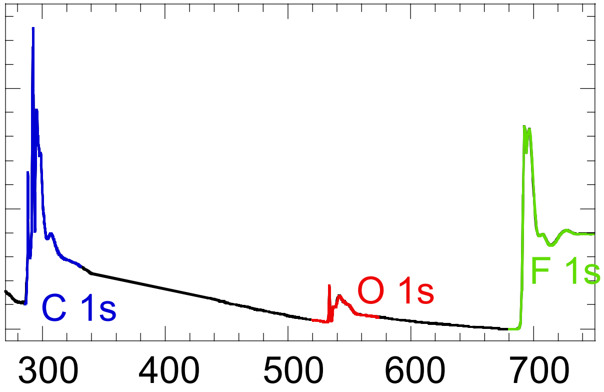
X-ray Magnetic Circular Dichroism



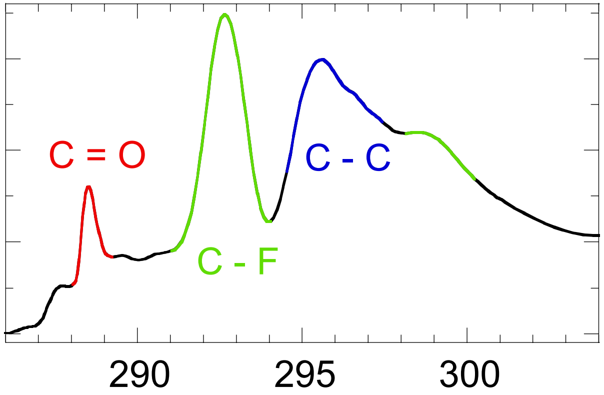
X-ray Magnetic Linear Dichroism



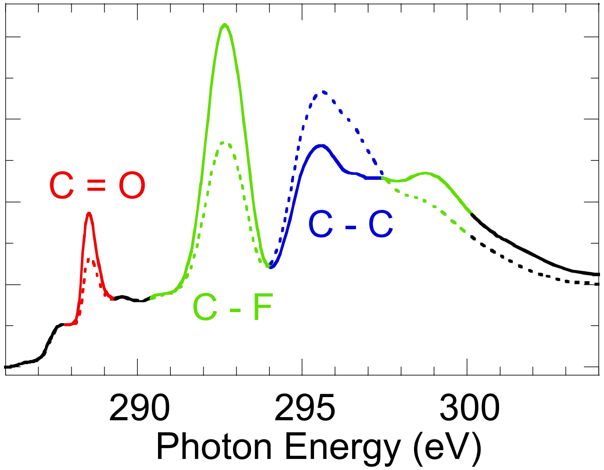
Elemental Specificity



Chemical Sensitivity



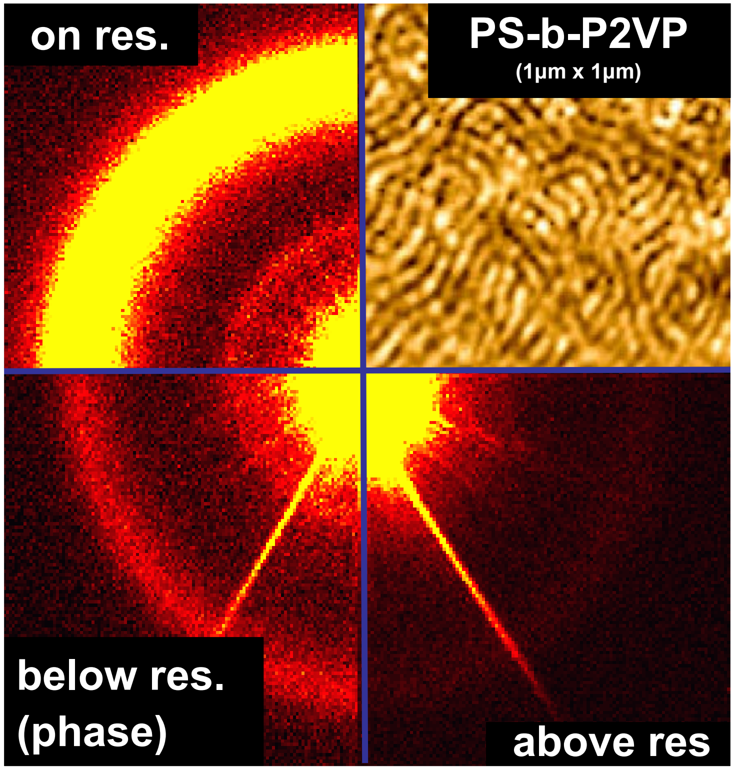
Orientation Sensitivity



Tuning of cross sections

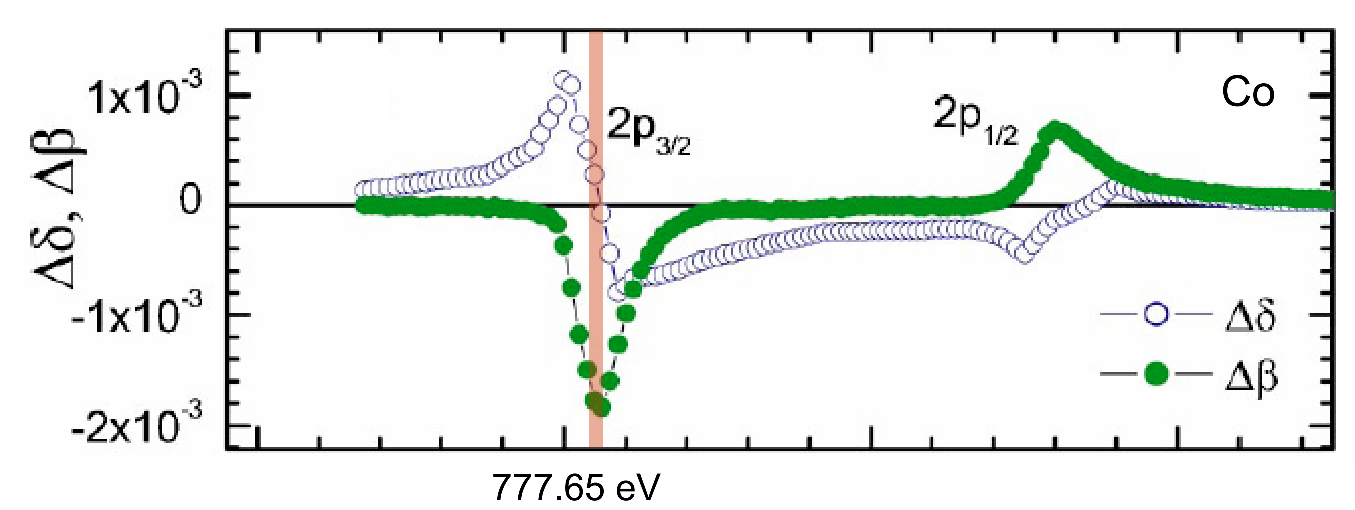
- Sensitivity of absorption cross section to elemental/chemical composition and presence of charge/spin ordering exploited in soft x-ray spectromicroscopy

- Scattering cross section exhibits same dependences, which can be used to tune the scattering contrast.



Phase contrast soft x-ray holography

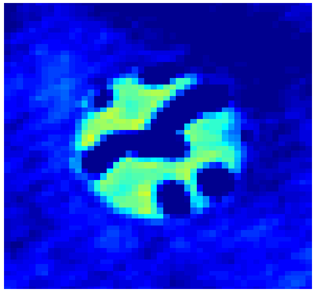
Refractive index is complex: $n = 1 - \delta + i\beta$



C. Mertens et al
PRB

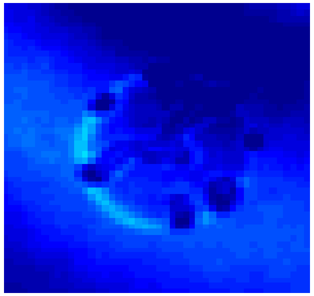
FTH yields autocorrelation (correlation theorem): $a * a = \mathcal{F}^{-1}(|\mathcal{F}(a)|^2)$

real part of AC
"Attenuation"



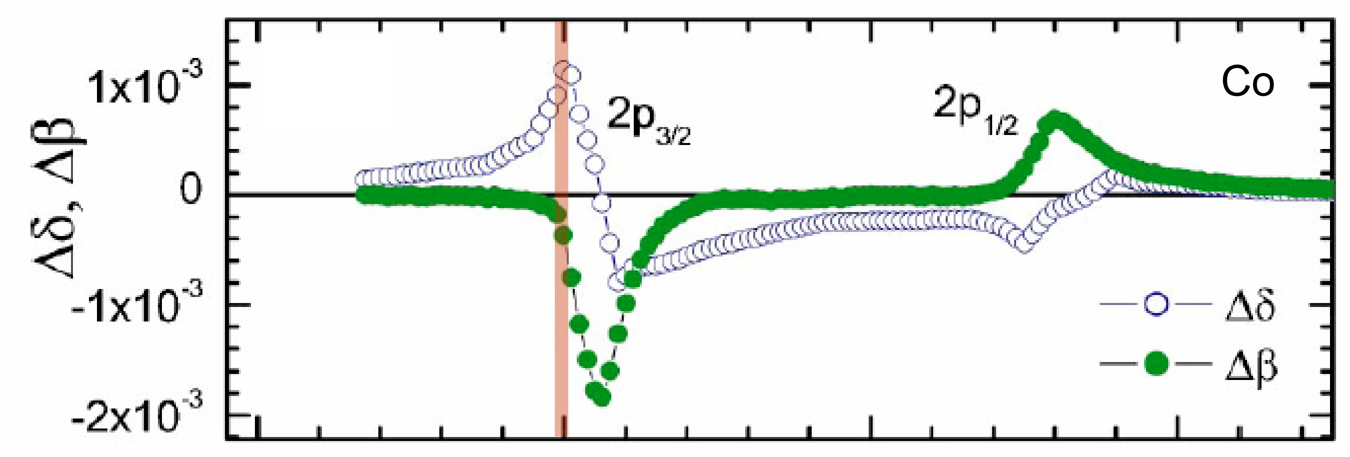
On resonance
$\lambda_x = 15$ nm

imaginary part of AC
"Phase shift"



Phase contrast soft x-ray holography

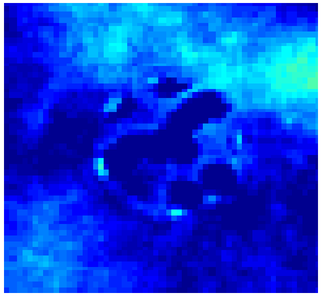
Refractive index is complex: $n = 1 - \delta + i\beta$



C. Mertens et al
PRB

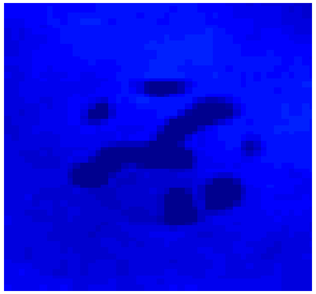
Imaging with phase contrast before absorption resonance reduces absorbed energy by factor of ~20

real part of AC
"Attenuation"

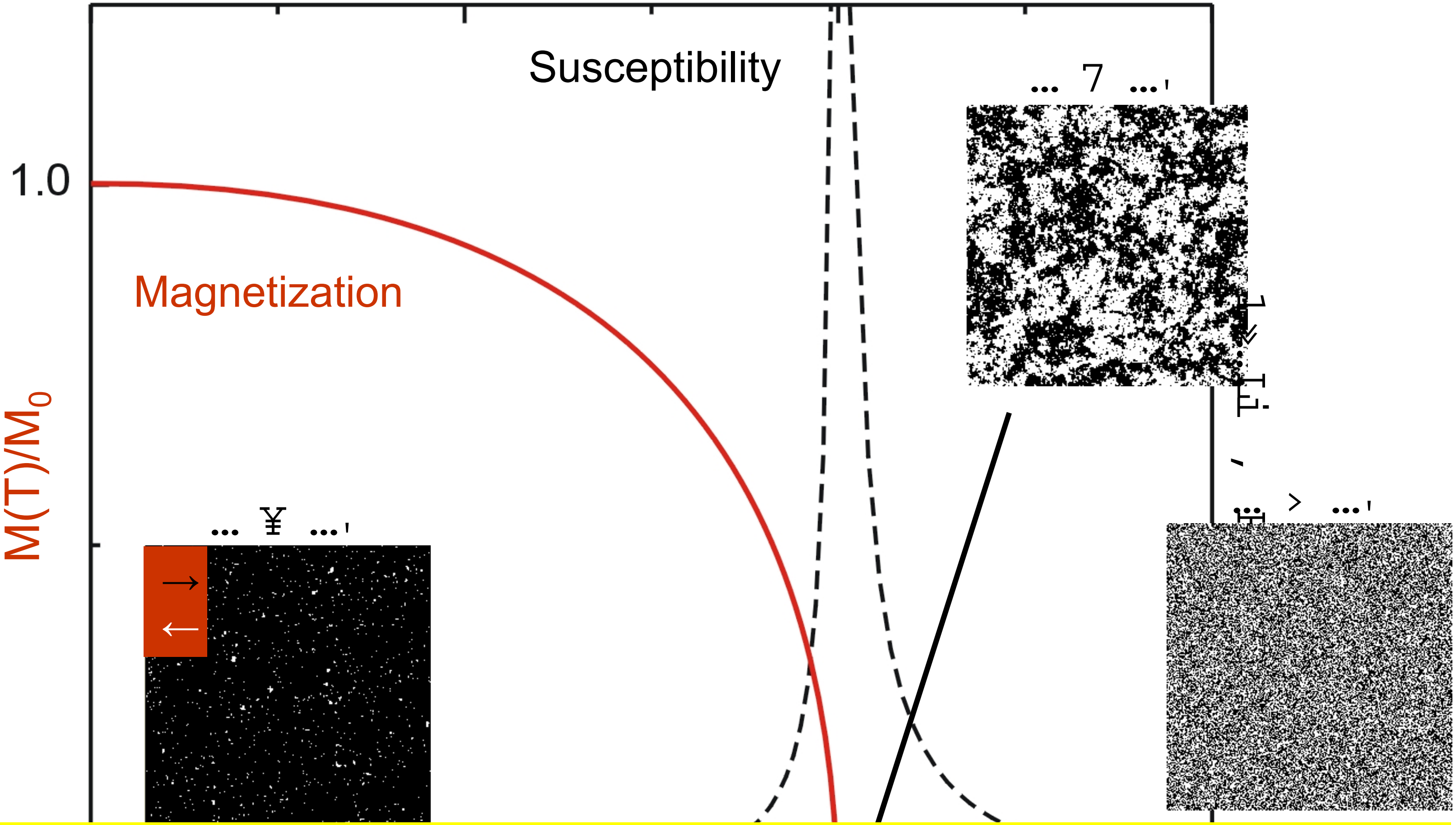


Before resonance
λ_x = 600 nm

imaginary part of AC
"Phase shift"

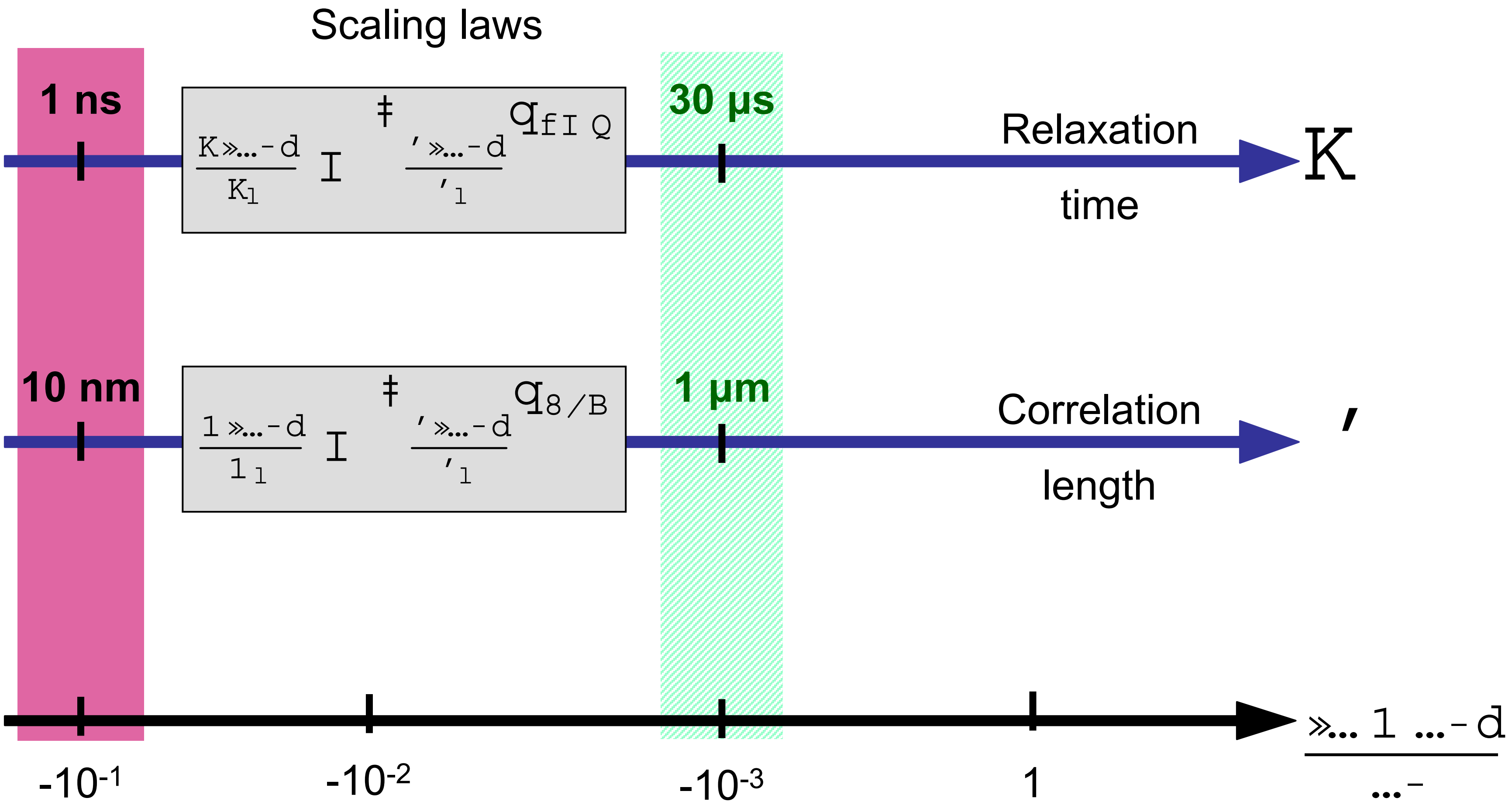


Magnetic phase transitions



- 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

2D critical magnetic fluctuations: Size and dynamics



Input for scaling laws:

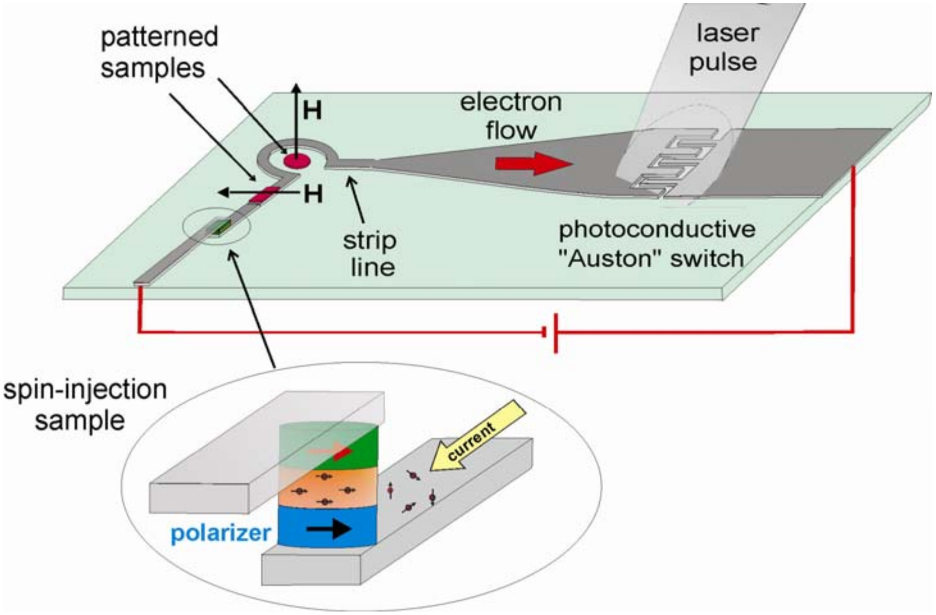
ξ_0 is range of spin correlation in ferromagnetic phase

Relaxation time K1 from FMR line width

Susceptibility 1(T) for 1.8 ML Fe/W from Back et al., Nature (95)

Magnetic field pulses by laser-generated current pulses

Photoconductive Auston Switch

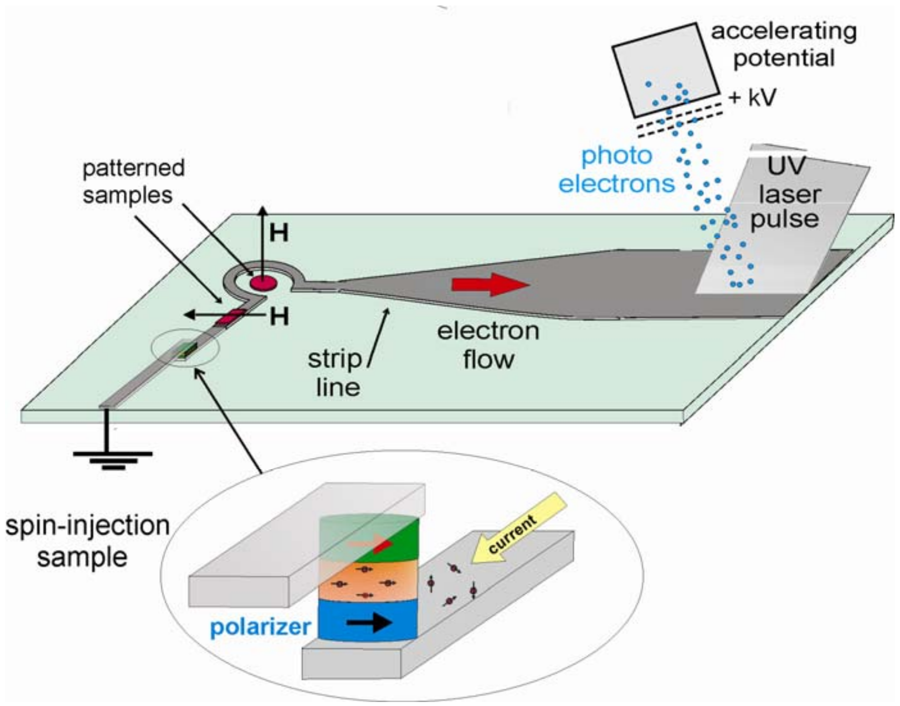


Peak Current: 100 - 1000 mA

Pulse Rise Time: 5 ps (30 ps currently)

Peak Field: .02 - .2 T

Photoemission into Vacuum



Potential for >>10 A peak current

Pulse in the sub picosecond range

Conclusions

- Lensless imaging is full field microscopy technique requiring coherent source

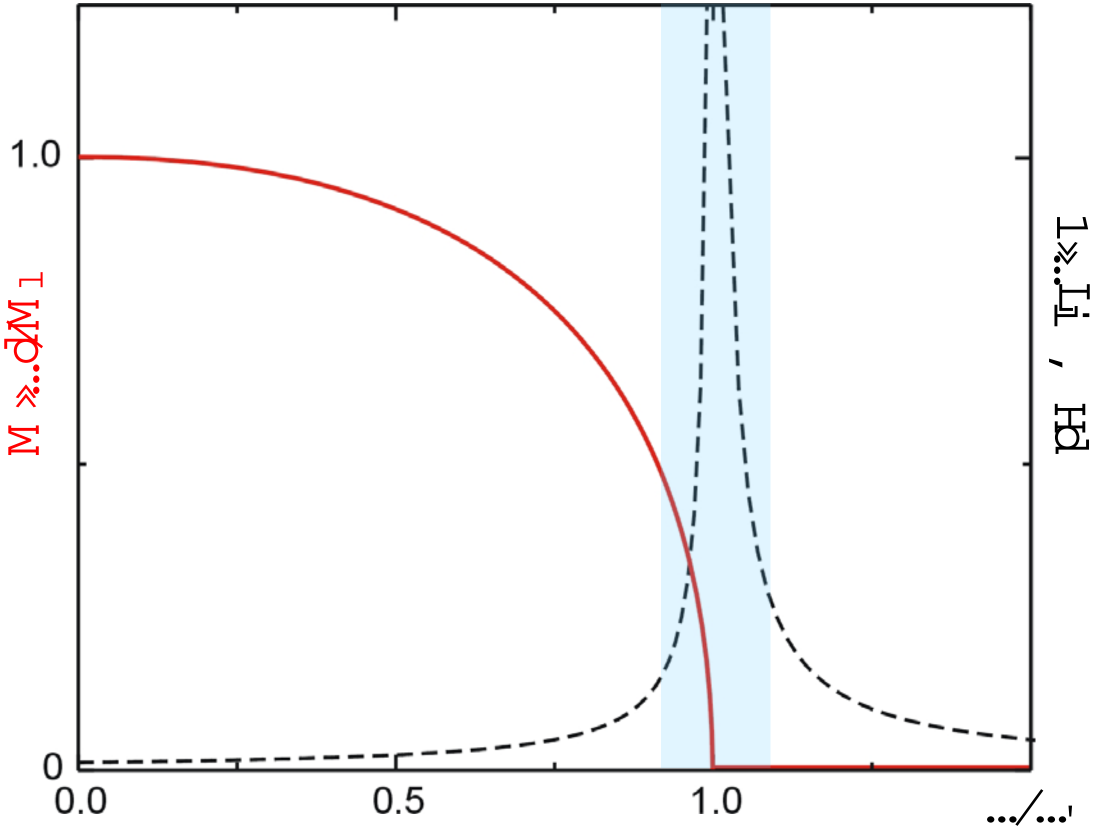
- Soft x-ray energy range contains relevant resonances of
  - *K* edges of light elements → organic matter
  - L_{23} edges of TM → magnetism

- Most important ERL characteristics
  - high coherent flux
  - fsec pulse length

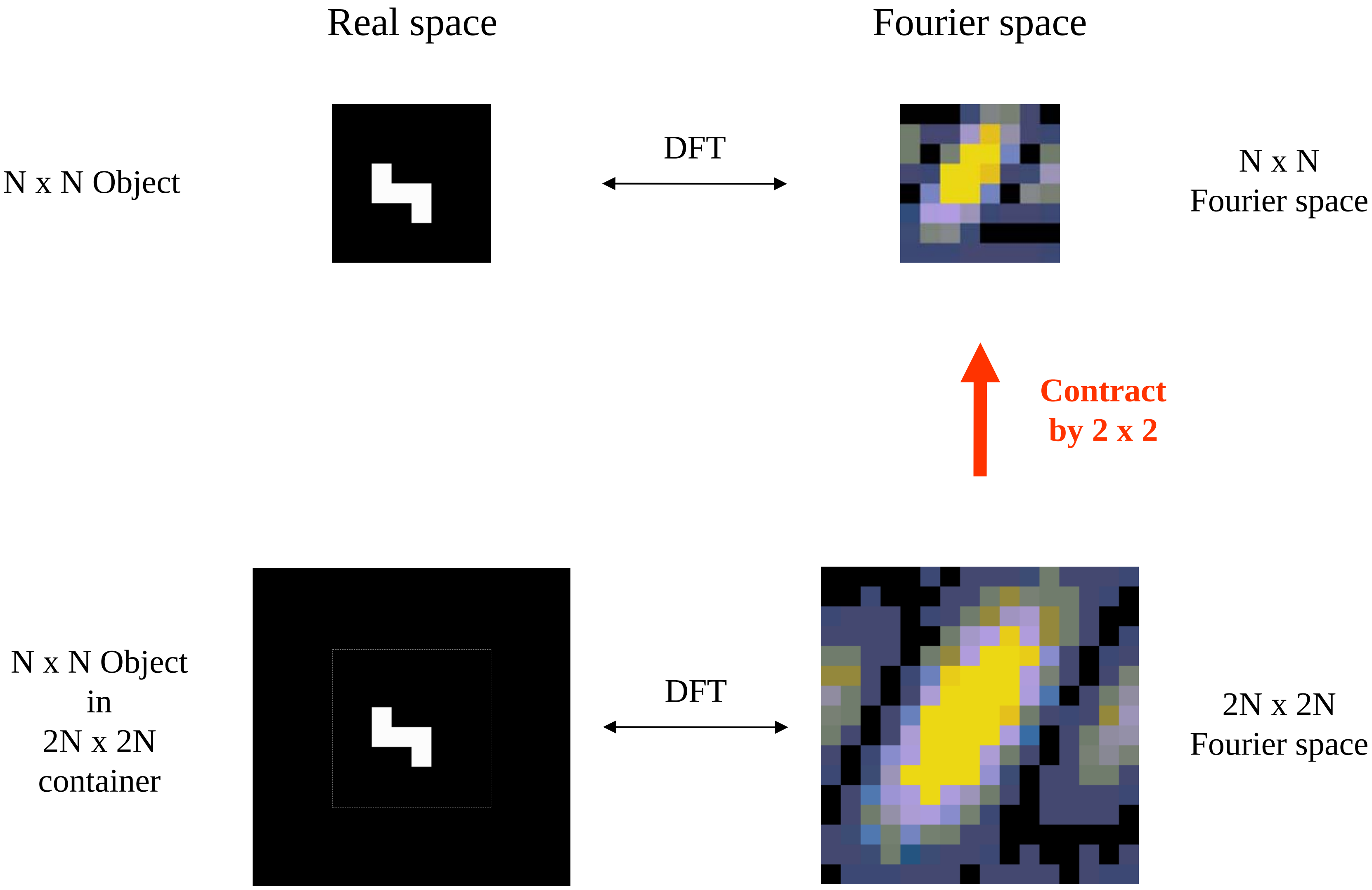
- Equilibrium and relaxation dynamics of magnetization phenomena

Experiment

- Grow thin film with T_c just above room temperature
- Let sample temperature drift slowly through T_c
- Measure
  - Magnetization
  - Susceptibility
- Record time dependence of I(**q**)



Object frame size and Fourier transformation



Real space

Fourier space

N x N Object

DFT

N x N
Fourier space

**Contract
by 2 x 2**

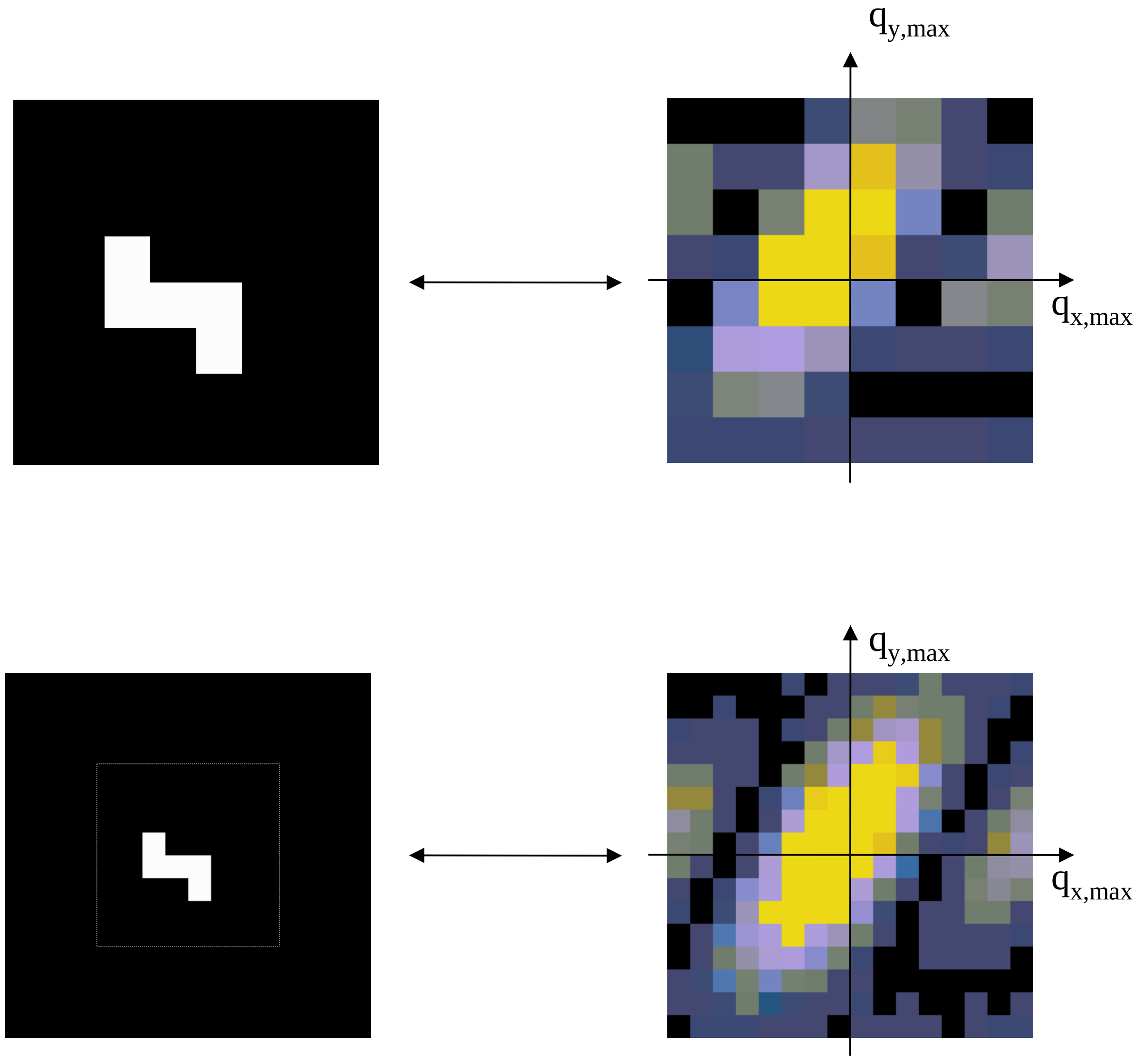
N x N Object
in
2N x 2N
container

DFT

2N x 2N
Fourier space

‘Oversampling’ overcomes X-ray phase problem

Sampling finer than the Bragg frequency (Shannon sampling)



N x N
Object

N x N
‘Detector’

N x N Object
in
2N x 2N
Container

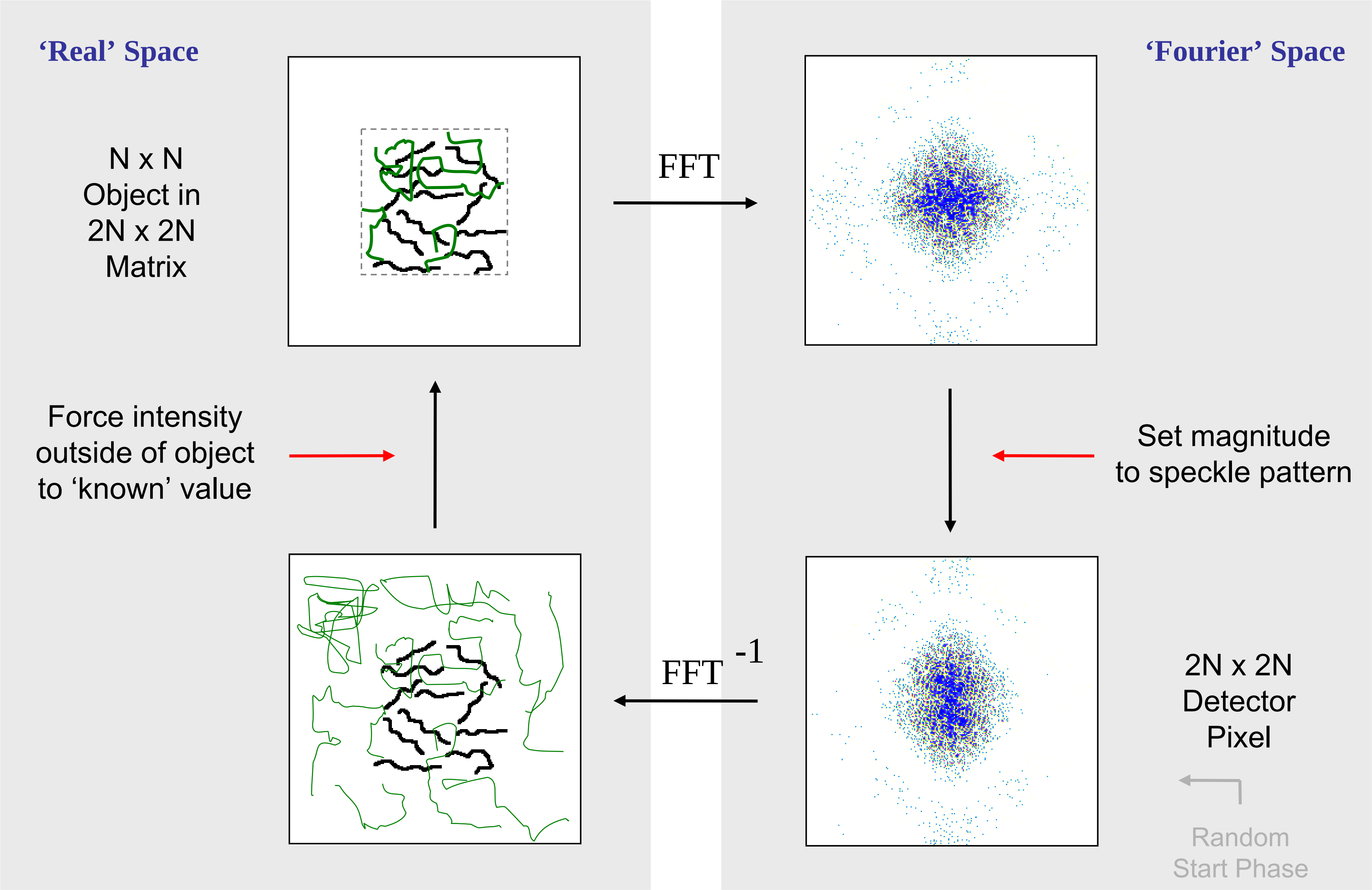
2N x 2N
‘Detector’

Real Space

‘K’ Space

Iterative algorithm for phase reconstruction

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



‘Real’ Space

‘Fourier’ Space

N x N
Object in
2N x 2N
Matrix

FFT

Set magnitude
to speckle pattern

FFT^{-1}

Force intensity
outside of object
to ‘known’ value

2N x 2N
Detector
Pixel

Random
Start Phase

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